division of technology for society

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Summary of reports, publications and patents.

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Contents

PREFACE

A national programme on coal research (NOK) has been developed, as well as a programme to locate coal deposits outside the region where coal was mined until the late sixties, to stimulate the re-introduction of coal in the energy supply of The Netherlands.

In this context the Netherlands Organization for Applied Scientific Research (TNO) has set up and is working on a large number of research and development projects and a major coal exploration programme. Moreover, TNO is participating in research activities of third parties. Some of these projects have been completed.
On the basis of the results obtained so far, new research projects with emphasis on the environmental aspects of the use of coal have been formulated and are now being carried out.

Many of the reports containing the results of the above work are or will be made available to third parties. This book has been written to promote the useful application of the knowledge which TNO has acquired so far. I hope that it will assist you in finding the information about the TNO work that may be of use to you.

Part of the information presented here has been obtained in conjunction with government departments, the Management Office for Energy Research (PEO) and the Geological Survey Department (Rijks Geologische Dienst).
The assistance of contributors from academia and industry is gratefully acknowledged.

W.A. de Jong
President

FIELD SEISMIC REPORT: BRUNSSUM, SOUTHERN LIMBURG, THE NETHERLANDS

Author : N. de Voogd
Report No. : 81-1
Date : March 8th, 1982
Language : English

SUMMARY

A reconnaissance reflection seismic survey, using dynamite as energy source, was conducted in southern Limburg, The Netherlands.
The program covered 20 kilometers of survey lines. Together with field tests and experimental sections a total of 25 line-kilometers was measured.
The survey was first part of a local reconnaissance program of 225 line-kilometers that will be concluded in 1983. The object was to derive fieldparameters and to collect data that will enable a definition of major geological data, such as overburden thickness, but if possible should also enable an evaluation of the coal field itself.

FIELD SEISMIC REPORT: SITTARD, SOUTHERN LIMBURG, THE NETHERLANDS

Author : J.A.C. Jacobs
Report No. : 82 - 1
Date : March 16th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Southern Limburg.
The program covered 27 kilometer of survey lines.
The survey was the second part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

FIELD SEISMIC REPORT: VENRAY, NORTHERN LIMBURG, THE NETHERLANDS

Author : J.A.C. Jacobs
Report : 82 - 2
Date : March 16th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Northern Limburg and the eastern part of Northern Brabant, The Netherlands.
The program covered 24.5 kilometers of survey line.
The survey was the third part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

FIELD SEISMIC REPORT: BROEKSITTARD, SOUTHERN LIMBURG, THE NETHERLANDS

Author : J.A.C. Jacobs
Report No. : 82 - 3
Date : July 20th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Southern Limburg.
The program covered 5 kilometers of survey lines.
The survey was the continuation of the second part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

FIELD SEISMIC REPORT: HORST-AMERICA, NORTHERN LIMBURG, THE NETHERLANDS

Author : J.A.C. Jacobs
Report No. : 82 - 4
Date : July 20th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Nothern Limburg and the eastern part of Northern Brabant, The Netherlands.
The program covered 28.1 kilometers of survey line.
The survey was the fourth part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

FIELD SEISMIC REPORT: LOCHEM, EASTERN GELDERLAND, THE NETHERLANDS

Author : J.A.C. Jacobs
Report No. : 82 - 5
Date : July 20th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Eastern Gelderland and the southern part of Overijssel, The Netherlands. The program covered 49.7 kilometers of survey lines.
The survey was the fifth part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

FIELD SEISMIC REPORT: BERGEN-HELENAVEEN, NORTHERN LIMBURG, THE NETHERLANDS

Author : J.A.C. Jacobs
Report No. : 83 - 1
Date : July 20th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Northern Limburg and the eastern part of Northern Brabant, The Netherlands. The program covered 62.4 kilometers of survey lines.
The survey was the sixth part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

FIELD SEISMIC REPORT: LOCHEM-2, EASTERN LIMBURG, THE NETHERLANDS

Author : J.A.C. Jacobs
Report No. : 83 - 2
Date : July 20th, 1983
Language : English

SUMMARY

A continuation of the coal reconnaissance reflection survey was conducted in Eastern Gelderland and the southern part of Overijssel, The Netherlands. The program covered 27.2 kilometers of survey line.
The survey was the last part of a coal reconnaissance program of 225 line-kilometers that will be concluded in 1983.
The objective was to collect data that will enable a definition of major geological features, such as overburden thickness, faults and general dip of coal seams.

RESEARCH ON THE COAL BENEATH THE NETHERLANDS

Authors : N. de Voogd
Chr. Staudt
Published in : Geologie en Mijnbouw 61 (1982), p. 359 - 366
Language : English

SUMMARY

A brief description is given of the programme for coal exploration in The Netherlands. The problem of detailing coal seams in the Dutch geological environment is described. With respect to resolution and lateral continuity promising results have been obtained using a field technique with the following main features: single deep shot with 0.5 kg charges, 10 m receiver group interval, and six 50 HZ geophones per group.

THIN-LAYER RESPONSE AND SPECTRAL BANDWIDTH

Authors : N. de Voogd
H. den Rooijen
Published in : Geophysics, vol. 48, No 1 (January 1983), p. 12 - 18
Language : English

SUMMARY

A derivation is presented of the response of an embedded thin layer to a vertically incident seismic pulse. The reflected pulse has the shape of the time derivative of the incident-wavelet, and its amplitude is proportional to the two-way traveltime in the thin layer and to a factor depending upon the ratio of acoustic impedances. The influence of spectral bandwidth on interface resolution and thin-layer response is investigated by means of zero-phase signals, and a filtering philosophy is proposed which enables thickness estimation either from amplitude or from peak-to-through time.

QUANTITATIVE INTERPRETATION OF COAL-SEISMIC AMPLITUDES

Authors : H. Veltmeijer
N. de Voogd

Presented at : The 46th meeting of the European Association of Exploration Geophysicists at London
Date : June 19-22, 1984
Language : English

SUMMARY

This investigation considers the possibility of deriving lateral changes in thickness of coallayers from measurements of their reflection amplitudes, starting from a calibration point.
Extensive use is made of the property of thin layers that, under some assumptions, the amplitude of a thin layer reflection changes linearly with a change in thickness of the layer.
Synthetic seismograms are used to study the effects of different changes in thickness in coal-layers.
A consequence of large changes in thickness of a coallayer appears to be that the transmitted wave through such a layer changes significantly in amplitude. This introduces a distorting influence on the amplitudes of the reflections from deeper situated coallayers. This distorting influence can be corrected, however, by filtering with the inverse of the transmission response of the shallower layers.

STATISTICAL DECONVOLUTION OF NON-WHITE MULTI LAYER DATA

Authors : R. Romijn
N. de Voogd

Presented at : The 46th meeting of the European Association of Exploration Geophysicists at London
Date : June 19-22, 1984
Language : English

SUMMARY

The conventional statistical seismic deconvolution technique assumes that the earth impulse response has a white spectrum. This assumption is examined with a modelling technique.
It is shown that an artificial 'reflectivity function', made from a borelog recording of a coal sequence in the Carboniferous in the Netherlands, is non-white, especially in the frequency range of interest. This is attributed to short delay peg-leg multiples and explained by interpreting the autocorrelation functions of cyclic layered sequences.
If a smoothed amplitude spectrum of the earth impulse response can be estimated from, for example a borehole log, it is possible to overcome the whiteness assumption in the statistical deconvolution and wavelet estimation technique.
With this wavelet estimation technique, various deconvolution filters (especially zero-phase filters) are designed and their performance is tested on synthetic and real seismograms of coal deposits. Thereby, it is concluded that it is not so much the specific waveform, but rather the bandwidth that determined the resolution.

FLUIDIZED BED COMBUSTION DEVELOPMENTS WITH TNO

Author : M.L.G. van Gasselt

Presented at : Symposium on "Development on AFBC in the Netherlands", October 7, 1982
"De Stoop" Spijkenisse and Shell Europoort (KIVI, KNCV, NIRIA)

Language : Dutch

SUMMARY

The combustion of coal in a fluidized bed with limestone has potentials as a new boiler concept to meet acceptable emission standards for safeguarding the environment from increasing pollution.

However, many areas have to be investigated to design a commercial f.b. boiler plant with clean and efficient combustion and high availability. Therefore, a 1 m x 2 m, 4 MW_{th} Atmospheric Fluidized Bed Boiler development facility has been built at TNO to carry out a R&D program to meet this goal. The AFBB is a sophisticated R&D tool capable of testing and evaluating design options arising from industrial boiler development efforts. It has been proved during initial experiments that the emission standards proposed by the Ministry of Housing, Physical Planning and Environment for nitrogen oxides, sulfur dioxide and dust are adequately met.

The commissioning tests completed in March 1982 are discussed and further development options are presented.

TNO/STORK FLUIDIZED BED COMBUSTION DEVELOPMENT

Author : F. Verhoeff (Stork boilers)
M.L.G. van Gasselt

Presented at : The 7th international conference on fluidized bed combustion, October 26, 1982
Philadelphia, Pennsylvania, USA

Language : English

SUMMARY

In this paper a new control system for industrial boilers with capacities of up to 250 tonnes per hour of steam is presented. With this control system the windbox under the fluidized bed is divided into one main bed and eight control sections. Coal is introduced in the main bed only, while the in-bed heat transfer tubes are located to a large extent in the control sections. With this control system turn-down over a large control range is possible with only small temperature variations.
To check this control system a series of tests were carried out in the 4 MW_{th} AFB Boiler of TNO, the Netherlands organisation for applied scientific research. A description of the test facility is given and test results are presented. Future developments are outlined.

OPERATING EXPERIENCE WITH THE TNO 4MWth ATMOSPHERIC FLUID BED BOILER FACILITY

Author : H.M.G. Temmink

Presented at : Coal Technology Europe
3rd European Conference & Exhibition on Coal Utilization
11-13 October 1983, Amsterdam
Volume 2, 99-123

Language : English

SUMMARY

The purpose of the research with the TNO 4 MWth AFBB pilot-facility is, to establish the parameters governing the fluid bed combustion of coal, so that industrial plants can be built with good combustion efficiency, at the same time complying with the emission standards in the Netherlands. The research related to the 4 MWth AFBB consists of four parts concerning:

1. The operation of the boiler, including maintenance, boiler characteristics, load control, fly ash recycling and other development work, the supplier of the boiler, STORK BOILERS, cooperates in it. This paper will deal with some aspects of this research.
2. The environmental problems associated with the combustion of coal, including measurement of flue gas components, biological effects of (PCA's in) ash from the bag filter and from the stack (mutagenity, teratogenity a.o.), reduction of limestone consumption and removal of fly ash from flue gas by means of an electrified bed filter.
3. Application of ash from fluid bed combustion of coal.
4. Chemical and physical characterisation of coal.

In this paper a review of the pilot-plant is given followed by the results of the commissioning tests and a year of operation. Eventually future research by TNO and Stork is discussed.

ADVANCED FLUIDIZED BED COMBUSTION

Authors : A.G. Melman
M.E. Reinders
Reference no. : 84-0418
File no. : 8728-10004
Date : March 1984
Language : Dutch

SUMMARY

This report gives a comparing survey of external circulating fluidized-bed combustion (CFBC) systems using coal and low-grade fuels.
The main advantages over the AFBC are: a better load following capability and a greater turndown ratio, due to the fact that heat production and heat transfer are separated. Moreover, a CFBC has a greater energy density and the number of feed points is smaller. Limestone efficiency and combustion efficiency will be only slightly better than those of the AFBC of the second generation with improved performance.
A disadvantage of the CFBC is its greater complexity, which makes the installation relatively expensive. The higher capital costs could be offset by scaling up and using cheaper fuel (low-grade coal and waste).
Probably application of the CFBC is not the best solution for the Dutch situation, as we have to import coal over long distances, which leads to the use of a higher-quality coal.

THE ATMOSPHERIC FLUIDIZED BED BOILER OF THE TNO COAL PROJECT

Author : H.M.G. Temmink
Published in : Energiespectrum, May/June 1984, 132-140
Language : Dutch

SUMMARY

As a result of the options of the Dutch boiler industry to construct medium size up to very big atmospheric fluidized beds, a 4 MW_{th} boiler has been constructed. The choice of fluidized bed technology is based on its advantages:

- the SO_2 emission can be limited effectively and cheaply by adding limestone to the fluidized bed
- NO_x emission is lower than from other combustion techniques as a result of the lower combustion temperature
- coal with high humidity and high ash content can be combusted without problems
- it is not necessary to grind the coal. Breaking to 6-9 mm is sufficient
- the heat transfer coeffecient is high, and, as a result, the boiler can be compact.

The construction of the $4MW_{th}$ fluidized bed is discussed. Experiments with this fluidized bed boiler have shown, that unattended continuous operation and load control are possible without difficulties. The requirements for the emission of SO_2, NO_x and dust (600 g/GJ, 300 g/GJ and 20 g/GJ) respectively can be easily met.

MASS-BALANCE, HEAT BALANCE AND EFFICIENCY OF THE TNO-AFBB

Authors : A.W.M.B. van Haasteren
J.A. de Jong

Report no. : 84-012300
File no. : 8726-10271
Date : October 1984
Language : Dutch

SUMMARY

The experiments described in this report are made with the TNO atmospheric fluidized bed boiler. The combustion efficiency was investigated as a function of the amount of recirculated fly ash.
In previous experiments, the combustion efficiency was measured as a function of the amount of recycled cyclone ash. In these experiments, ash collected in the cyclone as well as in the fabric filter was recycled, using an improved recycling system. During these experiments the SO_2 and NO_x emissions were also measured.
The mass- and heat balances are calculated. From the overal balances the balances over bed, freeboard and convection sections of the boiler are derived. Process parameters as boiler efficiency, combustion efficiency and carbon efficiency are derived from these balances made in five experiments with Virginia coal. The percentage of combustion in the freeboard is also measured.
A Sankey diagram is described.
Recycling 850 kg cyclone ash/h gave a combustion efficiency of 95%.
Recycling of 850 kg/h of a mixture of cyclone ash and fabric-filter ash gave a combustion efficiency of 97,1%. With an increasing amount of recycling the SO_2 emission increased at first, and decreased afterwards.
The observed increase of SO_2 emission can be explained by a decrease of the O_2-concentration and possibly by a $CaSO_4$ decomposition into SO_2 and CaO as a result of the high temperature in the freeboard ($970^{o}C$).
The NO_x emission decreased, caused by the decrease of the O_2 concentration. During the experiments the bed height was 1.06 m and excess air 20%.

APPLIED RESEARCH INTO FLUIDIZED-BED COMBUSTION

Author : H.G.M. Temmink

Presented at : Symposium Schone Verbranding van Steenkool
(Clean Combustion of Coal)
January 16, 17, 1985
Noordwijkerhout

Language : Dutch

SUMMARY

Within TNO (Applied Scientific Research) the Project Coal Technology has been started. This project has been devided in four sub-projects:

1. investigation into the optimalization of the AFBC design,
2. biological investigation into the environmental effects of (fluidized bed) coal combustion,
3. investigation into the application of waste,
4. development of new characterization methods.

In this paper attention is only paid to some aspects of the first sub project: the combustion efficiency and the resulting SO_2 and NO_x emissions. A survey is given of the experiments performed and the improvements made. The effect of the improvement of the efficiency on the emissions of SO_2 and NO_x is indicated. The measures to be taken for further improvement of the combustion efficiency and emission control are outlined.

OPERATING EXPERIENCE WITH A 4 MWth AFBB RESEARCH FACILITY

Authors : J.P. Meulink, Stork Boilers
H.G.M. Temmink, TNO
A.W.M.B. van Haasteren, TNO

Presented at : The Eighth International Conference on Fluidized Bed Combustion "Options and Issues" at Houston, Paper 2.1
Date : March 18-21, 1985
Language : English

SUMMARY

In December 1979, TNO, The Netherlands Organization for Applied Scientific Research, awarded to Stork Boilers the order for the construction and commissioning of a 4 MW_{th} atmospheric fluidized bed research facility for coal, gas and oil firing.
After commissioning and acceptance testing, the facility was transferred to TNO in April 1982.
From that moment, Stork Boilers and TNO have conducted a joint research program aimed at the design of industrial facilities up to steam rising capacities of 250 tons/h. Thanks to the experience thus acquired, Stork Boilers obtained the order for the construction, commissioning and testing of a 100 MW_{th} AFBB in December 1983.
After a general description of the boiler has been given, various aspects of the research with the 4 MW_{th} facility is dealt with:

- erosion protection of in-bed heat transfer tubes,
- design and testing of the grit refiring system,
- load-control system and possible use of velocity turndown,
- admissible stack temperatures in view of dew point corrosion and NH_4Cl fouling.

EVALUATION REPORT FLUIDIZED-BED COMBUSTION, NATIONAL PROGRAM COAL RESEARCH

Reference no. : 8175-0365
File no. : 8730-10269
Date : January 1985
Language : Dutch

SUMMARY

In this report an evaluation is made of research at TNO into the fluidized bed combustion of coal during the past two years.
The research involves a) the operation and maintenance of the 4 MW_{th} fluidized bed boiler and the experiments carried out on the boiler performance and environmental pollution and b) the development of components. Much attention was also paid to the application and removal of residues and the characterization of coal and coal residues.

The 4 MW_{th} AFBB was in operation for the benefit of industry and TNO research for 3,200 hours during the period covered. Much attention had to be paid to the erosion of the in-bed heat transfer surface. The configuration of the tube bundles as well as the construction material was varied. For the benefit of the load following, experiments were carried out to test the Stork control concept. At the same time it appeared that load control by means of bed expansion was only marginally possible.
The combustion efficiency could be increased from 95% to 97.4% without further optimalization by the combined reinjection of filter ash and cyclone ash with a newly installed reinjection system.
In all the experiments, the stack emissions were measured, whereby it appeared that the emission standards for SO_2 and NO_x could generally be adhered to.
The 4 MW_{th} AFBB was enlarged with a facility for testing flue gas filters. The fly ash reinjection system was renewed. The experience gained during the commissioning of this system was extremely instructive and showed once again how important it is to test such partial systems on this scale.

The sampling of flue gases and solids demands much continuous attention in order to be able to guarantee the high reliability that is necessary for a correct evaluation of experiments carried out.

An important start has been made in the development of adequate analysis techniques for the characterization of coal, additives and residues. A number of analysis techniques are already available while others need further research.

It became apparent that agglomeration of fly ash with conventional equipment can be achieved satisfactorily, at the same time reactivating the unused limestone. Reinjection of the agglomerates by means of the coal feed system, renders a reinjection system unnecessary, improves the combustion efficiency, and leads to a decrease in the limestone consumption.
The laboratory experiments now have to be verified in the AFBB. A patent application has been submitted for a limestone reactivation method in which the water present in the coal supply is used.

Cold extrusion is useful as a continuous, well controlable feed system for coal, coal-ash mixtures, and low grade fuels, against high pressures if desired.

Apart from a reliable feed system for AFBC and gasification, extrusion can be pre-eminently suited for application in the feed of the PFBC. The prototype designed should be built and tested in practice.

The corrosion research of uncooled construction materials has shown considerable differences between the various prospective materials. Even the best materials will have to be replaced regularly, unless suitable coatings are found. Under cooled conditions the 9/12% Cr steels appear to be clearly more resistant to erosion/corrosion than the lower alloys and carbon steels.

Using a cold model, freeboard internals were developed by which the residence time of particles in the freeboard is extended and therefore the chance of complete combustion increased. Two practical experiments in small fluidized bed boilers should give a decisive answer to the realistic gain in efficiency which can be achieved.

The development of a flue gas filter has led to a model of an electrostatic granular bed filter (EGB-filter). The efficiency of the EGB-filter is equal to that of good fabric material. The gas velocity is, however, higher and the pressure drop equal or lower. This can lead to a compact filter that at the same time is suitable for high temperatures. All attention is focussed at the moment at developing the best cleaning method.

The electrostatic separation of fly ash in a fraction rich in carbon and a carbon deficient fraction, using the equipment used in the coal upgrading research, has not produced good results. As fly ash separation is of great importance, not only for fluidized bed combustion but also for pulverized coal boilers, in view of the application of the fly ash, other separation methods will have to be found.

A desk study showed that a heat exchanger of polymer material offers promising perspectives for cooling the flue gases to below the acid- and water dew point, thus putting the heat produced to good use.

In the research into the application of AFBC residuals, a distinction was made between possibilities for the short, medium, and long term. In the short term, application as an asphalt filling proved possible but there is not much demand. Also the application as a raw material for cement production offers good perspectives given a number of technical and commercial conditions is fulfilled. For the application of AFBC residuals in the medium term exploratory research has been carried out into their possibilities in building materials. This showed that AFBC ash, with its typical high sulphate and residual carbon content, will not easily be used as a raw material for the traditional building materials.

Further research will have to be aimed at those building materials in which the specific AFBC residual properties can be used and where limited competition from other waste products is to be expected.
For the application of AFBC residuals in the long term, research has been conducted into the recovery of aluminium and aluminium products. The technical possibilities have been shown on a laboratory scale. The economic evaluation has shown that the manufacture of aluminium oxide is not (yet) economically attractive but that the production of aluminium sulphate looks promising.
In the context of research into the environmental effects of landfill of AFBC residuals, attention was especially focussed on the leaching of ash. A contribution has thus been made to the realization of the standard leaching procedure.

From the conducted PAH emission measurements, it was concluded that fluidized bed combustion of coal can be a clean and environmentally acceptable process if the boiler installation is well designed. The emissions are several times lower than those of small coal fired boilers not based on the fluidized bed principle, and comparable with those of a pulverized coal boiler.

Using a number of biological in vitro tests to determine the genotoxic and cytotoxic activities, it has been possible to make a contribution to the toxicological characterisation of inhalable fly ash. The results of the measurements should be seen as relative effects with comparitive values.
Types of fly ash and their connected combustion techniques can thus be distinguished as to their more or less positive properties for man and the environment. In the same way the influence of various process conditions can probably be judged, but this has not been adequately investigated.
From a comparison of the toxic characteristics in vitro of fly ash emissions of four installations studied, it would seem that the two small installations compare very unfavourably with the fluidized bed boiler and the pulverized coal boiler, as far as their mutagenous

emission is concerned. It has been shown that stack ash is far more cytotoxic and mutagenous than filter ash. This is of special importance when estimating the contribution of stack ash from coal fired installations to the already present mutagenous activity of ambient air (additional, or reinforcing, or weakening by chemical conversions). High priority should be given to the immediate vicinity of small coal fired installations with low stacks.

It has furthermore been shown that filter ash from some small boilers, depending on the type of installation and combustion conditions, can be less harmless than was until recently generally supposed. This aspect deserves further attention from a point of view of labour hygiene. From a point of view of environmental toxicity it is also of importance because of possible leaching of mutagenous materials into the soil and water from landfill of fly ash.

MICROSCOPIC AND X-RAY MICROANALYTICAL INVESTIGATIONS OF A NUMBER OF MATERIALS WHICH HAVE BEEN EXPOSED DURING 300 HOURS TO TEMPERATURES OF 800°-850°C IN THE 4 MWth FLUIDIZED BED BOILER

Authors : J. van der Veer
J.C. van Wortel
P.L.F. Rademakers

Report No. : 83M/0596/WOR/SGE
Order No. : 6136.5.0043
Date : January 20, 1983
Language : Dutch

SUMMARY

Seven heat-resistant materials have been exposed during 300 hours to a temperature of 800°-850°C in an atmospheric fluidized bed combustor. The results showed that all materials are covered with a duplex scale consisting of:

a) An inner layer which has penetrated into the steel. This oxide layer has been formed with oxygen and elements from the steel: mainly Cr and in less degree Mn, Si and Fe. No characteristic differences in composition are found between the materials exposed in the bed, the splash zone and the freeboard. The highest attack occured at the under side of the material facing the fluidized-bed. In the freeboard, the attack was more local than in the bed and the splash zone. Haynes Alloy 188 is attacked least (<30 µm), followed by AISI 446 → AISI 347 → AISI 310S → 253 MA. Most severely attacked were Hasteloy S and Alloy 800H (max. about 100 µm).

b) An outer oxide layer containing the elements from the fluidized bed, mainly Al, Si, Ca and S. The composition of the oxide layer in the bed, splash zone and freeboard was comparable.

