# Artificial lightweight aggregates as utilization for future ashes – A case study

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ABSTRACT

In the future, more electricity in the Netherlands will be produced using coal with co-combustion. Due to this, the generated annual ash volume will increase and the chemical composition will be influenced. One of the options for utilization if present markets are saturated and for use of fly ashes with different compositions, is as raw material for lightweight aggregates. This was selected as one of the best utilizations options regarding potential ash volume to be applied, environmental aspects and status of technology. Because of this, a study has been performed to assess the potential utilization of fly ash for the production of lightweight aggregate. Lightweight aggregate has been produced in a laboratory scale rotary kiln. The raw material consisted of class F fly ash with high free lime content. An addition of 8% clay was necessary to get green pellets with sufficient green strength. The basic properties of the produced lightweight aggregate and its behaviour in concrete have been investigated. The concrete has a good compressive strength and its leaching behaviour meets the most stringent requirements of Dutch environmental regulations. The carbon foot print of concrete will be negatively influenced if only the concrete itself is taken into account, but the reduction of the volume weight has advantages regarding design, transport emissions and isolation properties which may counteract this. In the Dutch situation the operational costs are higher than expected potential selling price for the LWA, which implies that the gate fee for the fly ash is negative.



## 1. Introduction

### 1.1. Background

Nowadays about 36% of the electricity in the Netherlands is being generated with coal (ECN, 2010). This share will increase in the near future. The Dutch coast is an attractive location for new coal fired power plants because of the availability of sea transport for fuel, cooling water and the availability of natural gas fields and gas transport grids for future $CO_2$ storage. Up to 2015 several new coal fired power plants will be commissioned in the Netherlands. Thereby, the total capacity will increase from 4.18 to 7.75 $GW_e$. The new power plants will be engineered to co-fire high percentages of biomass. It is expected that wood will be the main source for biomass and further some agricultural residues, depending on the future Dutch policy on subsidies (KEMA, 2010).

In the Netherlands, all ashes from coal fired power plants are utilized since the late eighties. Utilization of these ashes is also prescribed by the Dutch National Waste Management Plan, which is the reference for permits within the Environmental Management Act. Up to now, the major part of the generated fly ashes is used in concrete and cement industry as pozzolanic filler and as alumina source for production of Portland clinker. Further, fly ashes are used as asphalt filler and to a small extent as clay replacement in ceramics.

One of the options for utilization if present markets are saturated, is as raw material for lightweight aggregates, mainly for use in concrete. This was selected as one of the best utilizations options regarding potential ash volume to be applied, environmental aspects and status of technology.

Lightweight aggregates for use in concrete were produced in the Netherlands up to 2000 using the Aardelite and Lytag process technology. Both production plants were closed for economical reasons when more and more fly ashes were utilized as pozzolanic filler in concrete. However, 10–15 years later, time has come for a reassessment of the potential utilization of fly ash for production of lightweight aggregate as the ash volume may strongly increase in the future.

### 1.2. Lightweight aggregates from fly ash

Most production processes of lightweight aggregates (LWA) from both primary materials (clay or shale) and from secondary

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materials like fly ash and sludges have the same basic steps, namely:

- Mixing of the raw materials with water and additives to adjust the properties of the green pellet for hardening, transport, processing et cetera.
- Agglomeration to produce a granular aggregate (green pellet), which can be obtained by granulation or compaction.
- Hardening of the green pellets by sintering, hydrothermal or cold bonding to form the final product.
- After-treatment, which may consist of crushing, sieving and/or wetting.

Hardening will be performed at different temperatures depending on the type of binding of the LWA to be obtained. Basically, four ways of hardening/bonding are possible using fly ash (see also Table 1). Cold bonding and hydrothermal bonding are based on comparable reaction mechanisms as the bonding in normal concrete: formation of calcium alumina silicate hydrates. Sintering processes are based on the creation of a ceramic matrix. The matrix consists of alumina silicates, mainly mullite and amorphous phases. The sintering temperature for alumina silicate fly ashes is typically in the range 1100–1200 °C.

With increasing temperature more and more melted phases will be formed, where at the end of the range complete melting is obtained. In this study, sinter process technology is selected to be worked out in detail.

It is noticed that matrices can also be based on organics, like (recycled) thermoplastics in combination with a bloating agent (Slabaugh et al., 2007); these type of LWA are not further taken into account.