The total scale (a + b) consisted of calcium sulphate, quartz, metal oxides and complex silicates.
Most substrate materials showed depletion of Cr and Mn.
Hardly any depletion is observed in AISI 446, AISI 347 and HA 188.
All materials showed some internal sulphidation, except HA 188 and AISI 446. The sulphidation was most pronounced in the Ni-rich alloys Alloy 800H and Hasteloy S. In the bed and in the splash zone the sensitivity for sulphidation was significantly higher than in the freeboard, especially at the underside of the specimens. Two kinds of sulphides were found: chromium sulphides and manganese-chromium sulphides. Nickel sulphides were not present.

CORROSION/EROSION OF MATERIALS FOR FLUIDIZED BED COMBUSTOR APPLICATIONS

Authors : P.L.F. Rademakers
B.H. Kolster

Report No. : 82M/0307/613650043/RAD/JIB
Order No. : 36.5.0043
Date : October 14, 1983
Language : Dutch

SUMMARY

Various aspects of the atmospheric fluidized bed combustion of coal are studied extensively at TNO for which a.o. an atmospheric fluidized bed boiler (4 MWth) is used.
Part of the project deals with the exposure and evaluation of candidate fluidized bed materials.

The first 300 hours exposure of uncooled rings were realised. Examination of the dismounted rings revealed the following preliminary results:

- all specimens are covered by a deposit which consists of calcium sulphate, mixed oxides and complex silicates;
- erosion phenomena are not observed;
- at all locations (in bed, splash zone, free board) the maximum attack occurred at the underside of the rings
- a preliminary ranking order of decreasing resistance against the attack in the combustor is HA 188, AISI 446, AISI 347, AISI 310S, 253 MA, Hast S, and Alloy 800.

CORROSION/EROSION OF MATERIALS FOR AFBC APPLICATIONS

COST 501 PROJECT NL-9 Progress Report No. 1

Authors : P.L.F. Rademakers
L. Bos
J.C. van Wortel

Report no. : 83M/013687/RAD/SGE
Order no. : 6136.5.0101
Date : December 1983
Language : English

SUMMARY

The program, part of which already started in 1982, comprises the examination of uncooled rings and cooled tubes, which are exposed in the 4 MW_{th} Atmospheric Fluidized Bed Boiler (AFBB) at Apeldoorn.
In the progress report a description is given of the test facility, the methods, materials and the test conditions. Uncooled rings of several high temperature materials are exposed for various times in the bed, the splash zone and in the free-board. The coal which is burned contained 0,86% sulfur, and limestone is added as sorbent. The first results of metallographic and micro-analytical examinations, carried out after exposures of 300, 700 and 1000 hrs, are described. Metal losses of 0.02 to 0.2 mm are observed, the attack in the free-board being more pronounced than in the bed. All exposed materials showed internal sulphidation, and chromium and magnanese sulphides are formed.
Tests with uncooled rings are continued, and an additional period of about 1500 hrs is reached.
Cooled test tubes of ferritic boiler steels are made in addition. These tubes are placed in the bed and in the free-board, the test temperatures being between 400° and 600°C. The cooled tubes reached operation times of 1000 hrs. The tests are continued.

CORROSION/EROSION OF FERRITIC TEST TUBES EXPOSED IN THE 4 MW_{th} AFBB FOR 1260 HRS

Authors : P.L.F. Rademakers
L. Bos
J.C. van Wortel

Report no. : 10287 Job 2.1.15
Order no. : 36.5.0103
Date : May 1984
Language : English

SUMMARY

Uncooled rings and cooled tubes are exposed in the coal fired 4 MW_{th} Atmospheric Fluidized Bed Boiler (AFBB) at TNO Apeldoorn.
This report describes results of wall thickness measurements on test tubes from various ferritic boiler steels after a period of 1260 hrs. Low alloy steel (st. 35.8) and steels with chromium contents of 1%, 2¼%, 9% and 12% are exposed, the metal temperature lying between 400°C (st. 35.8) and 560°C (12% Cr-steel).
In the bed corrosion/erosion occurred. The wall thickness of the steels containing 2¼% chromium or less decreased with 0.56 to 0.73 mm, whereas the 9 and 12% chromium steels showed values of 0.10 mm.
In the freeboard similar differences between the materials were observed, albeit that the metal losses were restricted to 0.3 mm and 0.07 mm.
The mechanism involved obviously concerns the formation of oxide, and the simultaneous removal of the oxide by the erosive action of particles. Instead of a parabolic oxidation rate this may result in a high linear rate. Under the conditions encountered the erosion/corrosion resistance of the 9% and 12% chromium steels is significantly better, in comparison with C-steel and low-alloy steels.

HARDNESS MEASUREMENTS ON FBC-PARTICLES AND OXIDE/DEPOSITION SCALES ON TEST TUBES

Author : J.C. van Wortel
Report no. : 84M/010881/WOR/VLT
Order no. : 6136.50103
Date : August 10th, 1984
Language : Dutch

SUMMARY

Several materials, which have been exposed in the fluidized bed combustor are more or less eroded. There are indications that erosion of the metal is not the only factor, and erosion of the oxide scale plays a rule as well; so the process would be corrosion/erosion.
It is supposed, that the erosion velocity is not only influenced by the shape, the size and the velocity of the particles, but by their hardness as well.
However, there is hardly any knowledge concerning the relation between the hardness of the particles and the oxide scales. That is why the hardness of the oxide scales is investigated in comparison to the fluidized bed material.
It was found that various types of limestone showed significant hardness differences, but all types investigated were relatively soft (max. 160 HV). In addition the FBC particles consisted of silicates with hardness values of about 720 HV, hard particles reaching 900 HV and particles with hard scales reaching 1300 HV.
The hardness of the various steels was relatively low (130 HV for carbon steel, 240 HV for 12% Cr steel). However the oxide scales reached values of 635 HV resp. 700 HV. And high Cr stainless steels exposed at temperatures of about 850^{o}C developed scales with values of 900 HV to 1000 HV. By comparing the results it is expected that oxide layers can play an important role as a "natural" erosion resistant coating.

CORROSION/EROSION TESTS IN A 4 MW_{th} ATMOSPHERIC FLUIDIZED BED BOILER

Authors : P.L.F. Rademakers
L. Bos
J.C. van Wortel
B.H. Kolster

Presented at : 3rd International Conference Fluidised Bed Combustion
Is it Achieving its Promise?
London, 16/17 October 1984
The Institute of Energy

Language : English

SUMMARY

The behaviour of candidate high temperature materials in a 4 MW_{th} Atmospheric Fluidized Bed Combustor was investigated. The combustor, which is a test facility, was operated with medium sulphur coal and limestone. Rings of stainless steels, nickel base and cobalt base alloys were exposed in the bed, the splash zone and the freeboard at temperatures of 850° to 900°C for periods up to 2825 h.
In addition cooled test tubes of different ferritic boiler steels were exposed for 1260 h. The metal temperature varied from about 400°C, at the C-steel end of the testtubes, to about 560°C for the 12% chromium steel. Metal losses were measured, and metallographic examinations were carried out. The results are evaluated and possible mechanisms of attack are discussed.

CORROSION/EROSION OF MATERIALS FOR AFBC APPLICATIONS COST 501 PROJECT NL-9 Progress report no. 2

Authors : P.L.F. Rademakers
L. Bos
J.C. van Wortel

Report no. : 85M/33/0123/RAD/VLT
Order no. : 33.5.3012
Date : January 1985
Language : English

SUMMARY

Uncooled samples and cooled test tubes are exposed in a 4MWth Atmospheric Fluidized Bed Boiler (AFBB) which is fed with low sulphur coal under limestone addition.

Uncooled rings of stainless steels, nickel and cobalt base alloys were exposed in the bed, the splash zone and the freeboard for periods up to 2810h at temperatures of 820° to 900°C. Metallographic examinations revealed outer calciumsulfate containing scales and inner chromium rich oxide layers. All materials showed internal sulphidation, and the formation of chromium and manganese sulphides.

For all materials the attack was most severe in the freeboard, and was most pronounced at the bottom of the rings. The best performance was shown by the materials 16Cr5AlY, HA 188, AISI 310 and Alloy 800 H, and 253 MA.

Cooled test tubes of different ferritic boiler steels were exposed for 1260h. The metal temperature varied from about 400°C, at the C-steel end of the test tube, to about 560°C for a 12% chromium steel. Under the applied conditions, in the bed, the steels containing 2¼% chromium or less showed erosion/corrosion. The 9% and 12% Cr-steels behaved significantly better. In the freeboard similar differences were observed, albeit that the material losses were much lower. The corrosion/erosion mechanisms are discussed.

A FREEBOARD INTERNAL FOR COAL FIRED FLUIDIZED BED BOILERS

Author : J.W. Pohlman
Reference no. : 85-071
File no. : 8726-10292
Date : January 1985
Language : Dutch

SUMMARY

The combustion efficiency of a coal-fired fluidized bed boiler depends among other things on the residence time of the solid particles in the freeboard.
TNO has developed a freeboard internal by which the residence time of the solid particles can be extended. The residence time depends on the height of the internal, the particle size, and the superficial gas velocity.
In an internal with a height of 2 meter the mean residence time of 100 µm particles increases with 0.9 seconds at a superficial gas velocity of 2.3 m/s.
This results in an increase of at least 2% of the combustion efficiency of a fluidized bed boiler without recycling.
The internal has a positive influence on the mixing of the various gases and possibly on the velocity distribution, too.

IMPROVED FLUID BED COMBUSTOR EFFICIENCIES THROUGH FREEBOARD INTERNALS

Authors : L.M. Rappoldt
M.L.G. van Gasselt

Presented at : The Eighth International Conference on Fluidized Bed Combustion "Options and Issues", Paper 3.2
Date : March 18-21, 1985
Language : English

SUMMARY

A system of plates and baffles inclined with respect to the vertical is arranged in the freeboard of an AFBC so that channels of two opposed arrays of superimposed inclined plates are obtained. With this system of internals a prolonged residence time of elutriated particles in the freeboard is obtained as well as a more uniform freeboard flow distribution.

In a cold model in which the internals have been developed the particle hold-up has been measured to be two times larger than in a freeboard without internals. The real effects on combustion phenomena have been measured in a 3 MW small boiler unit. The improvement of the combustion efficiency of $\pm$ 90% to $\pm$ 95% with this prototype internal assembly is very promising. The possibilities for further improvement are outlined.

FLUIDISED BED COMBUSTION APPARATUS

European patent application

Inventor : M.L.G. van Gasselt
L.M. Rappoldt

Application no.: 84200196.8

SUMMARY

A fluidised bed combustion apparatus in which in the free-board above the bed at least one system of plates or baffles inclined with respect to the vertical is arranged so that at least one upwardly directed channel having internally at least one group of two opposed arrays of superimposed inclined plates is formed, which plates at least alternately extend obliquely downwardly from the outside to the inside and are closely connected with the outer boundary of the channel.

Preferably all plates of each array extend obliquely downwardly at an angle with the vertical within 20^{o}-50^{o} from the outside to the inside from a vertical wall defining said channel whereas the plates of the one array are off-set over a distance "s" in the vertical direction with respect to the plates of the other array.

EXTRUSION OF COAL, PART I & II

Authors : C.A.J. Bultink
J.L. den Otter
Report no. : 201/'83
Order no. : 200 472 101
Date : August 11, 1983
Language : English

SUMMARY

The use of an extruder to feed combustion facilities for heat production is attractive for a number of reasons. Firstly it is to be expected that the continuous feeding can be made insensitive to all kinds of contaminations/impurities such as excessive moisture, pieces of rubber or other dirt, and large variations of particle size in the run-of-mine coal. In addition all kinds of additives can, to some extent, be continuously mixed with the coal. Obvious examples are: additives to limit emission of SO_2 (limestone), part of the ash for reburning and other fuels such as sulphur contaminated oil, wood or paperwaste, plastic-waste and so on. Extrusion would be particularly attractive for feeding plants in which a reaction takes place under elevated pressure, as in gasification plants, because it enables continuous feeding under high pressures.

Extrusion of coal is a new technology, however, and the few experiments that have been performed have shown that the difficulties in maintaining a continuous troublefree operation are manifold. As a result, and because a systematic trial and error investigation of extrusion is very time-consuming, any investigation of extrusion of coal will be quite expensive. We therefore used systematic rheological measurements to predict the extrusion conditions.

The materials chosen for our investigation are the two coal types used in the Netherlands in experimental atmospheric fluidized bed boilers (AFBB), viz. the Zolder coal from Belgium and a mixed coal of Polish origin. An advantage of this choise is that the two coals are very different in nearly all respects.

The most attractive way of feeding an AFBB seems, at first sight, to be extrusion at high temperature (< 400°C), where coal behaves plastic. This would enable us to reach the high pressures obtainable in the extrusion of plastic melts (up to more than 1000 bar) and to mix the additives intensively with the coal. The gases coming from the degradation of the coal can be burned separately or used otherwise. Hot extrusion proved possible with the Zolder coal, but not with the Polish coal. We found that part of the conveying problems with the Polish coal occurred in the feed zone, which is at a relatively low temperature, even in a hot extruder. We therefore continued our investigation of feeding problems at low temperatures.

Cold extrusion was investigated on a 60 mm extruder, which is equiped with the latest novelties in the field of single screw extrusion, such as grooved cylinders with various geometries. Under cold extrusion conditions the extruder is a solids-conveying pump. The extrusion was used to investigate cold extrusion as such, and the feeding problems with hot extrusion. After initial difficulties, we finally succeeded to obtain high outputs at room temperature with a reliable and stable output, which could be sustained against a high pressure.

Because larger AFBB's must be fed simultaniously from several nozzles, we tried to use the extruder to feed pipes of various lengths. This proved to be difficult because at high temperatures (hot extrusion) the residence time of the coal in the pipes easily exceeds the time the coal remains stable. This problem has not yet been solved. At low temperatures (cold extrusion) "dry" friction in the pipe results in a very unstable situation; this problem can only be solved by using lubricants, such as oil-waste.

From our experiments we conclude that hot extrusion can be an attractive solution for feeding reactors and a possible solution for feeding small scale installations, where long pipes are not necessary. Cold extrusion offers an economic and reliable feeding system for AFBB-systems. As compared to pneumatic feeding systems the costs seem similar, but the reliability is better. The output of the 60 mm extruder was about 170 kg/hour with wet Polish coal (water content 7.5%). More work is needed to investigate the transport through pipes, since the difficulties seem surmountable.

TNO

EXTRUSION OF COAL - Part III Extrusion

Auhtors : C.A.J. Bultink
J.L. den Otter

Report No. : 201/'83
Order No. : 200472101
Date : July 20, 1983
Language : English

SUMMARY

Hot extrusion tests for feeding coal in its plastic state to a combustion facility were carried out on Polish and Belgian coal. In these experiments, carried out on a 30 mm and a 50 mm single screw extruder, Belgian coal was extrudable at temperatures of about 500°C. The maximum output found on the 50 mm extruder at 60 rpm was about 90 gr/min of a foamy plastic coal. The process was not suitable for Polish coal and for extrusion at high pressures. On a 60 mm single screw extruder the coal extrusion process was further investigated.
Cold extrusion tests for feeding coal in its solid state to a combustion facility were carried out on Polish coal. For stable extrusion of coal it was necessary to use a fully grooved cylinder. In these experiments coal was extrudable against high pressures (about 20 bars) and with additives such as limestone, water, oil and impurities like rubber and wood. The output of 60 mm extruder at 150 rpm varied between 4000 gr/min for dry coal and 2700 gr/min for wet coal (water content 7.5%). For stable hot extrusion of coal in its plastic state it was concluded that a single screw extruder should have a fully grooved cylinder in those parts of the extruder where the coal is not plastic.
Scouting tests were performed for cold extrusion through pipes and for the splitting of a coal stream, coming out of the extruder, into two pipes.

ELECTROSTATIC SEPARATION OF THE ORGANIC AND ANORGANIC COMPONENTS OF FLY ASH FROM COAL FIRED FLUIDIZED-BED BOILERS

Authors : J. Bouwma
V.L. Vlasblom

Report no. : 345/84
Project no. : 200372106
Date : September 14, 1984
Language : Dutch

SUMMARY

We have investigated the possibility to separate fly ash, originating from incomplete fluidized-bed combustion of coal, electrostatically into carbon and mineral enriched fractions. A method was used based on tribo-charging of fly ash particles in a stainless steel cyclone, and subsequent deflecting in an electric field.
Starting from unground coarse fly ash captured with a cyclone (cyclone ash, approximate particle size range 10-400 μm), carbon enriched fractions of fine particle size are collected at both electrodes. This separation result is primarily due to the different particle size of the fine organic and the coarser inorganic constituents, especially the added calcium compounds. The coarser particles are hardly deflected. Sieving (150 μm sieve) gives a similar separation result: a coarse mineral and calcium enriched fraction (20-25%) containing less than 5% organic carbon is obtained.
The electrostatic separation of ground cyclone ash results in a carbon enriched fraction collected near the positive electrode. The added calcium compounds are, far more than the minerals, deflected to the negative electrode.
For both unground and ground cyclone ash with 32% organic carbon, concentrates with about 50% organic carbon are obtained containing about 60% of the organic carbon present in the feed (i.e. a recovery of about 60%). We did not succeed in concentrating the organic carbon with a high recovery (e.g. 80% or more) in a fraction with more than 80% organic carbon, even not when using a second separation step. This is due to the

insufficient separation of the organic and the mineral (not the added calcium compounds) matter, which probably is caused mainly by the similar charging behaviour.

The very fine fly ash captured with a fabric filter (fabric filter ash, approximate particle size range 1-40 µm) cannot be separated with the electrostatic method used. Besides the reasons mentioned before, this is probably mainly due to predominant interparticle contact charging, which is less selective.

COAL CLEANING USING ELECTROSTATIC SEPARATION

Authors : J. Bouwma
B.H.M. Kuypers
V.L. Vlasblom
A. van der Waal

Presented at : International symposium: Electric and Magnetic Separation and Filtration Technology, Antwerp, 23-25 May 1984

Language : English

SUMMARY

Three methods for electrostatic cleaning of finely ground coal (80% w/w- 200 mesh) have been investigated. The methods differ mainly in the way of charging the coal particles viz. tribocharging in a cyclone, contact charging during fluidization and corona charging via a drum.
The cleaning processes are hampered by the fineness of the coal and by the broad particle size distribution. The best cleaning results are obtained using tribocharging in a cyclone. Cleaning by this method results for some coal types in about 90% reduction of the pyrite and the ash content of the coal while the recovery of coal or calorific value exceeds 80%.

ELECTROSTATIC SEPARATION OF COAL AND FLY ASH COMPONENTS

Author : J. Bouwma
Presented at : The IEEE/IAS Congress, Chicago, October 1-4, 1984
Language : English

SUMMARY

Three methods for electrostatic cleaning of finely ground coal (80% w/w - 200 mesh) have been investigated. The methods differ mainly in the way of charging the coal particles, viz. tribocharging in a cyclone, contact charging during fluidization, and corona charging on a drum. The cleaning processes are hampered by the small particle size and the broad particle size distribution of the coal. The best cleaning results are obtained using tribo-charging in a stainless steel cyclone. Cleaning by this method results for some coal types in about 90% reduction of the pyrite and the ash content while the recovery of coal or calorific value exceeds 80%.

Electrostatic separation of mineral and maceral components of fly ash using tribo-charging in a cyclone has also been studied. The fly ash investigated results from atmospheric fluidized bed combustion of coal. Only moderate separation results are obtained due to the low selectivity of charging and, for relatively coarse-grained fly ash, also due to the low charge-to-mass ratio.

WORKING SYSTEM AND EQUIPMENT FOR THE COMBUSTION OF A FUEL WITH A FLUIDIZED BED

Patent application

Inventor : M.L.G. van Gasselt
Application no. : NL 8302112

SUMMARY

A fluidized bed apparatus for the combustion of fuel, preferably coal, where the flue gas and the entrained particles are conducted into a space where the particles are separated from the gas stream and recycled into the combustor, characterized in that, next to the collection of the particles from the flue gas, the carbon and carbon containing particles are separated from the ash particles, and the coal (containing) particles are recycled exclusively into the fluidized bed.
Working system according to this claim characterized in that the coal (containing) particles and the ash particles are separated by conducting the flue gas stream into a cyclone where the particles are separated and be electrically charged by friction. The particles then go into an electrostatic precipitator, where the coal (containing) particles go to one electrode and the ash particles leave the precipitator on the side of the other electrode. The coal (containing) particles are then re-introduced into the combustor.

TNO

CHEMICAL COMMINUTION

Author : A. Ansems
Reference No. : 82-012192
File No. : 8722-50067
Date : December 1982
Language : Dutch

SUMMARY

This report gives a survey of the knowledge relating to chemical comminution especially the comminution of coal with ammonia.
The purpose of this investigation was to find the optimum conditions for maximal dissolution of mineral matter from various kinds of coal for the Dutch market.
Comparing chemical comminution with pulverisation, it is concluded that more pyritic sulphur and almost as much ash is disclosed by the chemical comminution method, this at the same particle size after size reduction.
There is no commonly held opinion about the mechanism of chemical comminution.
The study of Syracuse Research Corporation shows that the velocity and extent of comminution are dependent on:

- ammonia concentration
- pressure
- temperature and
- time of contact.

The "novelty" aspect of the investigation by TNO is that the optimal process conditions for the comminution of coals that are important to the Dutch-related situation have been established. Attention has also been given to comminution with a maximum liberation of minerals.
Apart from the "sink-float" method as an applied cleaning technique it should be possible to combine chemical comminution with other cleaning techniques.

TNO

CHEMICAL COMMINUTION (OPTIMALIZATION OF THE PROCESS)

Authors : A. Ansems
R. Bloebaum

Reference No. : 83-08424
File No. : 8722-50067
Date : June 1983
Language : Dutch

SUMMARY

This report describes the results of an investigation into the optimal situations for the physical-chemical comminution of coal by means of ammonia.
During penetration of ammonia into coal, bonding forces in the coal are broken up and the coal breaks.
The results and conclusions of our own research are compared with the data and results of research carried out at Syracuse Research Corporation and Catalytic Incorporated.
The main conclusion of this investigation is that the method is coal-selective and that it is unattractive to Holland as a coal-importing country to proceed developing comminution as an alternative to pulverizing.
Some additional conclusions are:

- it takes only some minutes to achieve comminution;
- particle size sets a limit to the comminution process: under a certain particle size comminution is no longer possible;
- the degree of comminution decreases with an increase in volatile content;
- there is (still) no relationship between maceral analysis and the degree of comminution.

CHEMICAL COMMINUTION

Authors : A. Ansems
R.K. Bloebaum
Published in : Powder Technology, 40 (1984), 265-268
Language : English

SUMMARY

Liberation of mineral components of coal can possibly be achieved by chemical comminution of coal. Combined with a suitable separation process, this may eliminate the need for removal of sulphur and ash during or after combustion.
Comminution is affected by contacting the coal with a low-molecular weight substance. Ammonia has been chosen because it is supposed to be the most effective comminution agent. The coal used in these experiments was bituminous coal.
Conclusions of this research were:

- The chemical comminution method is coal selective and therefore economically less attractive to be developed as an alternative for mechanical comminution.
- Although in the literature conclusions have been drawn concerning the selective liberation of mineral particles, it is not proved because of the lack of a suitable analysing technique.
- The research conducted demonstrates that large lumps of coal can be fractured.
- The degree of comminution of "medium volatile bituminous" and "high volatile bituminous" coal increases with an increase in content of vitrinites and a decrease in content of exinites and inertites.

STUDIES ON THE ORGANYLATION OF COAL

Author : H.A. Meinema

Proceedings : Internationale kohlenwissenschaftliche Tagung
110-113
Düsseldorf, 7-9 September 1981

Language : English

SUMMARY

In recent years reductive alkylation has been found to be a relatively mild procedure for breaking up the coal molecular structure. In principle both carbon and oxygen bonded alkyl groups are introduced. Recently Liotta et al. reported on an non-reductive highly selective alkylation procedure by which O-acidic protons in coal are specifically exchanged for alkyl groups. This report contains some results of a comparative study on a number of coals which have been methylated both by a reductive and a non-reductive methylation procedure. Furthermore, for the first time functionally substituted organic groups have been introduced into the coal structure, resulting in the formation of a water soluble coal product.

TNO

COAL LIQUEFACTION - A TENTATIVE LITERATURE STUDY

Authors : T.G. Meerbeek,
H.A. Meinema

Report No. : 83/690/4102-10288/TM/IH
Project No. : 10288
Date : May 31, 1983
Language : Dutch

SUMMARY

This report gives a short survey of recent developments in the research on coal liquefaction.
From literature data it can be concluded that apart from economic factors, insufficient knowledge of the chemical composition and reactivity of the various kinds of coal is hampering the development of commercially applicable coal liquefaction processes.
Subsequently the effects of the various kinds of coal, solvents and catalysts are discussed.
In recent years much work has been done on the catalysis of the minerals in coal.
A short survey is given of the refining of crude products from coal liquefaction to liquid fuels and chemical base materials.

METHOD AND DEVICE FOR PREPARING A FLUID HYDROCARBON PRODUCT FROM COAL

European patent application

Inventor : J.L. den Otter
Application no.: 83200644.9

SUMMARY

Coal is continuously fed into an extruder and reacts therein at elevated pressure and temperature and possibly in the presence of a catalyst with hydrogen and/or hydrogen containing compounds added to the contents of the extruder on at least one spot situated at a distance of at least three times the inner diameter of the extruder in front of the end of the screw. From the reaction product leaving the extruder hydrocarbons which are fluid under normal conditions are separated.
Part of these hydrocarbons which are liquid under normal conditions may be recycled to the extruder.

PARTIAL MICROBIAL OXIDATION ON PYRITE FOLLOWED BY OIL-AGGLOMERATION

Author : J. Doddema

Presented at : The International Symposium of Biohydrometallurgy
Cagliari, May 1-3, 1983
Language : English

SUMMARY

Oxidation of pyrite surfaces in coal may lead to enhanced pyrite rejection during oil agglomeration of coal. In this study growth of Thiobacillus ferrooxidans on pyrite, the partial oxidation of pyrite in coal by this culture, and oil-agglomeration of the treated coal were studied.

A mixed culture containing T.ferrooxidans showed irreproducable growth on pyrite. Only at very low ionic strength and pH appreciable quantities of pyrite are oxidized. At higher ionic strength and pH large quantities of basic ferric sulphates are deposited.

For the partial oxidation of pyrite surfaces in coal the mixed culture, grown on pyrite or sulphur, was mixed with a coal slurry under growth conditions. All cells adhered to the coal particles preventing recirculation of cells.

Oil-agglomeration of the microbiologically treated coals did not show enhanced pyrite rejection when compared with untreated coals.

Based on the results obtained so far, partial microbial oxidation of pyrite in coal followed by oil-agglomeration does not promise to be an attractive coal cleaning process under practical conditions, with the low pyritic sulphur coals used in this study.

MICROBIAL DESULPHURIZATION OF COAL: PARTIAL MICROBIAL OXIDATION OF COAL FOLLOWED BY OIL-AGGLOMERATION

Author : H.J. Doddema
Report no. : 83/01408/4302-50052/HD/GK
Order no. : 4351
Date : October 1983
Language : Dutch

SUMMARY

Oxidation of pyrite surfaces in coal may lead to enhanced pyrite rejection during oil-agglomeration of coal. In this study growth of Thiobacillus ferro-oxidans on pyrite, the partial oxidation of pyrite in coal by this culture, and oil-agglomeration of the treated coal were studied.

A mixed culture containing T.ferrooxidans showed irreproducable growth on pyrite. Only at very low ionic strenth and pH, appreciable quantities of pyrite are oxidized. At higher ionic strength and pH, large quantities of basic ferric sulphates are deposited.

For the partial oxidation of pyrite surfaces in coal the mixed culture, grown on pyrite or sulphur, was mixed with a coal slurry under growth conditions. All cells adhered to the coal particles preventing recirculation of cells. Oil-agglomeration of the microbiologically treated coals did not show enhanced pyrite rejection when compared with untreated coal.

Based on the results obtained so far, partial microbial oxidation of pyrite in coal followed by oil-agglomeration does not promise to be an attractive coal cleaning process under practical conditions with the low pyritic sulphur coal used in this study.

MICROBIAL COAL DESULPHURIZATION

Authors : H.J. Doddema
T.F. Huber
C.H. Kos
P. Bos

Presented at : Coal Technology Europe
3rd European Conference & Exhibition on Coal Utilization, 11-13 October 1983, Amsterdam
Volume 3, 183-192

Language : English

SUMMARY

Desulphurization of coal by microorganisms can be done in two different ways: by the complete oxidation process and by the microbial agglomeration process.
Comparing the two varieties of the microbial desulphurization process, one can conclude that although a detailed feasibility study has not been made, the complete oxidation process offers more possibilities than the microbial agglomeration process. The complete oxidation process is not especially coal specific and high pyrite removal percentages have been obtained.
Using oil agglomeration only low removal percentages have been reached.
In the complete oxidation process some bottle-necks have been noticed. First of all, the long residence times. These may be reduced by running the process continuously because the lag phase of growth will be avoided. Theromophilic pyrite oxidizing bacteria may also be useful in reducing the residence times. In this context Detz and Barvinchak (1979) and Kargi (1982) report the use of Sulfolobus species. Another problem is the precipitation of jarosites in the coal. An improvement can be achieved by using a lower pH and by minimizing the ionic strength of the process water.
Independent of whether microbial desulphurization processes will be feasible, in coal technology we must deal with microorganisms. Thio-

bacillus ferrooxidans and related organisms will become active in every system where pyrite, water and air are present. We may expect the occurence in coal piles, especially where the coal is finely dispersed, of analogous microbial processes to those in some coal mines, which result in a highly polluted drainage water with a low pH and rather high concentrations of heavy metals (acid mine drainage water). Especially when transport and storage of pulverised coal and coal-water slurries are considered Thiobacillus ferrooxidans will offer a serious environmental and corrosion problem.

PRELIMINARY CALCULATIONS OF THE SO_2, SOOT, NO_x AND CO CONCENTRATIONS IN THE ATMOSPHERE WHEN CHANGING OVER THE GREENHOUSE HEATING IN "HET WESTLAND" FROM GAS TO COAL

Authors : C. Huygen
J. Hulshoff
C. Veldt

Report no. : CMP 82/03
Date : March 1, 1982
Language : Dutch

SUMMARY

Firing coal in stead of gas in the greenhouses of "the Westland" will resulting in a large decrease in air quality.
The MAC values for soot will possibly be passed and certainly those for SO_2 when no energy-saving measures are taken or the emissions are reduced.

TNO'S ENVIRONMENTAL PROGRAM ON FLUIDIZED BED COMBUSTION

Author : P.J. van Duin

Presented at : The IEA AFBC technical meeting at Petten, The Netherlands, May 1982

Language : English

SUMMARY

TNO carries out an environmental research program on fluidized bed combustion of coal. Tests are executed with a 4 MW experimental atmospheric fluidized bed boiler. To reduce the emission of sulphur dioxide, coal is mixed with limestone. The limestone turned out to be used partially, due to the formation of a calcium sulphate layer on the surface of the limestone particles. The conclusion was confirmed by X-ray mapping. Fly ash was investigated on harmful properties. When collected at low temperatures fly ash showed substantial genotoxicity.

AIR POLLUTION FROM COAL

Author : W.A.M. den Tonkelaar
Report no. : G 1236
Project no. : 7.2.04 1045-017
Date : February 1983
Language : Dutch

SUMMARY

The consequences for the air quality are investigated by means of the Calculating System Air Pollution when changing to coal combustion in a fluidized-bed combustor or a chain grate in the Dutch heated greenhouses. The conclusion is that with a stationary background concentration of the air pollution the limits for SO_2, NO_x, NO_2 and soot will not or only locally, be exceeded.
Calculations of the increase of the annual average concentrations of benz(a)-pyrene give in the "Westland" higher values than the VDI standard when using the highest value of the maximum emission factor found in the literature.

AN AIR QUALITY MANAGEMENT SYSTEM AS A TOOL FOR ESTABLISHING A SO_2- AND NO_x-POLICY

Authors : M. Bovenkerk
P.J.H. Builtjes
S. Zwerver

Published in : Atmospheric Environment, 18 (1984), 3, 519-529
Language : English

SUMMARY

A description is given of the development and use of an Air Quality Management System (AQMS) directed to the support of policy decisions made with respect to the conservation and improvement of clean air in the Netherlands. The AQMS consists of three separate modules oriented to economy, transmission and impact assessment.
Application of the AQMS to SO_2 has led to the construction of an environmental decision scheme, in which maximum acceptable emission levels are given as a function of the primary and secondary SO_2-air quality standard, the S import-export balance and acidification.
Together with information on the socio-economic impacts and the energy options this formed the basis for the political decision to limit future SO_2-emissions in the Netherlands to a value not higher than 500×10^6 kg/year, in fact to establish a ceiling value for the emission of SO_2.
For NO_x the AQMS is still under development, so at the moment it is not possible to formalize a complete NO_x-environmental decision scheme. Up till now the emphasis has been on the development of the transmission module of the AQMS for NO_x, i.e. the impact of domestic and foreign emissions. The results of this module are presented in the paper.
The results of the AQMS-calculations point out the importance of long-range transport of NO_x and photochemical precursors and products for the countrywide NO_2-pattern. The results emphasize the need for the abatement of NO_x (and especially hydrocarbons (HC)) on a European scale. Also abatement of automobile exhaust is needed in order to prevent violation of the NO_2-standard at sites with high circulation of traffic.

PLUME DISPERSION MODELS OF COAL-FIRED POWER PLANTS WITH RESPECT TO CHEMICAL TRANSFORMATION AND DRY DEPOSITION

Authors : P.J.H. Builtjes
G.L.H. Beugeling
J. Vijge

Report no. : 85-01234
File no. : 8724-50055
Date : January, 1985
Language : Dutch

SUMMARY

A chemical reactive plume model for tall stacks directed to NO-NO_2 conversion is discussed and its results are compared with measured values of power station plumes.
The model predicts the NO/NO_x ratio for 74% of the measurements within the reliability range. At short distance from the source the prediction is alternately too high and too low, at long distance too low.
The model is described and the found discrepancies are explained.
Calculations show that the influence of the dry deposition on calculated NO_x-concentrations for tall stacks can be neglected for distances up to about 50 km from the source.
A very simple model, accounting for only one terminating reaction for the NO-NO_2 conversion, predicts the NO/NO_x ratio poorly and shows the advantage of the proposed model.

LIMITATION OF THE NO_x-EMISSION FROM COAL-FIRED FLUIDIZED-BED BOILERS

Author : F. Bergsma
Report No. : CL 79/68
Date : July 13th, 1979
Language : Dutch

SUMMARY

A literature investigation has been made into the limitation of NO_x-emissions from coal-fired installations, especially fluidized-bed boilers. An investigation has been carried out in order to plan further research with the object of limiting NO_x-emissions.

LIMITING THE NO_X-EMISSION IN COAL-FIRED FLUIDIZED-BED COMBUSTORS

Author : F. Bergsma
Report No. : CL 81/190
Order No. : 50025
Contract No. : 4.351-2.1
Date : December 29th, 1981
Language : Dutch

SUMMARY

As a result of a study of recently published literature the technological possibilities have been indicated of decreasing NO_x-emissions from fluidized-bed combustors. Some improvements have earlier been proved to be effective, like two stage combustion and fitting internals (baffle screen). Other technologies the effectiveness of which has not yet revealed itself are: an improved coal feed and/or a separate feed of char and volatile constituents.

It is not yet possible to make predictions by means of a calculation model, although considerable progress has been made with mathematical descriptions of combustion, transport and reactions in fluidized-bed combustors.

The total of the concentrations of NO, NH_3 and HCN should be minimal to reach a maximal conversion of fuel-N into N_2.

ABATEMENT OF NO_x FROM COAL COMBUSTION

Author : F. Bergsma
Report no. : P 82/86
Date : November 25th, 1982
Language : Dutch

SUMMARY

The chemical background of NO_x abatement at the combustion of coal is described. The results so far obtained by reducing NO_x produced by pulverized coal combustion and those originating from the combustion of coal in fluidized bed combustors are analysed. The results are used to make suggestions for further reduction of NO_x emissions from fluidized bed combustors.

INVENTORY OF MEASUREMENTS OF NITROGEN MONOXIDE OXYDATION IN STACK PLUMES

Author : P.E.J. Vermeulen
Reference no. : 83-01620
File no. : 8724-50055
Date : April 1983
Language : Dutch

SUMMARY

Chemical-reactive plume measurements are analyzed and inventorized. The measurements considered were measurements on flue gas plumes of electric power plants. Eleven measurements were selected as test cases for the evaluation of chemical reactive plume models.

ABATEMENT OF NO_x FROM COAL COMBUSTION; CHEMICAL BACKGROUND AND PRESENT STATE OF TECHNICAL DEVELOPMENT

Author : F. Bergsma
Published in : Ind.Eng.Chem. Process Des.Dev., 24 (1985), 1, 1-7
Date : January 1985
Language : English

SUMMARY

The chemical background of NO_x abatement at the combustion of coal is described. The results so far obtained by reduction of NO_x emissions from pulverized coal combustion and those originating from the combustion of coal in fluidized bed combustors (FBC's) were analyzed. The discussion is restricted to process modification techniques. Flue gas purification is not included. The results were used to make suggestions for further reduction of NO_x from FBC's.

OPTIMIZING THE USE OF SULPHUR SORBENTS IN THE FLUIDIZED BED COMBUSTION OF COAL

Author : D. Schmal
Report No. : CL 82/77
Order No. : 50900
Date : July 6th, 1982
Language : Dutch

SUMMARY

Within the scope of research on fluidized bed combustion of coal and its environmental aspects the sorption of the sulphur dioxide released during the combustion, has been studied so far. Aim of this research is to optimize the use of sorbents in such a way that their quantities needed are minimal, that the sulphur dioxide sorption is maximal and that the possibilities for application of the remaining waste are optimal. Research of this kind is needed, because application of sulphur sorbents enlarges the quantities of waste significantly. Besides, future bottlenecks concerning the availability of suitable sorbents play a role. On the basis of a previous literature survey it has been decided to carry out research on the re-activation of the waste from fluidized bed combustion. This re-activation consists of a treatment with water vapor (or water in the liquid state) or possibly other gases, which restores the reactivity of partly sulphated sorbents.

To get an insight into the processes taking place, at first a laboratory research program was started on the sulphation and re-activation of the sorbents used. Next, the same kind of research was started with the waste from TNO's 4 MWth atmospheric fluidized bed boiler (AFBB). This waste contains, apart form partly sulphated limestone, also ash and unburned coal.

Apart from the results of these laboratory experiments, this report also contains the results of the first experiments concerning the sulphur retention in the 4 MWth AFBB. Furthermore, by means of experimental data, an American method to predict the sulphur retention in a fluidized bed combustor, on the basis of relatively simple laboratory experiments, is discussed.

From the laboratory experiments it can be concluded that, after re-activation, the conversion of sorbents will be greatly enhanced. On the basis of these results and those of experiments still taking place, a pilot plant re-activation unit will be designed and built. Such a unit will be used to examine the technical and economical feasibilities. Reliable data for an unambiguous interpretation of the results concerning the first tests with the 4 MWth AFBB and that of the mentioned prediction methodology for sulphur retention, are still lacking. The efforts to obtain those data will be continued after further runs of the 4 MWth AFBB.

It has, however, become clear form these experiments that, even without special provisions and with relatively small quantities of sorbent, the sulphur dioxide emissions can be restricted to such an extent that they do not reach the maximum (230 g SO_2/GJ) proposed in 1981 by the Dutch Ministry of Housing, Physical Planning and Environment for new installations.

IMPROVING THE ACTION OF SULPHUR SORBENTS IN THE FLUIDIZED BED COMBUSTION OF COAL

Author : D. Schmal
Report No. : P 83/7
Date : March 19th, 1983
Language : English

SUMMARY

As part of an investigation into the fluidized bed combustion of coal and its environmental aspects, TNO is studying the sorption of sulphur dioxide as a combustion product. Special attention is being paid to reactivation of fluidized bed ash by treatment with water or water vapour, followed by heating to fluidized bed temperatures.
A laboratory research programme was started on sulphation and reactivation of sorbents on their own and of sorbent containing ashes from TNO's 4 MWth atmospheric fluidized bed boiler (AFBB).
The experiments show that reactivation can be a versatile method for improving the efficiency of the sorbents.

THE SULPHUR BINDING PROPERTIES OF AGGLOMERATED LIMESTONE

Authors : P.J. van der Driest
D. Schmal
Report No. : R 82/81
Order No. : 10274
Date : May 11th, 1983
Language : Dutch

SUMMARY

For the sorption of sulphur dioxide in a fluidized bed combustor mainly either limestone or dolomite is used. So as to run a plant economically, the necessary quantities of sorbent have to be restricted. The applicability of the solid matter also plays a role in this process.
Under normal conditions only 15 to 30% of the sorbent available is used. This is mainly caused by pore plugging effects of the calcium sulphate formed, which in turn is a result of its relatively large molecular volume. The sulphur sorption can be enhanced by improvement of the pore structure. In this report the results are discussed of laboratory research into the sulphur sorbing properties of limestone which has been ground and agglomerated beforehand. For a comparison also the properties of the same limestone, but now in its usual form (broken), were studied.
From the experiments it can be concluded that agglomeration beforehand will give much better sulphur sorption properties. Despite the higher costs of agglomerated limestone, this can result in lower limestone consumption and thus lower costs and a reduction in the quantities of waste.
Because (the difference in) attrition properties of both types of limestone (agglomerated and broken) are not known, a definitive assessment can only be done after experiments in a fluidized bed combustor of an appropriate size.

METHODS FOR THE REDUCTION OF THE QUANTITIES OF SULPHUR SORBENTS IN FLUIDIZED BED COMBUSTION

Author : D. Schmal

Presented at : Coal Technology Europe,
3rd European Conference & Exhibition on Coal Utilization,
11-13 October 1983, Amsterdam
Volume 2, 145-160

Language : English

SUMMARY

One of the main aspects of the air pollution caused by the combustion of coal is the formation of sulphur dioxide. In view of the increasing use of coal and the adverse effects of sulphur dioxide on the environment and on human health it is necessary to prevent a substantial part of the sulphur dioxide from escaping to the atmosphere. In fluidized bed combustion this is accomplished by adding sulphur dioxide sorbents, such as limestone or dolomite to the bed.

Owing to pore plugging of the sorbent particles, only some tens of per cents of the available sulphur sorption capacity is used and therefore several methods have been proposed so far to improve the sulphur sorption in order to restrict not only the quantities of sorbents needed but also the amount of the solid waste. So as to be succesful these methods should be simple and cheap and without adverse effects on the combustion process. These conditions severely restrict the choice of methods. Regeneration processes, for instance, are relatively complicated and the regeneration temperature is higher than the temperature in the fluidized bed combustor.

Our research is focussed on two, relatively simple, methods to restrict the quantities of sorbents. One method is based upon reactivation of the partially sulphated sorbents. The other on the use of pulverized and subsequently agglomerated limestone. In the experiments the reactivity of the limestones and ashes to sulphur sorption was determined with a thermogravimetric analyzer.

IMPROVING THE ACTION OF SULPHUR SORBENTS IN THE FLUIDIZED BED COMBUSTION OF COAL

Author : D. Schmal
Published in : Ind.Eng.Chem.Process Des.Dev., 24 (1985), 1, 72-77
Language : English

SUMMARY

As part of an investigation into the fluidized-bed combustion of coal and its environmetal aspects, TNO is studying the sorption of sulphur dioxide as a combustion product. Special attention is being paid to reactivation of fluidized-bed ash by treatment with water or water vapor, followed by heating to fluidized-bed temperatures. A laboratory research program was started on sulphation and reactivation of sorbents on their own and of sorbent-containing ashes from TNO's 4 MW atmospheric fluidized-bed boiler (AFBB). The experiments show that reactivation can be a versatile method for improving the efficiency of the sorbents.

A METHOD FOR WATER TREATING SULPHUR BINDING ADDITIVES UPON BURNING SOLID FUELS

European patent application

Inventor : D. Schmal
M.L.G. van Gasselt

Application no.: 84200379.0

SUMMARY

In a fluidized bed combustion of solid fuels the efficiency of sulphur dioxide binding additives is improved by contacting the partially sulphated additive with fuel to be burnt and including the required amount of water.

TNO

POLYCYCLIC AROMATIC HYDROCARBONS FROM COAL-FIRED INSTALLATIONS

Author : A.C. Besemer
Report No. : CL 82/146
Order No. : 50053
Date : October 25, 1982
Language : Dutch

SUMMARY

In this report a survey is given of the literature dealing with emissions of polycyclic aromatic hydrocarbons (PAH's) in flue gases of coal-fired installations and the sampling methods and analytic methods used, together with the problems involved. Measurements carried out up to now have shown that big pulverised-coal fired installations contribute only slightly to the total emission of PAH's; the emission factor is about 0.01-120 mg PAH/ton of coal combusted.
Smaller conventional installations can give a relatively large contribution to the total emission of PAH's; the maximum emission factor can be 1,100 mg PAH/ton of coal combusted.

ISOLATION OF BENZ(a)ANTHRACENE-^{14}C AND BENZO(a)PYRENE ADSORBED ON FLY-ASH BY MEANS OF LIQUID EXTRACTION

Authors : A.C. Besemer
Th. Lems
Report no. : R 84/76
Order no. : 10273
Date : May 28th, 1984
Language : Dutch

SUMMARY

Polycyclic Aromatic Hydrocarbons are absorbed by fly-ash. The amount absorbed depends of the character of the hydrocarbon. Benz(a)anthracene (4 rings) is less absorbed than benzo(a)pyrene (5 rings). The character of the fly-ash plays a role, too. As a result of these investigations it was found, that the absorption increases with the amount of C in the fly-ash. When a solvent is added to the ash, it has been found that the PAH divides between the solvent and the ash depending on the concentration. There is a saturation of the fly-ash surface when big amounts of PAH are added. The desorption behaviour of PAH from fly-ash was investigated with the purpose to remove the PAH as completely as possible. The solvents hexane, cyclohexane, dichlormethane and acetone were not suitable to remove PAH. However, good results were obtained with toluene and xylene.

With a load of 100 µg PAH/g fly-ash, the PAH could be nearly quantitatively recovered by ultrasone extraction with xylene at 60°C. At a low load (5 µg/g) 60% of the benz(a)pyrene was recovered with 3 extractions with toluene. It was found that the absorption of benz(a)anthracene is not so strong as that of benzo(a)pyrene and, consequenty, the desorption was easier.

The desorption is easier, too, when the C-content of the fly-ash is lower. This means that quantitative recovery from FBC-ash with a high C content is difficult.

ENVIRONMENTAL ASPECTS OF COAL FIRING IN SMALL INDUSTRIAL INSTALLATIONS

Authors : J.A. Heil ECN
P.T. Alderliesten ECN
F.C. Poot TNO
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Presented at : Symposium clean combustion of coal
Leeuwenhout Congrescenter, Noordwijkerhout,
16, 17 January 1985

Language : Dutch

SUMMARY

A research program has been carried out at some coal fired drying installations and heated greenhouses.
In this paper the following three aspects are discussed:

1. The composition of the ash produced in relation to the type of coal fired.
2. The flue gas emissions regarding:
   - macrocomponents SO_2, NO_x, C_xH_y, CO
   - dust particles, heavy metals and halogenes
   - PAH in connection with boiler type, operation mode and coal type.
3. The cytotoxicity and mutagenicity of emitted fly ash and fly ash collected.

TOXICOLOGICAL PROPERTIES OF RESPIRABLE FLY ASH FROM VARIOUS COAL-BURNING INSTALLATIONS

Authors : G.J. Vink
P.J. van Duin
P.B. Davis
R.N. Hooftman

Report no. : D 84/114
Date : February 1, 1985
Language : Dutch

SUMMARY

An investigation of the toxicological properties of captured fly ash and stack ash originating from various combustion techniques has been carried out as part of the Dutch National Coal Research Program. Samples from the following installations were investigated: a pulverized coal combustor (PCC, 120 MW_e, an experimental atmospheric fluidized bed combustor (AFBC-E, 4 MW_{th}) and two coal-fired installations in use for greenhouse-heating, an underbedfeeder (UBF, 3.8 MW_t) and an atmospheric fluidized bed combustor (AFBC-G, 3.4 MW_t).
This report summarizes and evaluates the results of the investigations carried out in the period 1982 - 1984; more detailed information is presented in several separate reports.

The toxic samples were investigated in tests for genotoxicity (mutations in the Ames Salmonella test and in a test in Paramecium, chromosomal aberrations in CHO cells) and cytotoxicity (survival and effect on phagocytosis in bovine alveolar macrophages). Some samples from PCC were also investigated in single application tests for primary skin and eye irritation, and for sensitization. Some environmental toxicity tests were carried out with aqueous or organic extracts of PCC fly ash. The polycyclic aromatic hydrocarbons (PAH) and inorganic content of a number of samples was also investigated.
Organic extracts of stack ash samples were genotoxic. Mutagenicity in the Ames test (strain TA98S9, expressed as revertants per m^3 flue gas) was highly dependent on the installation. Low values were found for

AFBC-E (<20), intermediate values for AFBC-G (400-650) and PCC (780-1160, in one case 12500) and very high values for UBF (20.000 under full or partial load and 64.000 at low energy (and low flue gas) production). The intermediate values are of the same order of magnitude as reported in literature for other installations.

Organic extracts of captured flyash from PCC and AFBC-E were not mutagenic in the Ames test, but samples from AFBC-G (75 revertants/mg ash) and UBF (1300) were highly mutagenic. Only a small part of the mutagenic activity (1-6%) was emitted to the atmosphere from these latter two installations; by far the greater part was recovered in the bagfilter-ash.

Metabolic activation had generally little effect on the mutagenic activity of bag filter or stack ash. About half of the mutagenicity was nitro-reductase dependent, indicating that nitro-compounds are partly responsible for the effects.

Samples from UBF and AFBC-G which were very mutagenic in the Ames test caused a number of chromosomal aberrations in CHO-cells; samples from the other installations are still under investigation.

Captured fly ash from the electrostatic precipitators (ESP) from the PCC. which showed no mutagenicity in the Ames test, appeared to be mutagenic in Paramecium, an unicellular organism able to ingest fly ash particles. This result indicates that inorganic compounds can probably also be responsible for the effect.

Captured fly ash samples, from all four installations showed only slight cytotoxic effects in bovine alveolar macrophages, free lung cells which ingest inhaled viable and non viable particles. These effects were comparable with those caused by inert material such as glass beads. However the ash samples did contain a small percentage of fine particles which were far more cytotoxic than the bulk ash samples. Stack ash from PCC appeared to be more cytotoxic than ESP ash; stack ash samples from the other installations have not yet been investigated in the macrophage test. The adverse effects of PCC stack ash are comparable with or even higher than those of α-quarts. The adverse effects on phagocytic capacity are generally more pronounced than those on cell viability.

The in vivo irritation tests revealed that total PCC ESP ash, final zone ESP ash (enriched in the smallest particles) caused very slight skin and eye irritation effects and that the corresponding stack ash caused

moderate eye irritation. None of the samples gave rise to sensitization. Irritation effects have never been demonstrated in workers with PCC fly ash, and can probably be neglected.

With regard to the environmental toxicity tests ESP ash extracts from the PCC appeared to be practically non toxic in the chronic Daphnia test; slight effects were found with extracts of final zone ESP ash. Slight effects were observed in a plant seed germination/growth test with the same ESP ash samples; this toxicity was probably caused by a high boron concentration.

Chemical analysis revealed low carcinogenic PAH emission for PCC and AFBC-E (10-30 ng/m^3) and high emission for AFBC-G and UBF (200-600 and 73.000 ng/m^3 resp.). The carcinogenic PAH content of ESP ash from PCC (₹0.14 mg/kg) and bag filter ash from AFBC-E (0.04-0.14 mg/kg) are very low. A morphologically new type of particle has been discovered in AFBC-G bag filter ash; these particles (carbonaceous cenospheres) contain at least 75% carbon as a small membrane around a skeleton of minerals.

Calculations indicate that the toxic properties of PCC stack ash hardly contribute to the present mutagenicity of aerosols or to the elemental concentration in ambient air. The smaller installations (AFBC-G and UBF) may be expected to make a significant contribution to the ambient air concentrations on account of their highly mutagenic stack ash and the small stack height resulting in low dilution of fluegas. This point must, however, still be verified by investigations in situ.

Health effects from handling of highly mutagenic captured fly ash, as found at UBF and AFBC-G, cannot be excluded. More data on real exposure of respiratory organs and bioavailability of fly ash components under physiological conditions (in the lungs) are needed for better evaluation of health risk of the smaller coal-burning installations.

The results for the samples from the experimental AFBC-E indicate that fluidized bed combustion can be a rather clean process. The influence of combustion conditions on the toxic properties of fluegas has yet to be investigated.

TRACE ELEMENT EMISSIONS FROM PULVERIZED-COAL FIRED POWER PLANTS

Author : M.E. Reinders
Reference No. : 82-03068
File No. : 8728-40142
Date : April 28th, 1982
Language : English

SUMMARY

Coal consists of minerals and organic matter (macerals). Trace elements like copper and cobalt are incorporated in the crystal grid of iron sulphide. Clay compounds can likewise contain trace elements in their crystal grid. During combustion of the macerals the minerals in the coal melt and ash is formed. The mineral droplets solidify during flue gas cooling and leave the stack as fly ash. All SO_2 and NO_x passes the precipitator, and so does 70% of the mercury, and more than 90% of the bromine. Some of the trace elements show an increase in concentration as a result of the transition from coal to precipitator ash, and the transition from coal to fly ash, dependent on the volatility of the trace elements.
During the combustion of coal radio-active compounds are emitted, too. The concentration of radio-active compounds in the fly ash is higher than in the coal.

TRACE COMPONENTS: EMISSIONS FROM COAL FIRED POWER PLANTS

Authors : C. Veldt
C. Huygen
J.A. van Jaarsveldt
D. Onderdelinden

Report no. : G 1251
Date : March 1983
Language : Dutch

SUMMARY

The purpose of the NOK project LUK-C3 "Background concentration measurements" is to assess the present background concentrations of organic as well as inorganic trace components and to estimate a possible contribution in future from the coal fired electric utilities. The environmental significance of this contribution is not considered in this project.

It has been found, that reasonable estimations were possible based on coal composition and efficiency of the fly-ash collectors. Some elements are emitted quantitatively or for the main part as a gas (B, Br, Cl, F and Hg). The emissions of the other elements can be estimated by means of the enrichment factors. Elements with a greater volatility at the temperature of the furnace (As, Cd, Ga, In, Pb, Se, W, Sn, Zn, Tl) have an enrichment factor of about 5; the elements with a smaller volatility (Ag, Ba, Co, Cr, Cu, Mo, Ni, U, V, Ge, Li) have an enrichment factor of about 3. Elements with a negligible volatility (Al, Be, Ca, Cs, Fe, Hf, K, Mg, Mn, Na, Rb, Sc, Sr, Ta, Te, Th, Ti, Zr, rare-earth elements) have an enrichment factor of about 1. Based on these data, the data of the coal combustion, a dust collection efficiency for fly ash of 99,7% and an expected coal consumption of 30×10^6 tons/year, the emissions expected and the concentrations in flue gas have been calculated.

It is more difficult to estimate the emissions of organic (trace)components. This resulted in some more or less reliable data with a considerable variation. The PAH emission is the best known from literature. The scarce data indicate that the main part of the organic emission consists of (not-identified) acids. It can be concluded that the emissions of organic compounds will not increase (with the possible exception of organic acids), based on the comparison of the expected emissions from the power plants with the total Dutch emissions. However, it is possible that the emissions of some trace elements will increase strongly: B, Be, Hg, Sr, the halogens as well as Al, As, Co, Mn, Sb, Ti and the other (earth)alkali metals.

The relative contributions to the annual concentrations at ground level have been calculated with two dispersion models, based on the present situation of the power plants, and some other simplified propositions; the results give only some relative indications.

CAPTURE OF NaCl AEROSOL BY AN ELECTROFLUIDIZED GRANULAR BED FILTER

Author : P.H. de Haan
Report No. : 333/'82
Date : September 30, 1982
Language : English

SUMMARY

The possibility of the capture of submicron fly ash particles from flue gas is an important condition for the use of coal burning installations. We determined the capturing efficiency of a granular bed filter electrified with an external field. The filter has been tested using NaCl-aerosol. The best results with the electrofluidized filter are obtained when the aerosol is precharged with a corona charger. At a certain air flow velocity the efficiency is optimal.
This velocity depends on the grain diameter. At a flow velocity of v_s = 0.35 m/s and sand grains of 0.5-0,6 mm the efficiency amounts to $\eta > 99\%$ for particles with a diameter $d_p > 0.35$ µm. The pressure drop across the bed is 620 Pa. The high efficiency for the capture of submicron particles at the relatively high flow velocity and low pressure drop is encouraging for the further development.

COLLECTION OF FLY-ASH FROM THE FLUE GASES OF AN ATMOSPHERIC FLUIDIZED BED BOILER BY MEANS OF ELECTRET FILTERS

Authors : P.H. de Haan
J.L.B. de Groot

Report No. : 145/83
Project No. : 145/83
Date : May 18th , 1983
Language : Dutch

SUMMARY

The greater part of the fly-ash formed with coal combustion is collected from the flue gases by cyclones, electrostatic precipitators or by fabric filters. The inhalable part of the fly-ash (paricle sizes from 0.2 to 2 μm) is hardly collected by these filters, however. To collect also this fraction new filters are being developed.

In this report a number of experiments have been described for collecting the inhalable fly-ash particles by means of electrostatic collectors. Most experiments have been carried out using a sand-bed filter; in one experiment a fiber-bed filter has been used. The sand grains and the fibers are polarized by means of an electric field ($E = 5 \times 10^5$ V/m). The fly-ash particles charged by a corona in front of the filter are collected by the polarized grains of fibers with high efficiency as a result of the strong electrostatic forces.

The filters have been tested with flue gas originating directly from the flue duct of a coal-fired fluidized bed boiler. Flue gas with a very high dust concentration (> 10 g/m^3) and flue gas with a low dust concentration (< 10 mg/m^3) were used. The fiber bed was tested with the low dust concentration only.
The collection efficiency, η, was very high: $\eta > 99\%$ for particles of 0.2 μm and $\eta > 99.99\%$ for particles of 5 μm, at a gas velocity of 0.15 m/s. The initial pressure drop was about 0.5 kPa.

FILTRATION OF INHALABLE FLY-ASH PARTICLES BY AN ELECTRIFIED GRANULAR BED FILTER

Authors : P.H. de Haan
J.L.B. de Groot

Presented at : Coal Technology Europe,
3rd European Conference & Exhibition on Coal Utilization,
11-13 October 1983, Amsterdam
Volume 4, 149-160

Language : English

SUMMARY

To capture the inhalable fraction of fly-ash particles (0.2 - 2μm) from coal burning installations efficiently, new types of filters are being developed. In this paper some filtration experiments are described with a sand-bed filter. The sand grains are polarized by a high electric field. The fly-ash particles, which are corona charged before the filter bed, are efficiently captured because of the electrostatic forces between the fly-ash particles and the polarized sand grains.
Several bench-scale filters were tested with flue gas of the Atmospheric Fluidized Bed Boiler (AFBB) of TNO at Apeldoorn. The filters were charged with flue gas both with a high (> 19 g/m3) and a low volume concentration (≃ 10 mg/m3) of dust. The capturing efficiency, η, is high, viz. $\eta > 99\%$ for 0.2 μm particles and $\eta > 99{,}9\%$ for 2 μm particles. The face velocity of the gas is 0.15 m/s, and the initial pressure drop over the filter about 0.5 kPa.

EFFICIENCY AND PRESSURE DROP FOR THREE TYPES OF FILTER MATERIAL: GLASS FIBER FELT, GLASS PAPER, ELECTRIFIED GRANULAR BED

Author : P.H. de Haan
Report no. : 169/'84
Date : May 17, 1984
Language : Dutch

SUMMARY

The efficiency and pressure drop during dust loading has been measured of three different filter materials, namely glass fibre felt, glass paper, and an electrified fixed granular bed filter. In all cases a flat clean sample of material has been used. The samples are loaded with resuspended fly ash from a pulverized coal combustor. The gas velocities were 0.05 m/s and 0.2 m/s, and the dust concentration about 1 g/m^3.

The initial pressure drops across the glass fibre felt and the granular bed filter are approximately equal. The initial pressure drop across the glass paper is a factor of 6 to 8 higher.

The capture of dust in the glass fibre felt is based on "cake filtration". The initial penetration is high, but is reduced drastically by the formed dust cake.

In the glass paper filter, too, the deposition of dust occurs at the surface of the filter, but the penetration is low, also at low dust loading. The increase of pressure drop is equal to that of the glass fibre felt.

The dust capture by the electrified granular filter is based on in-depth filtration. A surface layer of dust is hardly formed. The increase of the pressure drop is less than in the case of the fibre filters and independent of the gas velocity. The penetration for submicron particles is higher, but almost independent of the dust loading of the filter. The granular filter shows a somewhat lower filter efficiency, but also a lower pressure drop than both the fibre filters.

AN ELECTRIFIED GRANULAR BED FILTER AS A FLY-ASH COLLECTOR FOR COAL FIRED COMBUSTORS

Author : P.H. de Haan
Published in : Energiespectrum, 9 (1985), 1, 23-29
Language : Dutch

SUMMARY

Coal contains 10-15% minerals which form for the greater part fly-ash during combustion. There are indications that fine fly-ash particles (0.2-2 µm) are dangerous for health. These particles are inhalable; they can contain many toxic compounds. This is the reason that inhalable fly-ash should be removed from the flue gas, before the flue gas is emitted from the stack. Dust collectors now in use in coal combustors are electrostatic precipitators, cyclones and wet collectors. For the total fly-ash, they have a collection efficiency of 99%, however, they have a poor collection efficiency for fine fly-ash dust.
Fabric filters have a high collection efficiency. Disadvantages are high pressure drop over the filter, low working temperature (< 200°C) and the high cost of the fabric material.
A new development for collecting small particles is the electrified granular bed filter. It has a small pressure drop. The dust particles are electrically charged and then collected into a granular bed, which granules are electrically polarized. By means of these electrostatic fields a high dust collection efficiency can be reached.
In this publication the results are discussed of an investigation into the possibilities to use this filter for collecting inhalable fly-ash.

WIND-TUNNEL INVESTIGATIONS INTO STORAGE YARDS FOR BULK MATERIALS

Author : G.Th. Visser
Published in : PT/Procestechniek, 37 (1982), 12, 29-33
Language : Dutch

SUMMARY

When storing bulk materials in the open air, attention should be given to dust emissions as requirements by law are more and more severe. By means of wind-tunnel investigations the amount of dust can be measured the wind blows from the storage piles as a function of a number of factors of influence, such as wind velocity, and humidity of the stored material. The effect of dust reducing measures can be evaluated.

DUST EMISSIONS FROM STORAGE AND TRANSHIPMENT

Authors : L.A.M. Janssen
J. den Boeft
Reference No. : 82-07624
File No. : 8724-30053
Date : June 1982
Language : Dutch

SUMMARY

Investigations have been carried out into the way of measuring emissions and concentrations of coal dust.
In the wind tunnel and during field measuring, techniques have been applied for determining concentrations and deposits and investigated on their practical applicability.
From the results obtained conclusions have been drawn with respect to wind velocities at which emissions can occur as a result of wind erosion under practical conditions.
Moreover an insight has been gained into the load levels in practice at low wind velocities (< 7 m/s) on the basis of information obtained by the measurements in the field.
The limited scope of these investigations does not allow an overall impression to be obtained of the possible load levels of coal dust caused by handling. Some recommendations are made on the basis of the results for further research.

TRACER AEROSOL GENERATOR: FEASIBILITY STUDY PART ONE INFLUENCE OF THE HETERODISPERSITY OF THE TRACER AEROSOL AND ESTIMATION OF THE REQUIRED SOURCE STRENGTH OF THE GENERATOR

Author : F. Oeseburg
Report no. : PML 1984-C 7
File no. : X 3109
Date : August 1983
Language : Dutch

SUMMARY

The effect of the heterodispersity of the tracer aerosol on the results to be expected within the framework of the OWS-2.1-project (sampling efficiency of instruments for the measurement of particulate matter) and the project "Validation of models for the atmospheric dispersion of coal dust, using tracer aerosols (TVS-project)" (still to be programmed) was determined by means of a computer simulation. The required source strength of the aerosol generator was also estimated by using a simulation.

During the simulation process two particle size distributions were handled to describe both the regular particles and the satellites. These distributions were represented by "zero-order-logarithmic-distributions". The transport of the tracer aerosol from the source to the measuring instruments was described by means of a modified Gaussian plume model, in which the deposition velocity of the particles was taken into account. Provisional sampling efficiencies were used during the calculation of the masses collected by the instruments. These were obtained from literature. Three instruments (high volume sampler (HVS), Sierra and sampling tube), forming a cross-section of the equipment used, were considered within the project OWS-2.1. The rotorod was chosen as a reference sampling instrument in both projects. The detection limit of the method of (chemical) analysis, the blanks of the filter of the instruments and the background concentration of the tracer aerosol were regarded as limiting factors during the calculation of the minimum source strength of the aersosol generator. Bouncing of particles as a

result of too high a loading of the rotorod was taken into account in the calculations of the maximum source strength. For this purpose a simple mechanistic model simulating the consequences of bouncing was developed. The required source strength was determined on the basis of the minimal and maximal values.

SAMPLING TECHNIQUES OF FUGITIVE COAL DUST

Author : W.M. ter Kuile
Report no. : F 2117
Date : September 1983
Language : Dutch

SUMMARY

A review and evaluation of the principal sampling techniques for concentrations measurements of fugitive coal dust is given. The sampling instruments under evaluation in the "fugitive dust"-program, cover all groups of dust sampling techniques suitable for either determination of dispersion of (fugitive) coal dust in a short distance of the source or health related sampling (see ISO-TR7708-1983) of particle size fractions of the coal dust. The influence of atmospheric turbulence at prevailing wind velocities on the sampling efficiency will be determined as a part of the BEOP-project on evaluation of dust sampling instruments for outdoor measurement of fugitive coal dust. The possible importance of additional windtunnelresearch will be considered there. Since the development of new dust samplers is running this review is limited to the most relevant dust samplers at the moment.

Though it is expected that high windspeeds (hard to stormy wind) will cause most annoyance by fugitive coal dust, probably representative dust concentration measurements are not possible under these circumstances with commercial available dust sampling instruments.

Based on frequency distribution of the windspeed at places near the border of the Netherlands it is recommended to test the sampling efficiency of fugitive coal dust samplers for windspeed up to 12 m s^{-1} at 1.5 m above ground level. However, for wind velocities greater than 10 m s^{-1} reference instruments for sampling coarse dust will have to be developed. The present study gives no reason to consider serious changes in the running projects of the "fugitive coal dust" program.

TRACER AEROSOL GENERATOR: feasibility study part 2, final report

Author : F. Oeseburg
Report no. : PML 1984-C-38
Administr. no. : X 3109
Date : October 1983
Language : Dutch

SUMMARY

A selection of commercial available monodisperse aerosol generators with source strenghts comparable with the required source strengths mentioned in Reference 4 was made (projects: Sampling efficiency of instruments for the measurement of particulate matter (OWS-2.1); Validation of models for the atmospheric dispersion of coal dust using tracer aerosols (TVS)). Suitable tracers were selected taking into account the critical relative humidity and the solubility of the compound, the concentration of the background aerosol, the blank of the filter material, the detection limit of the method of analysis (AAS) and the toxicity (TWA-value). The feasibility of the technical concept, including the generation of a primary spray consisting of a solution of the tracer in water and the subsequent evaporation of the spray, was studied for both projects (OWS-2.1; TVS) by means of a theoretical analysis. An evaporation model was developed for this purpose. Moreover an estimate of the required power was made and this estimate was compared with a previously fixed limit. A concise description of the methods of analysis of the tracer aerosol (mass concentration; particle size) is given.

SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART CONCERNING CONCENTRATION AND DEPOSITION LEVELS MEASURED IN THE VICINITY OF COAL HANDLING AND STORAGE FACILITIES

Author : R.W. Lanting
Report no. : G 1280
Date : January 1984
Language : Dutch

SUMMARY

Data on concentration and deposition levels measured in the vicinity of coal handling and storage facilities as well as on the physical and chemical characteristics of fugitive coal dust were collected from available reports and publications.

Fugitive coal dust consists mainly of relatively coarse particles. From the theoretical considerations supported by wind tunnel experiments it is concluded that under dry conditions coal particles with diameters up to 260 µm can become air-borne. As the moisture content of the coal rises, the fraction of coal susceptable to wind erosion decreases. For some coals this fraction can range from 1 to 10%. For dry powdered coals the mass fraction of particles smaller than 260 µm is higher (60-90%). At several tens of metres from the source the mass median diameter of fugitive coal dust is roughly 50-70 µm. The fraction of coal particles < 10 µm in the total mass of wind-blown coal dust is very small (<1%).

Owing to the rapid fall-out of hypermicron particles the concentration levels of coal dust decrease rapidly with the distance (a factor of 6 within 100 metres).
There is a large contribution of wind-blown soil dust to the concentrations of total dust (all particle sizes). Wind erosion of stabilized coal piles is only small compared to other coal handling activities.
Most studies presenting measuring results lack in providing clear-out data on the type of activities on the coal handling facility as well as on the physical characteristics of the coal handled. Hence generalization

of the concentration and deposition levels measured is hardly possible. At very short distance (0-5 metres) from the source peak concentration levels of total dust amounting to several tens of milligrams per m^3 were measured in dust plumes. The dust contained 60-80% coal. At distances ranging from 5 to 100 metres concentration levels measured vary to a large extent, which is mainly caused by differences in climatological conditions, the type of coal handled, the type of activities and the geometry of the stock pile. The highest values of total dust measured are in the order of magnitude of several milligrams per m^3. On the average the dust contained 30% coal. At distances of 300-500 metres from coal handling facilities the emissions of coal dust contribute only for 10% to the concentration levels of total suspended particulate matter. At these distance violations of the EPA air quality standard for total suspended particulate matter are incidental and mainly caused by fugitive dust from unpaved roads.

Deposition levels measured all indicate that deposition fluxes decrease rapidly with increasing distances from the source.
The coal dust content at distance of 100-300 metres varies between 6 and 50% and is further reduced to approximately 10% at distances of 600-1000 metres.
The largest particles occasionally found in deposition samples taken at a distance of 50 metres from a source had particle diameters ranging from 450 to 550 µm.
Resuspension of once deposited particles might play a role, however, its contribution to deposition fluxes is unknown.
Wet depositions is of insignificant importance. During rainfall the source emissions are reduced considerably. Besides, the mass fraction of small coal particle which remain in susênsion for a long time and are transported over long distances is only small.
Violations of air quality standard for deposition fluxes occur occasionally in industrial environments at distances up tp 750 metres from the source. However, under these circumstances the contribution of coal dust is only small (< 10%).
Besides noise, soiling is the most important nuisance aspect from coal storage and handling facilities. Complaints about soiling occur when

deposition fluxes of black dust exceed values of 1 to 2 $g.m^{-2}.month^{-1}$. Under well-controlled handling operations these values usually are not exceeded at distances of 150 to 500 metres from the source. One case is reported where during handling of cokes exceedance occured at a distance of 1000 metres.

A SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART OF SAMPLING AND ANALYSIS OF FUGITIVE COAL DUST

Author : J. den Boeft
Report no. : G 1316
Project no. : 11941
Date : May 1984
Language : Dutch

SUMMARY

1. SAMPLING INSTRUMENTS

In the literature studied a large number of samples and direct reading instruments which have been used for concentration and deposition measurements of coal dust were found.
Despite their shortcomings in sampling efficiency, traditional samplers like EPA Hivol and dust fall buckets are still used on a large scale.
Depending on the measurement objectives a selection of instruments is made. Some instruments are not yet validated.

1.1 ASSESSMENT OF EMISSIONFACTORS

Point and line sources (handling and haulrouds)

- Isokinetic samplers (MRI, PEDCo, MEDUSA, Filter packs).
- Isokinetic tunnel and possibly also the fugitive dust sampler (TSI).
- Rotating impactors ((modified) Rotorod, rotating coarse dust sampler).
- Forward scattering cloud droplet spectrometer.

For the assessment of emission factors there is a need for a single instrument which seperates the total particles size distribution (0 - 200 µm) on the basis of aerodynamic diameter in seperate size fractions.

Area sources (wind erosion and handling)

It is presumed that emissions due to wind erosion occur at windspeeds

over 8 $m.s^{-1}$. The selection of samplers should be compatible with this limiting factor.

- Rotating impactors, like the modified Rotorod (high tip spees) for coarse particles, and the rotating arm collector for coarse and fine dust. The last mentioned instrument also collects fine particles on a filter.

1.2 DISPERSION AND DEPOSOTION

Concentration measurements

Preferably concentration measurements are made with omnidirectional samplers heads.

- Rotating impactors (Rotorod and modified Rotorod) for the coarse fraction.
- Dichotomous samplers with windspeed independent sampling heads (Wedding inlet) for the fine fraction (d_{ae} < 10 μm).

Deposition measurements

- Sticky plates with a large effective area.

1.3 EFFECT RELATED MEASUREMENTS

Health

. Total suspendend particulate matter
  - EPA Hivol or LIS/P-LIB when air quality standards have to be met.

. Thoracic fraction
  - Hivol with size selective inlet
  - Dichotomous sampler with size selective inlet.

. Respirable dust
  - Samplers for respirable dust such as
    - Piezobalance
    - Simslin
    - β dust monitor with preseparator

  which have been developed for occupational hygiene applications (mines).

For quantitative measurements in the ambient air these instruments are less well suited. However, for relative measurements such as event detection during exposure profiling these instruments can be used.

Soiling

- Sticky plates with a large effective area.

There is a lack of sampling and analytical techniques which are representative for soiling nuisance.

1.4 EVALUATION OF SAMPLERS

In combustion with reference methods field experiments may give important information on the sampling efficiency of dust samplers under real conditions. Besides the meteorological variables influencing the sampling efficiency can be identified. If these relevant meteorological variables can be simulated in a windtunnel, this type of evaluation has the following advantage:

- evaluation at higher windspeeds;
- samplers are tested under reproducible conditions.

Up till now windtunnel studies on sampler performance have not been made in the Netherlands.

2. COAL ANALYSIS

In literature no analytical procedure was found which is specific for coal. Gravimetric bulk analysis is most frequently used for the total sample as well as for aerodynamically fractionated samples.

Light microscopy is a usefull tool for identification and particle size measurements. This method is time consuming. Automatic image analysis highly enhances the applicability of light microscopy. Like light microscopy electron microscopy in particular scanning electron microscopy is a suitable technique.

Elemental analysis of individual particles can be performed by scanning electron microscopy fitted with an energy dispersive X-ray analysis (EDAX) system. However, as EDAX only detects elements above atomic number 11 (sodium) coal cannot be identified positively. A recent development is computer controlled scanning electron microscopy (CCSEM).

SEM, EDAX and image analysis are computer controlled. Particles can be detected, sized and chemically characterized automatically.
For source-oriented measurements total carbon analysis has been used as a proxy for the coal content of a dust sample.
Analytically also a separation can be made between organic and inorganic carbon. The separation technique (thermal or chemical) used determines which fraction is organic or elementary carbon. For coal dust an aerodynamic separation during sampling can be made. The coarse dust fraction $d_{ae} > 2.5$ µm contains predominantely coal dust while the fine dust fraction consists mainly of organic carbon and soot.

3. RECOMMENDATIONS

- Evaluation of isokinetic samplers based on the Hivol principle.
- Development and evaluation of a particle size fractionating sampler for total dust.
- For testing the performance of samplers under reproducible conditions during high windspeeds a windtunnel is required.
- Development and evaluation of a deposition measurement technique which is relevant for soiling nuisance.

A SURVEY AND EVALUATION OF THE INTERNATIONALLY AVAILABLE KNOWLEDGE RELATING TO THE EMISSIONS AND EMISSION FACTORS OF FUGITIVE COAL DUST DURING COAL HANDLING AND STORAGE

Author : G.Th. Visser
Reference no. : 84-01340
File no. : 8724-10384
Date : May 1984
Language : Dutch

SUMMARY

In this study a survey and evaluation is given of internationally available knowledge relating to the emissions of wind-dispersed coal dust during coal handling and storage and the possible applicability to the situation prevailing in the Netherlands of the emission factors derived from literature hereon.
Both field measurements to determine total emissions and emissions for each particular activity or emission source and windtunnel and laboratory measurements are dealt with.
A short survey is also given of the value and reliability of emission control efficiencies known from literature for the various emission sources as the result of any emission-reducing measure.
It can be concluded from this study that there are hardly any reliable emission factors for coal handling and storage that are applicable to Dutch conditions, either for total emissions or with reference to the various individual emission sources.
Thus, dependent on the various quantities of influence determing the emission and the local situation and management, values for total emission are found in the literature ranging from 1 to 5000 $\mu g/m^2 s$. For conditions obtaining in the Netherlands with damp coal the emission level will probably be in the lower half of this range.
It appears that in case studies always emissions for each emission source are determined. Never the emission calculation is based upon emission factors for total emissions.
The spread in emission factor values for the different emission sources is generally large, the documentation of the measurements often in-

sufficient, and in many cases essential data are lacking that are required to appreciate the emission factors and, if necessary, extrapolate them to different situations.
The results of windtunnel and laboratory measurements known so far mainly give qualitative information on the various relationships between emissions and quantities of influence, and the spread in the few known quantitative measuring data is considerable.
The control efficiencies known from literature are mainly based on subjective estimates and are therefore highly unreliable. A relatively small number of control efficiencies is based on measurements with coal.

A SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART REGARDING DISPERSION AND DISPERSION MODELS OF FUGITIVE COAL DUST

Author : P.E.J. Vermeulen
Reference no. : 84-06399
File no. : 8724-10384
Date : May 1984
Language : Dutch

SUMMARY

This study deals with the aspects playing a part in the dispersion of coal dust blown up by the wind during the storage and handling of coal. It is concluded that the influence of atmospheric turbulence on the dispersion of fine coal particles is similar to the influence on gaseous contaminants, provided the particles are smaller than, say, 100 µm. Moreover, under the influence of gravity the particles fall to the ground at a velocity which, on an average, agrees with the velocity in free fall. Due to such factors as impaction and filtration the particles are held to the ground. The overall transport to the ground is defined by means of the deposition velocity.
Deposition velocity and velocity in free fall are not equal to one another in general. Mathematical dispersion models should therefore meet the requirement that the influences of gravity and dry deposition are individually modelled, with free fall velocity and deposition velocity as parameters.
For particles in excess of 100 µm the influence of atmospheric turbulence is smaller for the larger particles.
Coal dust blown up by the wind at storage and transhipment sites originates from any sources, with low emissions height in most cases. Dispersion of the dust mainly takes place in currents that are influenced by obstacles. It is therefore concluded that mathematical dispersion models should be capable of accounting for the influence of obstacles on dispersion.
It is found that the emission conditions (source strength, emission height) which are required as input parameters for mathematical models

can only be tentatively estimated.
It follows from an inventory of dispersion models that variants of the Gaussian plume model are available which are expected to be applicable to calculations of the dispersion of dust particles smaller than 100 μm.
Because of partical considerations also preference is given to a socalled partial-reflection model and to methods which have been derived from solutions of the dispersion equation for dust particles.

SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART CONCERNING DEPOSITION AND RESUSPENSION MODELS OF FUGITIVE COAL DUST

Author : P.E.J. Vermeulen
Reference no. : 84-03759
File no. : 8724-10384
Date : June 1984
Language : Dutch

SUMMARY

The dispersion of coal-dust particles can only be calculated if the dry deposition velocity is known.
This velocity indicates to what extent dust particles reaching the ground are actually "retained".
On the basis of a survey of a number of methods for the determination of the dry deposition velocity, Sehmel and Hodgson's method is recommended for use. This method is suitable for the determination of the dry deposition velocity above relatively smooth terrain such as grassland for example.
The dry deposition velocity above rough terrain such as high-growing crops or wooded country for example, cannot be ascertained accurately, and further research on this subject is recommended therefore.
Coal dust particles can also be removed from a dust plume by the action of raindrops (wet deposition). It is concluded that for calculation of the wet deposition of particles in excess of 1 μm reliable methods are available.
Deposited coal dust particles can be dispersed by the wind at an other point of time, which is frequently denoted by re-suspension. For the re-suspension emission of dust particles from grassland an empirical correlation has been drawn up. It has been found that the re-suspension emission from sites situated around a coal terminal is insignificant under normal conditions. Incidentally, and in particular with strong winds, this emission can be much higher, however. Thus, in carrying out measuring campaigns on emission factors for example, due account should be taken of re-suspension emissions, in particular with strong winds.

A SURVEY AND EVALUATION OF THE INTERNATIONAL SITUATION CONCERNING THE INFLUENCE OF FUGITIVE DUST FROM COAL STORAGE AND HANDLING ON THE HEALTH OF PERSONS NOT OCCUPATIONALLY EXPOSED TO COAL DUST

Author : H.C.M. Mulder
Report no. : D 90
Project no. : 4.351
Date : June 1984
Language : Dutch

SUMMARY

In the literature studies in the framework of this review and evaluation no research was found describing health effects on persons who are non-occupationally exposed to fugitive coal dust.
From limited data, concentrations of fugitive coal dust occuring near stock piles have been compared with existing standards for suspended particles in general.
As the occuring concentrations at distances of a few hundred meters from the source do not or hardly exceed the background concentrations of suspended particles no health effects are to be expected at this distance.
At very short distances from the source (< 200 m) existing standards may be exceeded, but also here no health effects are to be expected.
The many times described diseases among mine-workers, like silicosis and chronic bronchitis evolved after exposures to coal dust that are not comparable to exposures near stock-piles of coal.

A SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART CONCERNING THE INFLUENCE OF FUGITIVE COAL DUST ON THE BIOTIC AND ABIOTIC SYSTEMS

Authors : R.W. Lanting
H. Harssema (Agricultural University, Wageningen)
Report no. : G 1314
Project no. : 11941
Date : June 1984
Language : Dutch

SUMMARY

The following properties of fugitive coal dust were identified to be of importance for the effects on biotic and abiotic systems:

- high deposition velocity (small mass fraction of particles $d_{ae} < 2,5$ µm), strong variations in source strength both in time and space;
- low light reflection;
- chemical properties (content of sulphur, chloride and toxic metals);
- adhesive properties on smooth surfaces.

Noted effects occur on a local scale. No large-scale effects are expected. Soiling of objects was found to be the most important effect. Soiling complaints occur incidentally and are always closely related to emission events.
Using indicative dose-annoyance relations for soiling collected from literature, a preliminary mathematic description of the whole sequence from emission to effect seems to be feasible.
For the validation of such a model techniques for the measurements of soiling in relation to deposition (e.g. lightreflection of receptorsurfaces) are required.
Coal dust may play a role in the chemical deterioration (pit corrosion) of aluminium.
For all other physical actions of coal dust on materials the properties of coal dust are not significantly different from other particulate air pollutants. Preventive measures already taken are also effective for coal dust. There are no large-scale effects of fugitive coal dust on

visibility.

The effects on aquatic systems are of minor importance.
On theoretical considerations the leaching of toxic elements from deposited coal dust seems to contribute only slightly to the pollution of soils. Concerning the biotic environment effects on vegetation (e.g. decrease in yield of agricultural crops in greenhouses) may be expected at short distances from the source. Dose-effect relations are still lacking. The following gaps in knowledge are identified:

- The cleaning action of rain on adhering coal dust.
- Pollution of soils by toxic elements during long-term deposition of coal dust.
- The effects of decreased lighttransmission due to coal dust deposition on the cropyield in greenhouses and the efficiency of solar radiation collectors.

A SURVEY OF THE AVAILABLE LITERATURE REGARDING FUGITIVE DUST DURING HANDLING AND STORAGE OF COAL

Authors : P.E.J. Vermeulen
G.Th. Visser

Reference no. : 84-0457
File no. : 8724-10384
Date : June 1984
Language : Dutch

SUMMARY

This report lists the available literature regarding fugitive dust during the storage and handling of coal.
This review is made using a literature data base, specifically developed for the OWS-1 study.
It is concluded that most of the available references deal with the topics "emissions", "concentration- and deposition measurements", "nuisance/ perception of coal dust" and "abatement techniques for fugitive coal dust".

DEVELOPMENT OF A TRACER AEROSOLGENERATOR PART ONE: PATICLE SIZE DISTRIBUTIONS OF SPRAYS GENERATED BY THE MICRO-ULVA

Author : F. Oeseburg
Reference no. : PML-1984-C83
Date : July, 1984
Language : Dutch

SUMMARY

An aerosol generator for the generation of monodisperse solid aerosols is being constructed within the framework of the program "Fugitive Dust". This device will be used in experiments to determine the sampling efficiency of a number of samplers for solid aerosols. Sprays consisting of a solution of the solid tracer in water are generated by means of a spinning disk (Micro-Ulva) which is an important component of the tracer aerosol generator. The solid aerosol is created after evaporation of the water.

Particle size distribution of water sprays generated by means of the Micro-Ulva were measured. Sampling was carried out with the aid of impaction rods which were drwan manually through the spray (impaction velocity circa 6 to 10 $m.s^{-1}$). The rods were provided with a layer of magnesium oxide in order to achieve fixation during sampling. The analysis was carried out using a microscope. The "true" diameters of the particles were calculated using the spreading factor (0.85) reported by May. Volume particle size distributions were calculated starting from the measured number size distribution. Relevant parameters (mode, relative standard deviation) of the distributions of both the regular particles and the satellites were estimated. The relative standard deviation of the distributions of the regular particles were compared with the required value following from the feasibility study. It was concluded that the Micro-Ulva in an appropriate atomizer for the tracer aerosol generator.

SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART OF CONTROL METHODS APPLIED TO FUGITIVE COAL DUST

Author : P.G. Eggels
Reference no. : 84-02601
File no. : 8722-10384
Date : July 1984
Language : Dutch

SUMMARY

According to the number of reports and articles published on this subject, a considerable amount of thought has been given internationally to the abatement of fugitive coal-dust along the route taken by the coal from the time it is mined till the time of its receipt by the user. There is a lack of fundamental reasearch, however.

The control methods applied at one location often comprise a number of measures attuned to one another. A general characteristic of such a situation is that one or more spraying operations are resorted and very often some kind of enclosure is applied to the processing installation.

There are various sources of dust-emission at coal storage and shipment locations, consequently the measures taken at these locations differ in nature and intensity for each of the sources.

In the matter of differences in the abatement methods applied to comparable emission sources, it can be commented here that such differences can often be attributed to differences in the extents of the areas occupied for coal storage and/or transshipment, the type of coal involved, the moisture-content of the coal and the influence exerted by the prevailing climatic conditions.
In a number of cases, such differences in the control methods must be based on the (environmental) technical or economic assessment of the problem.
With the state of the technique as it stands at the present, it is of the general opinion that practically complete control of the fugitive

coal dust problem can be achieved. At the present time however total elimination of fugitive coal dust has hardly been achieved, this may be due to the financial consequences associated with the task.

There is a partial lack of reliable information in the relative literature concerning the effectiveness of the control methods and which is necessary in order to make an evaluation of the fugitive coal dust control. Some cost figures for an economic evaluation are not directly available either.

A SURVEY AND EVALUATION OF THE INTERNATIONAL STATE OF THE ART CONCERNING ANNOYANCE IN RESIDENTIAL AREAS FROM COAL STORAGE AND HANDLING

Author : J.E.F. van Dongen
Report no. : D 82
Date : July 1984
Language : Dutch

SUMMARY

In the literature studied in the framework of this inventarisation no investigations have been found which give more than a rough insight into the problem of annoyance from coal dust in residential areas near coal stockyards and transhipment locations. A dose-(annoyance)response relation with respect to coal dust cannot be developed yet. At best it can be concluded that at a same deposition black dust from coal, soot or fly ash sooner cause annoyance than from other kinds of dust. However, some dose-response relations of other kinds of dust are available.
Although from concentration and deposition measurements it appears that the dispersion of coal dust is limited to 1500 m, a conclusion that for those reasons problems of annoyance also will be limited is premature.
Both on the dosis side and the effect side intervening factors play a role, which insufficiently have been analysed with respect to the annoyance. This concerns items as the frequency of special weather conditions, the use of transport routes, emissions from other dust sources and the occurrence of other environmental effects like noise.
There are indications that annoyance sooner appears in new than in established situations.

TNO

SURVEY AND EVALUATION OF THE GLOBAL STATE OF THE ART REGARDING FUGITIVE COAL DUST: EMITTED AT COAL HANDLING AND COAL STORAGE FACILITIES

Author : B. Stork
Report no. : 84-013008
File no. : 8724-10384
Date : October 1984
Language : Dutch

SUMMARY

The Dutch organization for applied scientific research TNO has carried out within the framework of the National Coal Reseach Programme (NOK) a review and evaluation of the international state of the art of all aspects of fugitive coal dust occurring at the coal storage and handling. This report gives extensive results and conclusions extracted from 15 subreports which have been issued up to now.
The main conclusions are:

- the problem is local and is restricted to distances no further than 1,5 km from the source;
- annoyance by contamination is the problem, health effects are not likely to occur.

The report contains recommendations for emission factors, control efficiencies, dispersion and deposition models to be used but states at the same time that much further research is required in order to obtain reliable figures/models/final conclusions.
Recommendations for further work are presented.

SURVEY AND EVALUATION OF THE GLOBAL STATE OF THE ART REGARDING FUGITIVE COAL DUST, EMITTED AT COAL HANDLING AND COAL STORAGE FACILITIES

Author : B. Stork
Reference no. : 84-013019
File no. : 8724-12564
Date : November 1984
Language : English

SUMMARY

The Dutch Minister of Economic Affairs has established a National Coal Research Program (NOK) aiming at the elemination of technical, economic and ecological barriers to the re-introduction of coal as a main source of energy. Industries, engineering companies, (technical) universities and other R & D organisations, such as the Netherlands Organisation for Applied Scientific Research (TNO), are carrying out research projects within the framework of this program.
Part of NOK is research into the ecological aspects of fugitive dust caused by the storage and handling of coal. One of the projects to be carried out by TNO is a review and evaluation to the international state of the art of all aspects of fugitive coal dust.
The purpose of this project is to provide scientific support to the other research projects in NOK concerning fugitive dust.
Such a review comprises the following items:

- sampling/registration techniques
- emission
- dispersion models
- concentrations
- deposition
- resuspension
- abatement techniques
- annoyance
- effects on human beings, nature, agriculture, horticulture and materials
- R & D programmes carried out in other countries.

Up to 15 reports have been issued on the above mentioned items. Both these subreports and the final report of NOK-OWS-1 are written in Dutch. In this report an extended summary in the English language is therefore provided for those, interested in the contents of the various reports. Also the conclusions of the final report are presented.

TNO

RISKS CONNECTED WITH THE MINING, TRANSPORT AND USE OF COAL USED FOR ELECTRICITY PRODUCTION IN THE NETHERLANDS

Author : G. Snellink
Reference No. : 78-04884/MSW
File No. : 8713-3537
Date : March 1979
Language : Dutch

SUMMARY

A desk study has been made into risk-analysis literature concerning coal in its relation to the Dutch situation. The specific coal studies are only partly applicable to the Netherlands. To find additional data use has been made of other sources of information (data from the Netherlands Bureau of Statistics and the Netherlands Maritime Institute for example). The results found for the Dutch-related situation (up to 1985) show a statistic value of circa 1 to 1.5 seriously wounded persons per 10^6 tons of coal imported into the Netherlands. From these, 0.6 to 1.3 are persons engaged in direct labour (mining and transport), whereas 0.3 to 0.6 are persons belonging to indirect labour sources (traffic accidents). For the Netherlands part these figures relate to 0.07 persons involved in direct labour and 0.12 in indirect labour. The other fatal accidents occur for the greater part in countries where the coal is mined. The data so calculated are about 20% higher than those quoted in U.S.-studies, as a result of the extra transport over sea and in the Netherlands. This study is limited to the use of coal for electricity generation in power plants with currently used equipment.

A SIMPLE MODEL FOR THE SELF HEATING OF COAL

Authors : J.W. van Heuven
D. Schmal
J.H. Duyzer

Report No. : CL 82/32
Order No. : 50009
Contract No. : 4.351-2.1
Date : March 29th, 1982
Language : Dutch

SUMMARY

As part of an investigation into the self-heating of coal piles, we have developed a one-dimensional model describing the self-heating at relatively low temperatures (less than 80-90°C). The model aims to establish a relation between laboratory measurements on coal and the behaviour of coal piles. In addition, it should be useful in establishing those physical and chemical factors that play an important part in the heating of coal piles. We have for the time being opted for a simple one-dimensional model because it is yet unknown which of several processes contribute to the complex phenomenon of self-heating.
The (non-stationary) model takes account of the depletion of oxygen and development of heat by chemisorption of oxygen to coal inside a coal pile, of transport of oxygen through diffusion and convection, and of heat through conduction and convection. It consists of three differential equations, namely an oxygen mass balance, a heat balance, and a reaction rate equation. For a number of simple cases, the equations can be solved analytically. For non-stationary conditions, they can be solved numerically. Calculations (by both methods of solution) on the basis of practical data yield results describing the self-heating of coal piles in a qualitative sense. If the model is to yield results that are useful in a quantitative sense, more precise data will be needed of the factors operating in the process of self-heating. These factors include, first, the porosity of the coal pile and its ventilation rate, which strongly depends on porosity, and secondly, the reactivity of the coal and those factors on which it depends.

In its present form the model affords a satisfactory qualitative description of the behaviour of a self-heating coal pile. We therefore hope that it can serve as a basis for more elaborate models describing that behaviour quantitatively.

TNO

ROLE OF MICROORGANISMS IN THE DEVELOPMENT OF SPONTANEOUS HEATING

Author : H.J. Doddema
Report no. : MT 82/489/4301
Date : December 31, 1982
Language : Dutch

SUMMARY

In the framework of the research on the insidious tendency of some coals to self-heating while in storage, we tried to count the number of microorganisms on coal in coal storage-piles. Our aim was to achieve an impression of the oxidative capacity of microorganisms in coal and of the resulting heat production by microbial activity as a contribution to the initial self-heating of coal.
Coal from hot (60^{o} - 80^{o}C) areas in coal storage heaps was dried, milled and sieved. Consequently the number of microorganisms were counted using the Most Probable Number Technique.
In all coals tested the numbers of microorganisms were so low ($n<100$ per g) that it is unlikely that the microbial oxidation of reactive groups in coal will contribute significantly to the self heating of coal.

TNO

HEAT PRODUCTION DURING COAL STORAGE

Author : A.H. Heemskerk
Report No. : PML 1982-112
Date : March 1982
Language : Dutch

SUMMARY

The results of the investigation into the heat generation of coal, which is caused by degradation, are reported.
Temperature as well as moisture content play an important role during the self-heating process. This role cannot be explained by physical adsorption processes, but it is hoped that chemisorption processes can. During degradation chiefly carbon dioxide is formed. Even under oxygen-poor circumstances the production of carbon monoxide is small.

INVESTIGATION INTO SOME METHODS FOR THE DETERMINATION OF THE TENDENCY OF SELF HEATING

Author : R. Tadema Wielandt
Report no. : R 83/10
Order no. : 50087
Date : January 1983
Language : Dutch

SUMMARY

While carrying out work on the project "Calorific loss and spontaneous heating in coal-piles" there appeared to be interest into studying the heating behavior of various types of coal.
The purpose of the orientating research was to find a method by which the tendency of several sorts of coal towards spontaneous heating can be compared.
As the reactivity in oxidation reactions could be seen as a measure for the tendency towards spontaneous heating, a reactor was developed, wherein coal samples could be quickly oxidized with atomic oxygen. It was thought that the reaction could be followed by monitoring the chemoluminescense emitted during oxidation.
By measuring the light reflection of the coal surface, it was found that the chemoluminescence process occurred once the surface of coal was almost completely ashed. The developed chemoluminescence technique was, therefore, found to be both too unsensitive and too unspecific to be used as an oxidation monitor.
From experiments carried out using Fourier-transform-infrared-spectroscopy (FTIR) it was found that this technique appeared to be more suitable to specifically follow the fast oxidation process.

A MODEL FOR THE SPONTANEOUS HEATING OF COAL

Authors : D. Schmal
J.H. Duyzer
J.W. van Heuven

Report no. : R 84/16a
Order no. : 10565
Date : January 25, 1984
Language : English

SUMMARY

As part of an investigation into the spontaneous heating of coal piles, we have developed a one-dimensional model describing the spontaneous heating process at relatively low temperatures (less than about 100°C). The model aims at establishing a relation between laboratory measurements on coal and the behaviour of coal piles. In addition the model should be useful in establishing those physical and chemical factors that play an important role in the heating of coal piles in order to take rational measures against excessive losses in caloric value. For the time being we have opted for a one-dimensional model.
The ultimate non-steady model described in this report takes into account the depletion of oxygen and the production of heat by chemisorption of oxygen in the coal, the transport of oxygen via diffusion and convection and the transport of heat via conduction, convection and evaporation/condensation of coal moisture. It consists of four differential equations, namely equations for conservation of oxygen mass, of coal moisture and of heat and a reaction-rate equation of oxygen with coal.
For some simple cases, the equations were solved analytically. For non-steady conditions, they were solved numerically.

Calculations using data from laboratory and field experiments give results that describe the process of spontaneous heating in a semi-quantitative sense as it appeared from a comparison between the results of calculations and those of measurements in coal piles.

TNO

HEAT GENERATION DURING COAL STORAGE

Author : A.H. Heemskerk
Report no. : PML 1983-107
Date : January 12, 1983
Language : Dutch

SUMMARY

The results of the investigations in 1982 into the heat generation of coal as a consequence of degradation, are reported.
The heat generations of two types of Australian and American coal have been determined isothermally at several temperature levels. A direct relationship between the oxygen concentration and the heat generation has been established for oxygen concentrations between 10 and 100%. Only a marginal effect of the particle size on the heat generation has been found. The moisture balance in the coal plays a vital role in the self-heating process. According to adiabatic experiments the cooling effect of evaporating moisture is closely related to the varying ventilation rate, while also the relative humidity of the air is involved in the process.

CALORIFIC LOSSES AND SELF-HEATING OF STORED COAL

Reference No. : 83-0323
File No. : 8722-50009
Date : February 1983
Language : Dutch

SUMMARY

The project "Calorific losses and self-heating phenomena in stored coal" is jointly executed by TNO and KEMA. Research in practice is carried out by KEMA, TNO undertakes investigations in the laboratory. The calorific losses are defined by the weight loss of pure coal and the decrease in the heating value. For the average weight loss of 15 batches of loose dumped coal the result was: about 5% in the first year and about 2% per year in the following years. About 2% decrease in heating value per year was found.

In the laboratory the heat production was measured under similar conditions as prevailing in coal piles. Self-heating sensitive coal was used. A heat production of 40-70 mW/kg was found for Australian coal and of 150 mW/kg for US coal in air at 65°C. The activation energy of the reaction between coal and oxygen was 70-80 kJ/mol. Above 10% oxygen the reaction rate was proportional to the oxygen concentration to the power of 0.7. Humidity and particle size gave deviations in heat production of less than 20% and their influence on heat production was therefore slight.

TNO

SELF-HEATING OF SUB-BITUMINOUS COAL-INVESTIGATION 1983

Author : A.H. Heemskerk
Report no. : PML 1984-C35
File no. : M 9189
Date : February 13, 1984
Language : English

SUMMARY

The results of the investigation into the heat generation of coal are collected in this report.
The relationship between the heat generation and the partial pressure of oxygen has been investigated for low oxygen concentrations (0-20% O_2). An unequivocal relationship could be established, which is in agreement with the observations at high oxygen concentrations (10-100% O_2).
The loss in caloric value and in the activity of the coal has been established with long-term storage experiments performed at 305 and 323 K. It appears that the addition of such chemicals as sulphuric acid and iron salts that are frequently encountered in coal piles stimulate the heat generation.
Adiabatic investigations reveal an equilibrium between the moisture in the gas phase and in the solid phase, leading to an adiabatic saturation temperature. This temperature is dependent on the flow rate and appears to remain between 348 and 363 K under normal storage conditions.
It is evident from all experiments that the entire process of self-heating is determined by the chemical oxidation process generating heat and the physical process of evaporation consuming heat.

TNO

SPONTANEOUS HEATING OF COAL

Authors : D. Schmal
A.H. Heemskerk
A. Kok
J.W. van Heuven

Report no. : R 84/47
Oder no. : 10565
Contract no. : 4.351-2.1
Date : March 22, 1984
Language : English

SUMMARY

In this report a review is given of the results and their evaluation of investigations on spontaneous heating of coal performed in 1983.
The research programme consists of three parts:

- field measurements;
- laboratory measurements;
- development of a model.

A lot of information on the process of spontaneous heating was gained in the course of the project. Methods were developed to estimate the tendency of spontaneous heating and losses in calorific values of coal piles on the basis of measurements on gas composition and its flow velocities in the pile. Mean steam coal losses calculated from measurements vary between 0.1 and 4% per year depending on storage conditions. Local losses in calorific value in piles having temperatures of about $80^{o}C$ can be as high as 15% in one year. The making of an inventory of these losses will be continued.
Furthermore a method was developed to predict the spontaneous heating behaviour of a coal pile on the basis of laboratory measurements and a methematical model. As follows from a comparison of the calculating results with those of field experiments, it now gives semi-quantitative results. For a further quantitative verification, it is necessary to perform experiments, under well-defined conditions, with coal piles in various degrees of compaction.

From this study it can be concluded that the most important parameters in the process of spontaneous heating are: the porosity of the pile (degree of compaction), the process of evaporation and condensation of coal moisture, temperature of the coal at the beginning of storage and reactivity of the coal towards oxygen adsorption.

With the knowledge obtained so far about the relative importance of the different parameters, it is already possible to give general recommendations about storage conditions and measures to prevent or delay spontaneous heating for existing and new coal piles. The verification mentioned above will give such recommendations a more solid basis.

A MODEL FOR THE SPONTANEOUS HEATING OF COAL

Authors : D. Schmal
J.H. Duyzer
J.W. van Heuven

Publication no.: P 84/14
Date : April 5, 1984
Language : English

SUMMARY

As part of an investigation into the spontaneous heating of coal piles, we have developed a one-dimensional model describing the spontaneous heating process at relatively low temperatures (less than about 100°C). The ultimate non-steady model takes into account the depletion of oxygen and the production of heat by chemisorption of oxygen in the coal, the transport of oxygen via diffusion and convection and the transport of heat via conduction, convection and evaporation/condensation of coal moisture. It consists of four differential equations, namely equations for conservation of oxygen mass, of coal moisture and of heat and a reaction-rate equation of oxygen with coal.
Calculations using data from laboratory and field experiments give results that describe the process of spontaneous heating in a semi-quantitative sense as it appeared from a comparison between the results of calculations and those of measurements in coal piles.
The most important parameters in the process of spontaneous heating particularly for the time between dumping and spontaneous ignition are the porosity of the pile (degree of compaction), the initial temperature of the coal and the evaporation and condensation of coal moisture. The influence of other parameters (e.g. reactivity of the coal, heat conductivity, etc.) is much less pronounced.

A PREDICTION METHOD FOR THE SPONTANEOUS HEATING OF COAL

Authors : D. Schmall
A.H. Heemskerk
J.W. van Heuven
J.H. Duyzer

Report no. : P 84/25
Presented at : 4th European Coal Utilization Conference,
4-6 September 1984,
Volume 1, 171-183
Order no. : 10729
Date : July 24, 1984
Language : English

SUMMARY

When coal is stored it suffers slow oxidation. This causes spontaneous heating, resulting in loss of calorific value and, possibly, in self-ignition. The reintroduction of coal as a major source of energy in the Netherlands and the important part played by Dutch harbours in the transhipment of coal have prompted a research programme into the various aspects of spontaneous heating. The programme comprises three parts, viz.:

- laboratory studies of the effect of a number of variables on heat generation by (mostly sub-bituminous) "steam" coal
- measurements in coal piles of temperature, gas composition, gas flow velocities, and loss of calorific value
- development of a mathematical model describing the process of spontaneous heating in a coal pile.

A mathematical model that reliably predicts the heating behaviour of a coal pile can be used to

- prompt measures to prevent (excessive) spontaneous heating
- calculate loss of calorific value
- reconstruct the course of events in a coal pile (e.g. for insurance purposes).

This paper presents a brief description of the model and the methods of the laboratory measurements. Some examples will highlight those variables which play an important part in spontaneous heating. It furthermore discusses those aspects of the calculations which are important from a practical viewpoint (recommendations for storing coal piles etc.) Results of field measurements illustrate the importance of the spontaneous heating process from an economic point of view: calorific losses in compacted steam coal vary between 0.1 and 4% per annum. Local losses in loosely stored coal were as high as 15% annually, even in the absence of ignition.

DUST EXPLOSIONS

Author : J.P. Zeeuwen
Published in : PT/Procestechniek, 36 (1981), 9, 445-451
Language : Dutch

SUMMARY

Finely divided dust of many products mixed with air can cause a dust explosion. In this publication a survey is given of this phenomenon, and also of the factors influencing the dust explosion. A review is given of the preventive and corrective measures.

REVIEW OF CURRENT RESEARCH AT TNO INTO GAS AND DUST EXPLOSIONS

Author : J.P. Zeeuwen

Presented at : The international specialist meeting on fuel air explosion.
November 4-6, 1981
Montreal PQ, Canada

Language : English

SUMMARY

In this paper a review is presented of the research into gas and dust explosions, which is currently in progress at the Prins Maurits Laboratory TNO. Since most research is contract research, it is not possible to go into details, but in order to highlight the areas of interest, a few examples of ongoing or recently finished research are given.
In the area of dust explosions emphasis is on recent work on the minimum ignition energy requirement of dust clouds.
In the area of gas explosions in confined systems attention is paid to a study of the coupling between acoustic waves and combustion in both completely closed and vented large explosion vessels.
Flame propagation in the presence of repeated obstacles relevant to gas cloud explosions, is illustrated for different degrees of confinement.
An example is given of numerical simulation of real unconfined gas explosion experiments where the measured flame history is used as input for blast calculation codes and the measured and simulated pressure pulses are compared.

TNO

AN ORIENTATING STUDY OF DUST EXPLOSION RISKS WHEN HANDLING AND PROCESSING COAL IN THE NETHERLANDS

Authors : G.F.M. van Laar
J.P. Zeeuwen
D.J.G. van Dijen
J.P.J. Ruygrok

Report No. : PML 1983-112
Administration No. : G 2180
Date : January 1983
Language : Dutch

SUMMARY

At the request of Project Office for Energy Research, BEOP, an orientating study has been conducted into the possible (dust) explosion hazards during transport, storage, transhipment, handling and use of coal in the Netherlands. The study comprises a survey of the explosion properties of coal dust, safety measures against explosions, and the explosion hazards in the different systems of transport, transhipment, handling and use of coal. The present and the future situation are described. This study concludes with guidelines and recommendations to promote explosion safety.

COAL EXPLOSION HAZARDS

Authors : J.P.J. Ruygrok
G.F.M. van Laar
J.P. Zeeuwen

Published in : Bulk Syst.Int., 5 (1983), 6, 51-53, 55
Language : English

SUMMARY

The increasing use of coal as fuel in the Netherlands has prompted research into its safe transport, handling and use. The Dutch government has sponsored an extensive research programme covering explosion hazards in these areas, which is described in this article. Coal gas and coal dust explosion phenomenology is described, and dust control procedures are included. Preventive measures are suggested.

TNO

COAL AND EXPLOSION DANGER

Authors : G.F.M. van Laar
J.P. Zeeuwen
D.J.G. van Dijen
J.P.J. Ruygrok

Published in : PT/Werktuigbouw, 38 (1983), 11, 71-74
38 (1983), 12, 51-53

Language : Dutch

SUMMARY

An investigation is carried out into the (dust)explosion risk caused by the use of coal today and in the future. The result of this study is an inventory of the explosion characteristics of coal dust, and the explosion risks in the various kinds of systems for transport and handling and processing.
Explosions are possible in all kinds of equipment and during all processing, but can be prevented by taking adequate measures.

INVESTIGATIONS INTO DUST EXPLOSIONS

Authors : G.F.M. van Laar
J.P. Zeeuwen

Presented at : The TNO Symposium "Industrial Safety" at Noordwijkerhout
Date : 15-16 December, 1983
Language : Dutch

SUMMARY

In this paper the phenomenon of dust explosion is explained. The prevention of dustexplosions is discussed as well as the measures that can be taken to reduce the effect of a dust explosion. A survey is given of the research into dust explosions at the Prins Maurits Laboratory since 1970.

A PRACTICAL LOOK AT DUST EXPLOSION CHARACTERISTICS

Authors : J.P. Zeeuwen
G.F.M. van Laar

Presented at : The symposium "Explosion Protection in Practice" at Antwerp
Date : April 16-19, 1984
Language : English

SUMMARY

The main dust explosion characteristics that have to be used in the design of preventive and protective measures are presented with a brief outline of the test method.
The US Bureau of Mines classification scheme ("explosion index") is described and reasons are given why its use is not recommended anymore. Dust explosion data from open literature sources are presented for samples of various branches of industry to illustrate the range of data that can be encountered in practice.

A SURVEY OF THE RESEARCH AT TNO IN THE DUST EXPLOSION FIELD OVER THE LAST DECADE

Authors : J.P. Zeeuwen
G.F.M. van Laar

Published in : VDI-Berichte, 494 (1984), 81-86
Language : English

SUMMARY

In this paper the following items are discussed:

- Influence of particle size and moisture content on such important explosion properties of dust-air mixtures as maximum explosion pressure, maximum rate of pressure rise (K_{st}) and maximum ignition energy,
- Influence of blending proportions on these properties,
- Explosion venting of a room partially filled with dust-air mixtures.

From the experiments performed the following conclusions can be drawn:

- The moisture content and particle size of a dust sample only have a large effect on the minimum ignition energy of that sample. The minimum ignition energy will drop appreciably with drier and smaller particles.
- When two products are mixed, the dust with the highest K_{st} and the lower ignition energy tends to influence these explosion properties of the mixture more than is to be expected on the basis of their weight ratios.
- An exploding dust cloud partially filling an enclosure and situated at a considerable distance from the vent, behaves like an exploding dust cloud of a much larger volume than the actual cloud. This means that in the case of a partially filled enclosure the surface area of the vent should be calculated on a multiple of the volume of the actual dust cloud.

EXPLOSION RISK AND COAL HANDLING

Authors : G.F.M. van Laar
J.P. Zeeuwen
Published in : PT/Procestechniek, 39 (1984), 6/7, 37-41
Language : Dutch

SUMMARY

A preliminary study has been made regarding the explosions risks when transporting, storing and processing coal. Especially the aspects of coal processing are discussed and the required conditions for dust explosions to occur. The characteristics of pulverised coal and fluid bed boilers and coal preparation plants in relation to dust explosion hazards are given.

THE RADIO-ACTIVE EMISSIONS AND THE IMPACT ON THE PEOPLE IN THE NEIGHBOURHOOD OF A 600 MWe COAL-FIRED POWER PLANT

Authors : L.A.M. Jansen
J.J. Broerse
Reference no. : 80-014233
File no. : 8710-7579
Date : February 1981
Language : Dutch

SUMMARY

The radiation impact from a coal-fired power plant (600 MWe) with a maximum fly-ash emission of 700.000 kg per annum on the people in the neighbourhood has been calculated.
The total radiation impact on the total human body was found to be 2 mrem per annum. This is primarily caused by the food. Radiation impact on the bones has been calculated to be 4 mrem per annum. These calculated values can be considered as maximum possible values caused by the emission of the fly-ash.
Additional impact of 2 mrem per annum can be considered as acceptable, compared with the impact of natural radiation of about 100 mrem per annum. The calculated impact is also small compared with the maximum allowable impact according to the ICRP limit (160 mrem per annum for the total human body), which are also valid in the Netherlands.
The fact is emphasized that lowering the fly-ash emission up to the level prescribed is a necessity. Application of the fly-ash requires special attention as a result of the radio activity of the fly-ash collected.

ARE PATIENTS SUFFERING FROM CHRONIC LUNG- AND BRONCHIAL TUBE DISEASES MORE IN TROUBLE WHEN COAL IS FIRED AGAIN IN THE NETHERLANDS?

Author : P.E. Joosting
Published in : Project (1982), 3, 91-96
Language : Dutch

SUMMARY

The construction and function of our lungs and bronchial tubes are such that air pollutants are eliminated in several ways. It depends on the quantity and kind of air pollution whether health will be negatively affected.
It has been demonstrated, when firing coal on a big scale, that people suffering from asthma and bronchitis, have more troubles with their disease. The author does not expect that the experiences obtained in England will be repeated in the Netherlands as a result of the improvement of coal-firing techniques.

ENVIRONMENTAL TOXICOLOGICAL RESEARCH OF EXTRACTS FROM TWO SAMPLES OF FLY ASH FROM A PULVERISED-COAL COMBUSTOR

Author : D.M.M. Adema
Report No. : R 83/59
Order No. : 10293
Date : April 5, 1983
Language : Dutch

SUMMARY

Aqueous and organic extracts from fly ash samples originating from the combustion of Polish coal in a pulverised-coal fired power plant (PGEM, Nijmegen) were investigated regarding some environmental toxicological properties. One sample was a mixture of the zones 1 to 4 inclusive of the electrostatic precipitator. The other sample was only from zone 4, where the smaller particles are collected. The toxicity of the aqueous extracts was determined by means of the crustacea Daphnia magna (mortality, growth and reproduction), the hydro-polyp Hydra oligactus (growth rate) and the plants Avena sativa and Brassica rapa (germination, growth and leaf damage). The toxicity of the organic extracts was determined by means of the Daphnia test.

LUNG MACROPHAGES, SOME GENERAL INFORMATION AND A LITERUTURE SURVEY OF CYTOTOXICITY TESTS WITH FLY ASH

Author : R.N. Hooftman
Report No. : R. 83/89
Order No. : 10293
Date : May 24, 1983
Language : Dutch

SUMMARY

Research has started into the possible biological hazardous effects to human beings and the environment of inhalable fly ash. As an introduction to this research this report gives general information on lung macrophages and the results of tests with fly ash as far as they are known from literature.

INVESTIGATION INTO THE PRIMARY SKIN AND EYE IRRITATION EFFECTS IN RABBITS AND FOR SENSITIZATIONS IN GUINEA PIGS, OF FLY ASH SAMPLES FROM A PULVERISED-COAL COMBUSTOR

Authors : G.J. Vink
L. van Beek
M.K. Prinsen
H.P. Til
D.C. Veldhuysen

Report no. : R 83/147a
Order no. : 10293
Date : August 10, 1983
Language : Dutch

SUMMARY

The effect of three samples of fly-ash from a pulverised coal combustor (PGEM, Nijmegen, The Netherlands), collected simultaneously during combustion of Polish coal, were investigated in single-application tests for primary skin and eye irritation in albino rabbits and for sensitization in guinea pigs. The fly-ash samples were collected at the combined final discharge of the four zones of the electrostatic precipitators (filter-ash), at the discharge from zone 4 alone (filter-ash E4) and in the stack with a bagfilter at 120°C (stack-ash DF). In this sequence of ash samples a decrease of particle size and an increase of concentrations of heavy metals and organics (e.g. PAH) can be expected.

Although small differences were found, the three samples all appeared to cause slight skin irritation.
Both samples of filter-ash caused slight eye irritation. The lesions cleared up completely, however, during a seven-day observation period. Stack-ash DF caused moderate eye irritation and the lesions only recovered partly within seven days. Because of this observation stack-ash DF is considered to be an eye irritant.
Washing of the treated eyes appeared to be an effective remedy against irritation. None of the ash samples gave rise to increased hypersensitivity (sensitization).

IN VITRO MEASUREMENT OF THE CYTOTOXICITY OF SOME FLY ASH SAMPLES FOR LUNG MACROPHAGES

Authors : R.N. Hooftman
C.W.M. Arkesteijn

Report no. : R 83/182a
Order no. : 10293
Date : September 10, 1983
Language : Dutch

SUMMARY

A number of fly ash samples were tested for cytotoxicity with bovine lungmacrophages. The macrophages were lavaged from a lung lobe and exposed to fly ash samples in tissue culture tubes. Viability and the phagocytotic capacity were used as test parameters. Macrophages exposed to fly ash were investigated with the scanning electron microscope.
The fly ash samples investigated were stack samples as well as samples from electrostatic filters predominantly obtained from a pulverised coal fired power plant. A sample of the fluidised bed combustor at Apeldoorn (Netherlands) was also investigated. The stack ash samples appeared to be more cytotoxic for bovine macrophages than the samples of the electrostatic filters. A fine fraction (2.3 µm) of the electrostatic filter ash, however, was far more cytotoxic than the complete ash.
Cytotoxic effects were, however, found at concentrations higher than α-quartz, the tested model compound of which it is known that it causes fibrinogenic reactions to man and animal.
The adverse effect on the main function of the macrophage, phagocytosis, was considerable for some stack ash samples. It should be noted that the defense of the lung against inhaled particles and bacteria may be endangered by the decreasing number of phagocytosing cells.

A PRELIMINARY LITERATURE STUDY FOR A RESEARCH PROGRAMME ON HUMAN TOXICOLOGICAL EFFECTS OF STACK EMISSIONS OF COAL-BURNING INSTALLATAIONS

Author : G.J. Vink
Report no. : R 84/57
Order no. : 10293
Date : May 1st, 1984
Language : Dutch

SUMMARY

A preliminary study of literature data on human toxicological aspects of the stack emissions of coal-burning installations has been carried out for the Dutch "National Research Programme on Coal Technology". The aim of the study was to contribute to the formulation of a research programme on the biological effects of these emissions.
The Dutch government, and the Bureau of Energy Research Projects, intend to start a programme of research into the nature and the extent of possible effects of increased emissions of coal combustion products resulting from increased use of coal for energy supply, for man and the environment.
The following subjects have been successively considered: the concept of "human toxicological research", the quantitatively most important air pollutants in stack-emissions from coal combustion, the effects of these components on man or mammals, the effects of total emissions and stack fly ash on man, mammals or in-vitro test systems.
A survey of human toxicological research in the Netherlands on air pollutants is also given.
Comparison of the expected concentrations of important emission components in the air with "maximum immission concentrations" (MIC-values) and the "lowest effect concentrations" (LEC) quoted in the literature leads to the following conclusions:
Coal use of 20.10^6 ton per year results in SO_2 and NO_x concentrations which probably remain below the MIC- and LEC-values. In the most unfavourable case (high variant of contribution by coal combustion) concentrations up to 50% of the MIC-values can be reached.

Risks from these compounds may be expected to be highest where they are found together with stack fly ash with absorbed trace elements and polycyclic aromatic hydrocarbons (PAH). However the expected contribution of stack fly ash and the PAH from pulverised coal-burning installations is low, but could be substantial locally around small installations.
Uncertainties with regard to possible synergistic actions of these components in combination with fly ash form a problem for both short- and longterm exposure, which requires further research and risk analysis.
The elements with the greatest expected concentration increase in the air, Al, As, B, Hg and Sr, are (with the possible exception of Hg) not expected to show toxic effects for humans; toxicological data for Sr are, however, lacking.
No human effects are expected from (H)Cl or (H)F; (H)F can possibly affect cattle.

Data on epidemiological research around combustion installations are scarce.
Data about in-vitro research of fly ash supplies useful additional information on specific toxic properties and reactions but this must be evaluated for clear interpretation.
On the basis of these findings a preliminary research programme has been formulated.

SOME TOXOLOGICAL PROPERTIES OF FLY ASH IN RELATION TO PARTICLE SIZE

Author : G.J. Vink

Presented at : AshTech '84, Second International Conference on Ash Technology and Marketing, Barbican Centre, London, September 16-21, 1984

Language : English

SUMMARY

The toxicological properties of samples of fly ash of Polish coal from a pulverised coal combustor in the Netherlands were investigated. Studies were carried out with samples of both bulk fly ash, and fly ash from the final zone of the electrostatic precipitators.

Results on cytogenic effects in bacterial and cell culture test systems, on cytotoxic effects in a bovine lung macrophage, in vitro test system and on primary skin and eye irritation effects in rabbits are presented. Some eco-toxicological results concerning the influence of fly ash in test systems with plants and aquatic animals are given.

RE-INTRODUCTION OF COAL ACCOMPANIED BY INVESTIGATION INTO FLY ASH

Author : P.B. Davis
Published in : Project (1984), 393-395
Language : Dutch

SUMMARY

It is possible to calculate the distribution of fly ash in the atmosphere by means of mathematical models. It was found that the largest concentrations of fly ash were found at some kilometers north of the chimney. (0.3 $\mu g/m^3$ annual average). Whereas the back ground concentration is 40 $\mu g/m^3$. This shows that a 600 MW power plant will give no noticeable addition to the genotoxicity of the dust in the atmosphere, when there are no reactions giving an extra increment of the genotoxicity of the chimney ash.
Only when chemical reactions should cause a hundredfold increase of the genotoxicity of the chimney ash, the situation should be different. This has to be measured in future. Based on these measurements it will be possible to judge over the possible influence on human health. However, at this moment there is no reason to be anxious about. It is also necessary to investigate the neighbourhood of small installations. They have a low chimney and the fly ash has possibly a high specific mutagenicity.

CYTOTOXICITY OF COMBUSTION PRODUCTS FROM SMALL BURNING INSTALLATIONS FOR LUNG MACROPHAGES

Authors : R.N. Hooftman
P. Roza

Report no. : R 84/194
Order no. : 10293
Date : December 5th, 1984
Language : Dutch

SUMMARY

The cytotoxic properties of a number of bag filter ash samples and a cyclone ash sample from small coal fired installations in De Lier and Zevenhuizen for lung macrophages were investigated. The bag filter ash samples from De Lier and the cyclone ash sample from Zevenhuizen were not cytotoxic; the induced effects were comparable with those caused by inert material. The bag filter ash from Zevenhuizen caused a small cytotoxic effect; both survival of the cells and fagocytic capacity were slightly affected.

THE INDUCTION OF CHROMOSOME ABERRATIONS IN CULTURED MAMMALIAN CELLS BY COMBUSTION PRODUCTS FROM SMALL COAL FIRED INSTALLATIONS

Author : R.N. Hooftman
Report no. : R 84/195
Order no. : 10293
Date : December 5th, 1984
Language : Dutch

SUMMARY

The genotoxic properties of a number of combustion samples from small coal fired installations were investigated. The samples came from two installations used for heating greenhouses in De Lier and Zevenhuizen. Stack ash, bagfilter ash and (only in Zevenhuizen) cyclone ash were sampled and tested for their ability to induce chromosome aberrations in a genotoxicity test with a mammalian cell line (Chinese Hamster Ovary cells). With the exception of the cyclone ash, all samples induced a range of chromosome aberrations. Application of metabolic activation did not increase genotoxicity. The effects agreed to a large extent with those found in the Ames test, although some differences were found. It is likely that the effects in the two test systems are induced partly by different compounds on the fly ash.

It is believed that this is the first report of the induction of chromosome aberrations by fly ash. The results of the investigation described in this report warrant the immediate investigation of the ability of other fly ash samples to induce chromosome aberrations.

THE MUTAGENICITY OF COMBUSTION PRODUCTS FROM SMALL COAL-FIRED INSTALLATIONS, MEASURED WITH THE AMES TEST

Authors : P.B. Davis
H.A. Bakkeren

Report no. : R 84/198
Order no. : 10293
Date : January 15th, 1985
Language : Dutch

SUMMARY

The mutagenicity of extracts of fly ash from the cyclone, the bag filter and the stack of two small, coal fired installations was investigated in the Ames test, a test for the induction of mutations in special strains of the bacterium Salmonella typhimurium.

Organic extracts of cyclone ash from a 3,4 MW_{th} atmospheric fluidized-bed combustor in Zevenhuizen were not mutagenic. Organic extracts of both bag filter ash and stack ash from the same installation, and from a 3,8 MW_{th} underbedfeeder in De Lier were mutagenic.

The mutagenicity of the extracts of stack ash sampled with the installation under full or partial load was 400-600 revertants/m^3 flue gas (strain TA98, without metabolic activation) in Zevenhuizen (= 140-230 revertants/MJ) and 20,000 revertants/m^3 flue gas in De Lier (= 11,000 revertants/MJ). The mutagenity of stack ash sampled in De Lier when the installation was tempered (low energy production) was 64,000 revertants/m^3 flue gas (= 22,000 revertants/MJ); coal consumption, and thus also flue gas production, were low, however, under these conditions, so that the emission per unit time was low.

The mutagenicity of organic extracts of the bag filter ash samples in strain TA98 (without metabolic activation) was 75 revertants/mg (42,000 revertants/MJ) in Zevenhuizen and 1300 revertants/mg (= 130.000-300,000 revertants/MJ) in De Lier.

The emission of mutagenic compounds to the atmosphere in Zevenhuizen was of the same order of magnitude as reported in the literature for other installations. The emission to the atmosphere in De Lier is among the highest reported values. In both installations, however, only a small part of the mutagenicity (1-6%) was emitted to the atmosphere; by far the greater part of the mutagenicity was recovered in the bag filter ash.

Metabolic activation had generally little effect on the mutagenicity of bag house or stack ash. About half of the mutagenicity in strain TA98 (without metabolic activation) was nitro-reductase-dependent. Strain TA100 was somewhat less sensitive than TA98 for the mutagenicity of stack ash from both installations; this is in agreement with earlier results with stack ash from a pulverised coal combustor; TA100 was, however, relatively insensitive for the mutagenicity of the bag filter ash, which may indicate that different compounds are responsible for (at least a part of) the effect in the two types of ash. Heated bag filter ash was no longer mutagenic.

Aqueous extracts of the bag filter ashes were not mutagenic.

POSSIBLE APPLICATIONS OF WASTE FROM COAL BENEFICIATION

Author : J.I. Walpot
Reference No. : 83-01613
File No. : 8722-50068
Date : March, 1983
Language : Dutch

SUMMARY

In the framework of an investigation into the cleaning of coal prior to combustion (KRV) by using physical (HGMS and ESS) and biological processes, a review has been made of potential applications for the residual materials resulting from these KRV-processes. Among other things literature on coal flotation wastes is used for this review.
Applications offering the best prospects in this respect are:

- addition in pulverised coal combustion;
- fuels/raw materials in cement production;
- fuels/raw materials in brick manufacture;
- fuels in fluid bed combustion.

Particularly with regard to the last quoted method of processing some research work has still to be carried out to find out whether it will actually be possible to use the residual materials resulting from coal cleaning (KRV) for this application.

APPLICATION OF COAL ASHES FROM FLUIDIZED BED COMBUSTION

Authors : J.I. Walpot
B. Boesmans
B.G. ten Dam
A.O. Hansveit

Presented at : Coal Technology Europe
3rd European Conference & Exhibition on Coal Utilization
11-13 October 1983, Amsterdam
Volume 4, 31-55

Language : English

SUMMARY

More or less concurrently with the development of fluidized bed combustion as an energy-conversion technique, research is in progress with TNO concerning the processing and application of ashes from a 4MWth AFBB-installation. One of the most striking features of these ashes is their particularly high carbon content.
The leaching characteristics of the ashes are almost completely governed by the presence of lime. No hazardous biological toxicity/mutagenicity has been shown to exist in the leachates.
The ashes are employed as filler material for asphalt, and their use as a constituent of construction materials for building purposes is being investigated.
Laboratory investigations and preliminary economic evaluation have shown future recovery of aluminium from AFBC ashes to be promising.

APPLICATION OF FLUIDIZED BED COMBUSTION ASHES FROM COAL

Author : B. Boesmans

Presented at : AshTech '84, Second International Conference on Ash Technology and Marketing, Barbican Centre, London September 16-21, 1984
Proceedings pages 677-683

Language : English

SUMMARY

As a part of the TNO-programme on fluidised bed combustion of coal research is being executed into the possibilities of applying ashes. Based on literature and analyses of ashes from the TNO-4MW$_{th}$ AFBB-installation the research is focussed on applications as: bricks, concrete, artificial gravel, cement, road filler and metal ore. The typical problems and benefits of the utilization of fbc-ashes caused by the high contents of unburnt coal, calcium combinations and particle shape are discussed. The possibilities and difficulties of marketing fbc-ashes in relation to its value and to the competition of pcc-ashes and other recycling materials as demolition waste and gypsum have been set out.

TNO

LEACHING OF COAL ASH

Authors : R. Gerritsen
J.I. Walpot
Reference No. : 83-01610
File No. : 8722-50065
Date : February 1983
Language : Dutch

SUMMARY

In the framework of the National Coal Research Programme (NOK): fluid-bed combustion of coal, research has been carried out into the long-term leaching characteristics of fabric-filter ash, the ash being produced during operation of the 4 MWth AFBB installation. This research has been carried out on the instructions of the Bureau for Energy Research Projects (BEOP) of the Dutch Energy Research Centre (ECN), a foundation in the Netherlands.
Research has also been carried out on PCC-ash, to compare the properties of ash produced by conventional pulverised coal installations with those of AFBC-ash.
To make the results suitable for application in actual practice, use has been made of columns, the diameter, height and volumetric flow rate of which have been chosen such that they have different values to assess the influence of these parameters on the leaching characteristics.
In addition to the column tests, so-called cascade shaking experiments have been carried out, which are expected to yield a fair approximation of the column test results, but in a much shorter time. In order to determine the degree in which the components can be leached from the AFBC filter ash, leaching experiments have been carried out at fixed pH-values.

POSSIBILITIES FOR THE RECOVERY OF METALS FROM COAL-ASH

Author : J.I. Walpot
Reference No. : 5060-300
File No. : 80-011059
Date : September 1980
Language : Dutch

SUMMARY

The metals arsenic, beryllium, selenium, aluminium, gallium, germanium, molybdenum, nickel, scandium, uranium, vanadium, iron and titanium have been selected for an investigation into the economic feasibility of their recovery from coal ash, on the basis of data with respect to environmental pollution, data from literature concerning the recovery of these metals from coal ash or their intrinsic value in coal ash. The technological possibilities of recovery of the selected materials from coal ash and from other sources have been investigated, and a study has been made of possible market developments.

The results of this investigation show that an economic recovery of metals from coal ash is only possible for aluminium; the same may apply to iron, but this has not been proved so far, and the recovery of nickel and selenium can be attractive in the near future.

The processes to be preferred for aluminium recovery from coal ash are chlorination and lixiviation after sintering with a calcium-containing additive.

From the point of view of environmental impact the latter process is more favourable, with flue-gas desulphurization-sludge as a sintering additive and sulphuric acid as a lixiviating agent. Among metals recovered as by-products are: iron, titanium, uranium and manganese.

TNO

ALUMINIUM RECOVERY FROM AFBC ASH (PART ONE)

Authors : J.I. Walpot
R. Gerritsen

Reference No. : 82-09263
File No. : 8722-50063
Date : July 1982
Language : Dutch

SUMMARY

For the purpose of carrying out research into the fluid bed combustion of coal (4 MWth-afbb) in the framework of NOK and in pursuance of the results of a study of the possibilities of the recovery of metals from coal-ash residues, tentative investigations have been made on the recovery of aluminium from afbb-filter ash.
These investigations have been based on a process comprising leaching as a first step, and recovery of the final product from the solution by further treatment and purification.
This first stage of the research involved optimalisation of the leaching process. It has been found that the best way to effect leaching is with moderately concentrated sulphuric acid (approximately 2.0 M) at a temperature of approximately 100°C for about half an hour.
A rough evaluation of the economics of the process has indicated that there is a reasonable chance that the recovery of aluminium from afbb-ash can compete with the aluminium recovery starting from bauxite as a raw material.

ALUMINIUM RECOVERY FROM AFBC ASH (PART TWO)

Authors : R. Gerritsen
J.I. Walpot
Reference No. : 84-03757
File No. : 8722-10282
Date : March 1984
Language : Dutch

SUMMARY

A continued investigation has been carried out into the possibility of recovering aluminium from fluid-bed ash originating from the 4 MW_{th} AFBB-installation located at TNO-MT, Apeldoorn.

Chapter 1 and 2 give the scope and objective of the research carried out; chapter 3 discusses the plan of the experimental investigations. Chapter 4 deals with the results of the experimental research and gives a survey of the literature in the field of alternative aluminium recovery processes. It appears that about 70% of the aluminium present in the ash can be recovered in one single extraction step, and that the major contaminants going into solution consist of iron, magnesium and alkali metals.
The results of the research have been used to draw up a process scheme for the recovery of 25.000 tons per annum of aluminium from 160.000 tons per annum of AFBC-ash.
In chapter 5, on the basis of this process scheme, economy of the process is compared with the economy of other processes for the recovery of alumina. Moreover cost-price calculations have been made for the recovery of 50.000 tons per annum of aluminium sulphate (14 aq) from 54.000 tons per annum of AFBC-ash.
Cost-profit analysis show the production of aluminium oxide from AFBC-ash to be much more expensive than that from bauxite, but the production of aluminium sulphate can indeed be economically feasible.
Finally in chapters 6 and 7 conclusions are given and recommendations are made for further research.

INVESTIGATION INTO THE DEMAND OF BUILDING MATERIALS IN RELATION TO THE STREAM OF COAL WASTE TO BE EXPECTED

Author : B.G. ten Dam
Report No. : B-81-101
Order No. : 60.5.0001
Date : May 1981
Language : Dutch

SUMMARY

In this report the commercial situation in the building materials trade is outlined as far as it is important for the application of coal waste. Attention is given to:

- logistics;
- trade limitations;
- momentary use of coal waste in the Netherlands and possibilities of expanding markets;
- technical limitations for the use of coal waste.

POSSIBILITIES AND LIMITATIONS OF THE USE OF COAL WASTE IN THE BRICK INDUSTRY

Author : J.H. van der Velden
Reference No. : 81-09051
File No. : 8711-50600-50-00
Date : June 1981
Language : Dutch

SUMMARY

General requirements for coal waste for use in the brick industry are discussed. Attention is also given to the environmental effects of the use of fly-ash in the brick industry.
This report also gives a provisional and rough estimation of the maximum quantity of fly-ash, which can be used by the Dutch brick industry in future at a favourable cost price level of this material.

APPLICATION OF AFBC COAL ASH IN CONCRETE, ASPHALT FILLER AND ARTIFICIAL GRAVEL

Authors : B. Boesmans
R. Gerritsen
Reference no. : 83-09557
File no. : 8722-10283
Date : August 1983
Language : Dutch

SUMMARY

In this investigation attention was given into the application of AFBC coal-ashes in concrete, asphalt filler and artificial gravel.

The investigation into the application of AFBC-ash in concrete has been restricted to additional analyses and considering the technical and economic possibilities. Concluded was, that there are small-scale and large-scale possibilities.

Investigating the possibilities of AFBC-ashes as an asphalt filler, the asphalt qualities were checked. It was found that the ashes were a good filler, but the economic value is low and the market is small for asphalt fillers.

Investigation into the application of AFBC-ash in artificial gravel was carried out in a process where base materials as sand, pulverised-coal fly ash and lime are granulated and hardened by heat treatment. It was found that the granules, made in this way, could substitute gravel with a concrete quality B 22,5.

THE USES OF FLUIDIZED-BED ASHES IN CONCRETE

Author : A.J.M. Siemes
Report no. : B-84-684/60.5.0001
Date : February 15, 1985
Language : Dutch

SUMMARY

From the atmospheric fluid bed combustion ashes result for which a useful application is looked for. In imitation of the use of conventional fly ashes, the application in concrete can be considered. The composition of the fluid bed ashes differs from the conventional fly ashes in respect to the particle size, the absorption, the amount of pozzolanic material and the sulphate content.
From literature hardly anything is known of the effects of fluid bed ashes in concrete. For this reason a preliminary experimental investigation has been done with regard to:

- the need for water of the mortar;
- the air content;
- the workability;
- the compressive strength of the concrete;
- the shrinkage;
- the carbonation;
- the frost- and de-icing salts resistance;
- the erosion resistance.

The concrete properties are established after different curing times to get a good insight into the effects of the puzzolanic reactions. For the investigation three types of ashes have been used, a filter ash, a cyclone ash and a bed ash.

Starting from a standard concrete composition the following functions of the ashes are reviewed:

- the substitution of binder;
- the substitution of the fine part of the sand;
- the addition of extra aggregate.

The compressive strength of the concrete is reduced, mainly due to the higher water content of the mix. For the filter and the cyclone ash this reduction is becoming smaller in the course of time as a consequence of pozzolanic reactions. For the bed ash the reduction increases, probably due to the forming of ettringite.
Based on the results of the tests a survey is given of the concrete strengths that are possible with the ashes in mixes with different water cement ratios. The higher this factor is, the lower the relative strength will be. In that respect replacing cement by ashes gives the highest reduction. In general however, it is possible to make concrete mixes with acceptable strength levels.
One of the mixes (A1B2C2) in which 50 kilogram (per m3 mortar) sand was replaced and 50 kilogram extra ash was added, showed a remarkable good result. After one year curing this mix had a compressive strength comparable to the strength of the unmodified concrete. This concrete mix also showed good results in the tests for porosity, frost- and de-icing salt resistance and erosion. Optimization of this mix is advised.

The carbonation depth of the modified concretes is about three times higher than that of the unmodified concrete. The applicability of ashes in reinforced concrete is reduced because of this phenomenon. It should be evaluated if the carbonation rate will reduce in the course of time due to further hydration and pozzolanic reactions.

The resistance against frost, de-icing salts and erosion is lower for the modified concretes than for the unmodified ones. As mentioned before the mix A1B2C2 gives a better result.

Based on the test results an evaluation is made of the applications which can be given to the modified concretes.

INVESTIGATION INTO THE SULPHUR BOND IN COAL BY MEANS OF X-RAY FLUORESCENCE SPECTROMETRY

Authors : P.M. Houpt
W.G.J. van der Leest

Report No. : CL 80/91
Order No. : 5074
Date : September 11th, 1980
Language : Dutch

SUMMARY

The possibility was investigated of defining the concentrations of various bond forms of sulphur in coal by means of the Kβ- X-ray line. This line is formed by the return of an electron from the M-shell to the K-shell of the sulphur atom. Since the M-shell holds the valence electrons, the bonding situation of the sulphur will influence the energy of the M,K-transition and therefore the extent of Kβ-radiation.

The most frequently occuring bond in sulphur compounds in coal is an organic one; sulphur bound as sulphate and sulphur bound as iron sulphide. Model measurements have been executed on $CaSO_4$; $CS(NH_2)_2$; FeS_2 and S, and on mixtures of $CaSO_4$ + FeS_2 and of $CaSO_4$ + $CS(NH_2)_2$.

From the results of measurements it is concluded that it is possible to distinguish between the three individual bond forms of sulphur by means of the SKβ-line. However, in a mixture of several bond forms of sulphur, only sulphate-bound sulphur clearly manifests itself via the Kβ-line with the available equipment.

INVESTIGATION INTO COAL BY MEANS OF SCREEN ELECTRON MICROSCOPY TOGETHER WITH X-RAY MICRO-ANALYSES (REM/RMA)

Author : N. van der Burgh
Report No. : CL 81/41
Order No. : 50750
Date : April 4th, 1981
Language : Dutch

SUMMARY

Samples from seams D and N of the Emil Mayerlichs coal mine, belonging to the Eschweiler Bergbauverein, were investigated by means of screen electron microscopy combined with X-ray micro-analysis (REM/RMA). By investigating the fracture surfaces as well as the ground surfaces, data were collected on the morphological construction of the coal, the kind of minerals in the coals and the distribution thereof in the coal.
It was found that the minerals in the samples investigated were significantly different.
In seam D the minerals were found to be present in the form of small inclusions as well as very thin layers between the macerals. The minerals of layer N on the other hand mainly formed thin layers between the macerals.
Inclusions of pyrite, iron oxides or carbonates were found in seam D, however hardly at all in seam N. In seam N, spherical and cylindrical inclusions of kaolinite were found.
Plant structures could be clearly observed in both samples.
The results of the investigation into the ground surfaces were processed into dispersion diagrams. These diagrams clearly demonstrate, that in seam N there are more aluminium silicates than in seam D and that there are also more dispersions in seam N than in seam D. In the diagrams of seam D pyrite and other iron compound dominate. It has been shown that morphologic research with REM and spatial analysis of fracture surfaces, together with RMA of the ground surfaces can make an important contribution to the characterization of coal.

MONITOR FOR THE SPECIFIC DETECTION OF NICKEL CARBONYL IN FLUE GASES, BASED ON CHEMOLUMINESCENCE

Author : P.M. Houpt
Report No. : CL 81/112
Order No. : 50860
Date : September 28th, 1981
Language : Dutch

SUMMARY

The monitor described measures the intensity of the chemoluminescence rays at 500 nm, emitted by the reaction of nickel carbonyl with ozone and carbon monoxide. It is suitable for measurements on flue gases. Apart from the detection unit, the monitor contains a purification system to clean the carbon monoxide needed from the considerable quantities of impurities formed by nickel and iron carbonyls.
The detection limits for nickel carbonyl with the monitor are 1 ppb linearly up to some ppm's.
The interference of iron pentacarbonyl and nitrogen monoxide have been eliminated by the application of optical and spatial separation of the chemoluminescence and by the correct choice of the pressure in the reaction chamber.

INVESTIGATION INTO THE BONDING FORMS OF SULPHUR IN COAL BY MEANS OF X-RAY ANALYSIS COMBINED WITH SCREEN ELECTRON MICROSCOPY

Authors : J.M. Timmner
N. van den Burgh

Report No. : CL 82/59
Order No. : 50750
Date : May 4th, 1982
Language : Dutch

SUMMARY

In the first part of this research a number of standard samples has been investigated by means of a combination of screen electron microscopy and X-ray micro analysis (REM/RMA).
The object of this research is to determine what information this technique can provide concerning the bonding forms of sulphur in coal. The results have been compared with data from literature.
Attention has been given to the Kα-, Kβ- and L-spectra of sulphur respectively. An investigation has been made into the relationship between the nature of sulphur bonds and line displacements on one hand and the occurrence of satellite peaks and peak widths at half height of certain peaks on the other hand. The displacements of the Kα- and Kβ-lines as a result of the bonding situations of sulphur have the same trend as that mentioned in literature. The values of these displacements differ from the values quoted in literature, however, which for that matter also show important mutual differences. There is a more or less linear correlation between the intensity relations of the $K\beta_1$-line and the satellite line SKβ' ($IK\beta_1/ISK\beta'$) and the degree of oxidation of sulphur in the range of 0 to 6^+.
The L-spectra on the contrary do not give any useful information concerning the degree of oxidation of sulphur, but oxygenous sulphur compounds can be discriminated from elementary sulphur, organic sulphur, and sulphides by means of these spectra.

Furthermore, sulphates distinguish themselves from sulphides by a lower $ISK\alpha_4/ISK\alpha_3$ value.
The form in the L-spectra is more sensitive to differences in the bonding situation of sulphur than that of the K-spectra. The optimal acceleration voltage has been determined as being 10 kV for the examination of sulphur compounds with the aid of L-spectra.

A MONITOR FOR THE SPECIFIC DETERMINATION OF NICKEL CARBONYL BASED ON THE CHEMILUMINESCENCE REACTION WITH OZONE AND CARBON MONOXIDE

Authors : P.M. Houpt
A. van der Waal
F. Langeweg

Published in : Analytica Chemica Acta, 136 (1982), p. 421-424
Language : English

SUMMARY

The chemiluminescence, measured at 500 nm., allows nickel carbonyl to be measured in the range 1 ppb to several ppm in combustion gases. Interferences from iron pentacarbonyl and nitrogen monoxide are eliminated by optical and spatial discrimination and by choice of the pressure in the reaction chamber.

OPTIMALIZATION OF THE ANALYSIS METHODS FOR COAL AND FLY ASH

Author : M. Hissink
Reference No. : 83-0317
File No. : 8712-10297
Date : January 1983
Language : Dutch

SUMMARY

To support the experiments in the Atmospheric Fluidized Bed Boiler many analyses of samples of fuel, additives and waste are needed. Therefore it was necessary to obtain experience with regard to sampling, sample preparation and analysis of these materials.
For a correct interpretation of the experiments in the AFBB a knowledge of the accuracy of the results of these analyses is essential. This accuracy depends largely on the sampling method.
As a guide for the methods to be used the instructions in the relevant ISO Standards have been followed usually. A description is given of the procedures for sampling and analysis as well as of the equipment used. The accuracy that can be expected for the various results of analyses has been established experimentally. It was found in general that the requirements were met.

INVESTIGATION INTO THE APPLICABILITY OF A METHOD DERIVED FROM MICRO-ELEMENT ANALYSIS TO THE DETERMINATION OF THE TOTAL-SULPHUR CONTENT OF COAL PRODUCTS

Authors : G.J. Rotscheid
mrs. P.C.M. Staal
W.J. Buis

Report No. : 83/0454/1207/10298/GR/CK
Language : Dutch

SUMMARY

The purpose of this investigation was to check in how far an existing analysis method in use in micro-element analysis could be used for measurement of the total-sulphur content of coal products, to improve this method and to make it applicable to this kind of products.
The method is based on the principle of combustion in a tube, the sample to be analysed being destructed oxidatively at a high temperature in a pure oxygen atmosphere in a quartz tube and as a result of which all sulphur is oxidized to SO_2 and SO_3.

For the final determination of the sulphur oxides formed, use has been made of a wet chemical oxidation of SO_2 to SO_3, which reacts with water to H_2SO_4, after which the SO_4^{2-} is determined by a precipitation titration with Ba^{2+} ions.
It has been indicated in this report what alterations have been made to existing equipment and working method with the purpose to find a quick, simple and cheap method for measurement of the total-sulphur content of coal products with an accuracy which is at least comparable with that of the methods now forming part of the ASTM or ISO.

The investigations carried out so far have been mainly concerned with the conversion of sulphur in coal. The method of final determination used in these investigations has certain limitations in that it is less sensitive with the measurement of low sulphur contents (< 0.2%).

APPLICATION OF SCANNING ELECTRON MICROSCOPY IN COMBINATION WITH X-RAY MICROANALYSIS TO CHARACTERIZE COAL

Authors : N. van der Burgh
J.M. Timmner

Report no. : R 84/90
Order no. : 12583
Date : June 17, 1984
Language : Dutch

SUMMARY

In this report a survey is given of the work during the years 1981, 1982 and 1983 regarding coal characterization by means of scanning electron microscopy in combination with X-ray microanalysis (SEM/XRMA).
Attention was paid to the following subjects:

1. Composition of the coal as to minerals and macerals, apart from a qualitative analysis of the minerals, a study having been made of the way in which the minerals and macerals are bound.
2. The quantitative analysis of the elements occurring in the minerals, i.e. Al, Si, Cl, K, Ca, Ti and Fe, by the side of the determination of the total sulphur content and the content of organically bound sulphur.
3. The chemical binding of sulphur.

Extensive subreports on the subjects described were published in the course of the period of investigation.
A list of these subreports forms part of this report.

FULLY AUTOMATED MEASUREMENT OF ORGANIC SULPHUR IN COAL BY MEANS OF SCANNING ELECTRON MICROSCOPY COMBINED WITH X-RAY MICROANALYSIS

Authors : J.M. Timmner
N. van der Burgh

Report no. : R 84/89
Order no. : 12583
Date : June 5, 1984
Language : Dutch

SUMMARY

Scanning electron microscopy combined with energy-dispersive X-ray micro-analysis (SEM/XRMA) affords special possibilities in the structural investigation and analysis of coal.
This paper describes a fully automated version of SEM/XRMA for the determination of organic sulphur in coal. The values found by this technique are in good agreement with those found by the usual classical methods, provided the coal has a low content of pyrites. For coals high in pyrite, however, the classical methods give considerably lower values of organic sulphur than SEM/XRMA does. In the classical analysis, the values are obtained by so called subtraction methods. The discrepancy is very probable due to the incomplete extraction of pyrite inclusions in the wet analysis of coal. The result is a too low value for the pyrite content, and consequently, a too high value for organic sulphur.
Apart from primarily being a method for determining organic sulphur, the method presented affords additional information; for example, it may enable one to conclude whether a sample of coal is uniform or a blend of several types of coal.
An exploratory investigation into an improved wet-chemical method of analysing pyrite and sulphate, described in the literature, gave no satisfactory results. The method involves ashing of the coal in a low-pressure oxygen plasma, the organic sulphur escaping as sulphur dioxide. In contrast with the literature data, it is highly probable that some of the escaping sulphur dioxide combined with ash constituents to produce sulphates.

AUTOMATED DETERMINATION OF ORGANIC SULPHUR IN COAL; USE OF SCANNING ELECTRON MICROSCOPY AND ENERGY DISPERSIVE X-RAY MICROANALYSIS

Authors : J.M. Timmer
N. van der Burgh

Published in : Fuel, 63 (1984), 12, 1645-1648
Language : English

SUMMARY

Scanning electron microscopy combined with energy-dispersive X-ray microanalysis in fully automated form was used for the determination of organic sulphur in coal. The values found by this technique are in good agreement with those found by the usual classical methods, provided the coal has a low content of pyrite. For coals high in pyrite, however, SEM/XRMA gives considerably lower values of organic sulphur. The discrepancy is probably due to the incomplete extraction of pyrite inclusions in the wet analysis leading to a low value for the pyrite content, and consequently, a high value for organic sulphur. Apart from primarily being a method for the determination of organic sulphur, the method presented affords additional information; for example, it may enable a differentiation to be made between a single coal sample and a blend of several types of coal.

TNO

CHARACTERIZATION OF COAL ASHES

Authors : R. Gerritsen
W.M.A. Cox
J.I. Walpot

Reference No. : 81-07014
File No. : 8707
Date : May, 1981
Language : Dutch

SUMMARY

In this report the results are given of the chemical and physical characterization of various kinds of ash, like fly ashes, fluidized bed ashes and ashes from coal gasification. Furthermore, the results of the chemical characterization are compared with the limits given by the Chemical Waste Act. Arsenic in coal ashes in general, and non-converted sulphur sorbents (CaO) in fluidized bed ashes can give rise to serious problems.

Finally, five coal ash samples were investigated tentatively on their environmental impact (as deposits) by means of leaching. In the first period (10 days), leaching was carried out under similar conditions; in the second period these conditions were changed according to our own views independently of one another to gain optimal information from the experiments. In general it is not possible to obtain more than superficial information, due to the interaction of various factors of influence. For more definite conclusions a better structural planning of the experiments is necessary.

INVESTIGATION OF FLY ASH PARTICLES WITH RESPECT TO MORPHOLOGY AND SURFACE ENRICHMENT OF MATRIX AND TRACE ELEMENTS

Auhtors : A.J. Gay
A.P. von Rosenstiel
P.J. van Duin

Published in : "Microbeam Analysis 1981", pages 229-236
Roy H. Geiss, editor
Proceedings of the 16th Annual Conference of the Microbeam Analysis Society, Vail, Colorado, 13-17 July 1981

Language : English

SUMMARY

In this investigation a fly ash was used originating from a U.S. bituminous coal of relatively high ash content (17.9%) and low sulphur content (1.52%), burned in a pulverized-coal fired power plant. Four sized fractions were obtained. The eight most important matrix elements (Al, Ca, Fe, K, Mg, Na, Si, Ti) and sulphur, as well as fifteen arbitrarely chosen toxic trace elements (As, B, Be, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Se, Tl, V, Zn) were analysed in a received sample together with four sized fraction samples by scanning electron microsopy (SEM), energy dispersive spectroscopy (EDS), atomic adsorption spectroscopy (AAS), emission spectroscopy (ES), and instrumental neutron activation analysis (INAA). Surface analysis and depth profiling were performed by electron spectroscopy for chemical analysis (ESCA), Auger electron spectroscopy (AES) and secondary ion-mass spectrometry (SIMS). Furthermore, trace element concentrations of individual single particles were analysed by laser-microprobe mass analysis (LAMMA).

Morphologically the fly ash presents a standard appearance of predominantly spherical particles with some irregular ones. There is remarkable enrichment in S and C on the smaller particles, with a more or less constant composition for the other elements.

The results from the elemental concentrations as obtained by ESCA and after successive sputter etching of 50 and 350 Å show a high surface concentration of S and C and an increase of the matrix element concentrations of Al, Ca, Fe and Si with increasing depth.
Most remarkable are the nearly constant surface concentrations for all fractions, despite the large differences in bulk concentrations. Furthermore evidence was also found of the presence of Pb and Cd in the surface layer of the fly ash fractions, which means that these elements are present on the surface in concentrations of at least 1%.
The AES spectra clearly show the high surface concentrations of C, S and O. With increasing depth the C and S intensities decrease rapidly, whereas the matrix elements Si, Al, Ca and Fe increase. Data by SIMS show a thin surface layer with a number of trace elements such as As, Ba, Cd, Cr, Cu, Mo, Ni, Pb, V and Zn.
Analysis by LAMMA shows largely different trace element concentrations of Ba, Cr, Cu, V and Zn. Negative secondary ion LAMMA spectra show the presence of larger amounts of F, Cl, P and S as well as hydrocarbons, PO_2^-, PO_3^-, SO_2^-, SO_3^-, SO_4^- and HSO_4^-.

TNO

CHARACTERISATION OF SAMPLES FROM AFBB EXPERIMENT RUN 002

Author : J.I. Walpot
Reference no. : 82-03019
File no. : 8722-50062
Date : March, 1982
Language : Dutch

SUMMARY

During a combustion experiment carried out with the 4 MW_{th}-afbb, which lasted for a period of five days, samples were taken daily from the coal and lime supplied to the installation and from the ashes produced in filter, cyclone and fluidised bed, for the purpose of testing the operating stability of the installation. At the end of the testing period a large sample of filter ash was kept apart to make a further investigation into the possibilities of leaching, the recovery of metals and the preparation of useful products. Ash containers were sampled for comparison of daily samples.
Physical and chemical characteristics of these samples showed filter ash to be highly porous and toxic with respect to the "WCA (Wet Chemische Afvalstoffen)" on account of its high content of calcium(hydr)oxide.
Recirculation of cyclone ash resulted in a better combustion of the coal, although there were considerable differences in the extent of combustion of the individual particles. This may probably be remedied by choosing a more favourable distribution of the coal supply.
It proved impossible to draw conclusions with respect to matters such as operating stability of the installation and combustion- and sulphur capture-efficiency because there were not sufficient data for drawing up a (reliable) mass balance.

A STUDY OF THE COMPOSITION AND MORPHOLOGY OF FLY ASH FROM COAL-BURNING INSTALLATIONS

Authors : A.J. Gay
A.P. von Rosenstiel

Report no. : CL 82/55
Order no. : 20520, 50043
Date : April 22, 1982
Language : English

SUMMARY

The composition and morphology of fly ash originating from the same sort of coal (US bituminous), burned in two different processes (PCC and FBC), has been investigated.
Morphological differences are discussed. Bulk analysis of sized fractions has shown that the concentration of some elements increases with decreasing particle size. The increase is more marked in PCC fly ash than in FBC fly ash.
Surface analysis has shown a very high concentration of some elements on the surface of fly ash particles. The elements are present in the surface layer as oxides and sulphates.

THE COMPOSITION AND MORPHOLOGY OF FLY-ASH FROM COAL BURNING INSTALLATIONS (PCC AND FBC)

Authors : A.J. Gay
A.P. von Rosenstiel
P.J. van Duin

Presented at : Coal Technology Europe '82
2nd International Coal Utilization Exhibition and Conference, 22-24 September 1982
Bella Center, Copenhagen, Denmark
Proceedings pages 241-155

Language : English

SUMMARY

PCC fly-ash

For matrix elements there is an increase in magnesium and a decrease in titanium respectively with a decreasing particle size. Other matrix elements remain constant within the limits of experimental error. There is also an increase in concentration of sulphur and a pronounced increase of carbon with decreased particle size.

FBC fly-ash (without limestone)

The behaviour of matrix elements is similar to that of PCC fly-ash, except for the fact that there is no significant increase in the concentration of magnesium with decreasing particle size. Both sulphur and carbon show a marked decrease in concentration with decreasing particle size, however. For trace elements the trend is the same as for PCC fly-ash, i.e. an increase in concentration with decreasing particle size, although to a much smaller extend than is the case for PCC fly-ash. Arsenic and sélenium (both volatile materials) show the most defiant behaviour, i.e. an almost constant concentration, almost independent of particle size. Berillium shows a greater increase with decreasing particle size than is the case for PCC fly-ash.
Silicon, iron and titanium show no enrichment in their content in smaller particles. Magnesium shows a marked enrichment with decreasing particle size, whereas in FBC fly-ash it shows the opposite trend except for a slight enrichment in the smallest fraction. Calcium shows a trend

similar to that of magnesium, but its enrichment in PCC fly-ash is less pronounced. Potassium shows a slight depletion in both types of fly-ash. Sodium shows a slight enrichment in PCC fly-ash and quite a marked one in FBC fly-ash.

CHARACTERIZATION OF COAL SAMPLES BY MEANS OF REM/RMA

Authors : N. van den Burgh
J.M. Timmner

Report No. : R 83/52
Order No. : 61162
Date : April 8th, 1983
Language : Dutch

SUMMARY

The minerals of seven coal specimens were quantitatively analysed by means of screen electron microscopy combined with energy dispersive X-ray micro analysis (REM/RMA). These specimens were also characterized likewise with the aid of REM/RMA and special attention was given to the way in which the minerals and macerals are combined. There were significant differences in the relationship between macerals and minerals in the samples investigated.
Three different preparations were used in this investigation. It was concluded that it is sufficient to investigate one of these types, in this case polished cross-sections of 3 mm samples, to obtain a good average impression of the structure of coal specimens.

CHARACTERIZATION OF TWO COAL SAMPLES BY MEANS OF SCREEN ELECTRON MICROSCOPY COMBINED WITH X-RAY MICRO ANALYSIS (REM/RMA)

Author : N. van der Burgh
Report No. : CL 82/206
Order No. : 50680
Date : June 1st, 1983
Language : Dutch

SUMMARY

As part of a "round robin" type of investigation commissioned by the Dutch Centre for Coal Specimens, two samples of coal, designated 16.001.000 (501 GB 28) and 16.002.000 (503 BE 33), were investigated by scanning electron microscopy combined with X-ray micro analysis. The results were set against data on a sample (V7DE) which had previously been investigated as part of a coal purification project, and which had shown promising purifiability.

In the samples eight elements from the "mineral" fraction and sulphur were determined quantitatively. The samples were then ashed in a current of singlet oxygen generated by a high-frequency discharge ("cold ashing"), and the ashes where analysed likewise.

The structural relation of minerals and macerals in the samples was estimated by morphological examination together with X-ray micro analysis of fractured as well as polished specimens. As regards the structural relation of minerals and macerals, samples 16.001.000 and 16.002.000 appeared to be very similar.

The minerals were found to be present largely as granules intimately mixed with the macerals, not as layers between them. Sample 16.002.000 contained much less ferrous sulphide than sample 16.001.000, but in both samples the ferrous sulphide was present as inclusions in the macerals. In this respect they differed widely from sample V7DE, which has a markedly layered structure that allows macerals and minerals, including ferrous sulphide, to be more or less easily separated, and is therefore readily prepurifiable.

TNO

SAMPLING AND MEASUREMENTS ON BOILER 12 OF THE POWER PLANT AT NIJMEGEN

Author : W.F.M. Hesseling
Reference no. : 82-05884
File no. : 8726-50031
Date : May, 1982
Language : Dutch

SUMMARY

A sampling and measurement program was carried out at a pulverised-coal boiler of the Nijmegen electric power plant. During the combustion of Polish coal, samples were taken from the coal, pulverised coal, bottom ash, fly ash and flue gases. During the measurements and sampling the power was about 105 Mwe, the average dust content of the flue gases 15 mg/m^3 n.t.p., average flue gas temperature 120°C, average NO_x concentration about 740 mg/m^3 n.t.p. and the average concentration of SO_2 about 1600 mg/m^3 n.t.p. The average concentration of CO in the flue gas was lower than 10 mg/m^3 n.t.p.
The samples were used in various TNO investigations.

DETERMINATION OF THE INORGANIC MATRIX OF COAL ASH FROM AN ATMOSPHERIC FLUID-BED BY FTIR ANALYSIS

Author : R. Bosman
Published in : Fuel, 62 (1983), 9, 1009-1012
Language : English

SUMMARY

Three infrared methods for the quantitative determination of some inorganic compounds in coal and coal ash are evaluated. Attention is focused on calcium compounds occuring in ash from an atmospheric fluidized bed boiler. The IR measuring range was extended to 200 cm^{-1} where scattering was less a problem. A major difficulty in quantitative IR spectrometry remains the preparation of satisfactory KBr or CsI discs. However, the results are difficult to obtain otherwise.

TNO

PYROLYSIS-GAS CHROMATOGRAPHY-MASS SPECTROMETRY OF COAL

Author : E.Ch.Th. Gevers
Report no. : 84/16/1207-10303/EG/LH
Date : February 1984
Language : Dutch

SUMMARY

In this report an investigation into the usefulness of pyrolysis GC/MS is described as a technique for the characterization of coal. Samples of known elementary composition purchased from the Pennsylvania State Coal Data Base, were pyrolysed at a temperature of 750°C.
The pyrolysis temperature should be high enough to produce a sufficient amount of volatile compounds, but not too high lest a too far breakdown to undesirably small and unspecific molecules should occur. The actually chosen temperature, though not very critical, must be very well reproducible.
The pyrolysis products were analysed on line by means of gas chromatography-mass spectrometry. Among the pyrolysis products approximately forty different compounds have been identified: small molecules like H_2S, COS, SO_2 and CS_2; aromatic hydrocarbons like benzene, toluene, xylenes and naphthalene; heterocyclic aromatics like thiophenes and furanes and further phenols, alkanes and alkenes.
On the basis of literature data more than thirty pyrolysis products of each coal species have been quantified. The results have been subjected to chemometric procedures as principle component analysis and clustering. Comparison of the results leads to some conclusions regarding the chemical composition of the different coals.
In order to gain more insight into the relationship between the properties that are relevant for the use of different coals as a fuel and the composition of the pyrolysis products analyses of a larger amount of coal species is an absolute necessity. Besides the application of a novel technique like negative ion chemical ionisation mass spectrometry for coal analysis looks promising.

MORPHOLOGY AND CHEMICAL COMPOSITION OF SOME FLY ASH SAMPLES FROM A COAL FIRED POWER PLANT

Author : A.J. Gay
Report no. : R 84/91
Order no. : 10293
Date : June 19, 1984
Language : English

SUMMARY

Samples of fly ash collected from the electrostatic precipitators (ESP) and from the stack of a coal-fired power plant were analysed for morphology and chemical composition.
Morphology was studied by means of light and scanning electron microscopy. Concentrations of major and minor elements (Al, Ca, Fe, K, Mg, Na, Si, Ti), sulphur and carbon as well as those of trace elements (As, Be, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Se, V, Zn) in fly ash and coal were determined by various methods. Fly ash from the ESP shows morphological and chemical composition typical for its kind. Fly ash from the stack shows incongruous values for some major elements, sulphur and carbon, no enrichment in trace elements corresponding to its particle size, and is strongly contaminated with fibres from the filter and with iron oxide from the sampler. This suggests the inadequacy of the sampling method and apparatus used for this purpose.

A CONVENIENT METHOD COMBINING THE MORPHOLOGICAL AND CHEMICAL ANALYSIS OF COAL FLY ASH WITH PARTICLE SIZE DETERMINATION

Authors : A.J. Gay
P.J. van Duin
N. van der Burgh

Presented at : Proceedings AshTech '84, 2nd International Conference, September 16-21, London

Language : English

SUMMARY

Fly ash from a pulverised coal combustor (or any other combustor, e.g. fluidised bed) is examined with a scanning electron microscope. Micrographs thus obtained give information about the morphology of the fly ash.

Microanalysis with an energy dispersive X-ray spectrometer and a special computer programme for ZAF correction, in combination with scanning electron microscopy allows for a quick determination of the concentrations of up to 8 major elements.

From SEM micrographs the average particle size as well as its standard deviation, the distribution curve and the distribution histogram are determined using an HP 85 computer - HP 911A Graphics Tablet combination.

In this way satisfactory chemical and physical characterisation of fly ash can be performed in matter of hours and the results stored for further reference in the form of photographs and computer print-outs.

MORPHOLOGY AND CHEMICAL COMPOSITION OF SOME FLY ASH SAMPLES FROM THE TNO'S ATMOSPHERIC FLUIDIZED BED BOILER TEST FACILITY

Author : A.J. Gay
Report no. : R 84/139
Order no. : 10293
Date : September 11, 1984
Language : English

SUMMARY

Samples of bag filter ash and of particulate matter from the stack of the TNO's Atmospheric Fluidised Bed Boiler (AFBB) test facility were analysed for morphological and chemical composition. Morphology was studied by means of light and scanning electron microscopy. Concentrations of major and minor elements (Al, Ca, Fe, K, Mg, Na, Si, Ti), carbon, sulphur, nitrogen, hydrogen and chlorine, as well as those of trace elements (As, Be, Cd, Cr, Cu, Ni, Pb, Sb, Se, Zn) and several polycyclic aromatic hydrocarbons (PAHs) were determined by various methods. Samples of bag filter ash show morphological and chemical composition typical for fly ash from a fluidised bed combustor. Samples of particulate matter from the stack cannot be, however, regarded as fly ash. They contain large amounts of ammonium chloride (formed in the sampler) and are highly contaminated by fibres from the sampler filter and by material originating from the AFBB installation. This questions the adequacy of the sampling method and apparatus used for this purpose.
Concentrations of PAHs show considerable variations between runs. On the whole they are much higher in the particulate matter from the stack than in the bag filter ash.

STUDIES OF CARBONACEOUS CENOSPHERES FROM FLUIDIZED BED COMBUSTORS

Authors : A.J. Gay
R.F. Littlejohn
P.J. van Duin

Presented at : The 2nd International Conference on Carbonaceous Particles in the Atmosphere, Linz, September 1983

Published in : "The Science of the Total Environment", 36 (1984), 239-246, Elsevier, Amsterdam

Language : English

SUMMARY

Fly ash from fluidized bed combustion of bituminous coals has been shown to contain carbonaceous cenospheres of several forms including thick walled opaque, thin walled with well defined skeletal structure and single "balloons" with skeletons. The influence on cenosphere formation of factors such as particle size, temperature, rate of heating, petrographic composition etc. is reviewed. The probability of cenospheres surviving beyond the combustion zone is estimated from calculations of burn-out time for a range of conditions and the particular significance for fluidized bed combustion is demonstrated.

Particles from both experimental and industrial scale combustors have been studied using scanning electron microscopy. Differences in chemical composition between the three principal types of cenospheres were demonstrated by using SEM/EDS analysers.
The possible significance of cenospheres in the environment is discussed.

THE FORMATION OF CARBONACEOUS CENOSPHERES DURING FLUIDIZED BED COMBUSTION OF BITUMINOUS COALS

Authors : A.J. Gay
R.F. Littlejohn
P.J. van Duin

Published in : Fuel 62 (1983) 10, 1224-1226
Language : English

SUMMARY

The discovery of cenospheres in fly ash from fluidized bed combustors is reported and differences between the fluidized bed and more conventional combustors likely to affect cenosphere formation are indicated.

Morphological studies and qualitative chemical analyses obtained by scanning electron microsopy/energy dispersive X-ray spectrometry (SEM/EDS) techniques show differences in composition between the "window" areas of cenospheres and their "skeletons".

The formation of cenospheres from coal was first reported some sixty years ago, but, so far none have been found among the particles discharged by conventional combustion appliances. However, recent studies of particles carried in the off-gas from fluidized bed combustors burning two bituminous coals have revealed the presence of many typical coal cenospheres. The SEM micrograph shows some of those among the particles that were fine enough to remain in suspension after the gases had passed through a cyclone separator. They were recovered by isokinetic sampling and collection on a fine filter.

Factors influencing cenosphere formation are known to include heating rate, particle size, surrounding atmosphere and petrographic constitution. This last was also seen in the present work where it was found, that the proportion of cenospheres increases with the greater proportion of vitrinite. It has also been shown that cenospheres ignite more readily than their parent coal, so that survival after formation will require either low temperature or a low concentration of oxygen.