Several production processes have been developed in the past and some of these were commercialised. Most of these processes, like Lytag, Trefoil and Minergy are based on creation of a ceramic matrix (Van der Gaag and Wevers, 2003; Minergy, 2009; Lytag, 2005). More detailed information is given in Table 2. For agglomeration of the pellets commonly a granulation disc is used. Sintering/hardening is performed in a rotary kiln or a sinter strand at temperatures in the range of 1100–1200 °C. The Aardelite process is based on hydrothermal bonding whereby the pozzolanic properties of the fly ash are used, by adding lime as reactive agent. Eurolite is based on cold bonding using cement, with pozzolanic reactions as secondary mechanism. Both processes were also based on agglomeration with a granulation disc.

**Table 1**
Classification of LWA hardening processes based on temperature indication for inorganic matrices.

| Production process | Temperature (°C) | Binding |
|---|---|---|
| Cold bonding | 0–100 | Matrix with calcium alumina silicate hydrates |
| Hydrothermal | 100–250 | Matrix with calcium alumina silicate hydrates |
| Sintering | 1100–1200 | Material bridge bonding with alumina silicates |
| Complete melting | >1200 | Alumina silicates |

### 1.2.1. Important aspects for LWA production as utilization route for future fly ash surplus are

- Flexibility to process fly ashes with different chemical and physical properties. It is expected that in the future, ash composition will differ more from plant to plant, due to different co-combustion percentages and to different secondary fuels. This means that the process has to be flexible enough to use these fly ashes while still producing LWA with constant properties, which is essential for concrete industry.
- Sales assurance of the produced LWA, which is essential for effective ash management of power plants.
- Investment and operational costs versus selling price. According to the deliverer of a hydrothermal LWA production plant with a capacity of 250,000 tons per year, the operational costs will be about 12–15 EUR/ton (Aarding, 2008). The investment costs are about 8.3 MEUR. It must be stated that the lay-out of this plant is very basic. In the Dutch situation, the investment will be somewhat higher due to stringent environmental regulations (like dust emissions). The investment costs to produce a sintered LWA are higher, namely about 35 MEUR (Van der Gaag and Wevers, 2003). However this was designed for dredged sludge as raw material, which makes extended pre-treatment necessary. The operational costs were estimated to be about 20–55 EUR/ton. These data are updated with correction for 2.2% average inflation to 2009. The costs are exclusive sales and costs of area. In general it can be stated that the operational costs of LWA production based on sintering, will be about double as high as LWA production based on hydrothermal hardening.
- Status of technology. LWA production technology with fly ash is proven for both sintering and cold bonding on commercial scale. As far as we know, plants are in operation in Poland and USA.
- Environmental aspects. Advantages of the production and use of LWA made from or with waste materials are reduced use of virgin materials and re-use of waste materials. On the other hand the production and use of LWA influences its environment by emissions to the air of greenhouse gases and other compounds, emissions to surface water, emissions to soil and groundwater during its use and use of additives, fuels et cetera.

## 1.3. Utilization of lightweight aggregates in concrete

Since early ages materials which can be characterised as lightweight aggregates have been used in concrete (aggregates of

**Table 2**
Production processes for lightweight aggregates based on coal fly ash.

| | Aardelite | Eurolite | Lytag | Trefoil | Minergy |
|---|---|---|---|---|---|
| Raw materials | Fly ash<br>Lime | Fly ash<br>Cement | Fly ash<br>Bentonite | Fly ash<br>Sewage sludge<br>Clay | Fly ash<br>Sewage sludge<br>Paper sludge |
| Temperature (°C) | 80–90 | 20–50 | 1100–1200 | 1100–1150 | 1000–1200 |
| Matrix | CASH[1] | CASH | AS[1] | AS | AS |
| Particle density (kg/m$^3$) | 1450 | 1370–1400 | 1350–1650 | 1420–1520 | 1450–1700 |
| Water absorption 24 h (%) | 25 | 32–36 | 15 | 13–15 | 25 |
| Particle range (mm) | 0–25 | 0–25 | 0–14 | 0–16 | 10–20 |

[1] CASH = Calcium alumina silicate hydrates; AS = alumina silicates.

pumice, crushed bricks and lava). In WW II lightweight aggregate concrete was used to build ships in the USA. After WW II LWA concrete is being used for different types of concrete constructions and products. According to an ACI committee, most important reasons to select LWA concrete is reduction of dead load, flexibility in design, saving of construction cost and superior long term durability (EuroLightCon, 1998). Lytag was used in the Netherlands for both reinforced and pre-stressed concrete. In 1985, the Sacharov Bridge was successfully constructed using Lytag as LWA in the concrete. The reduction of dead load (also for the foundations) leads to 5–10% lower building costs (EuroLightCon, 1998). Other applications of LWAC with Lytag are:

– Building and pavement blocks with characteristic compressive strength 5–15 MPa.
– In place cast structural concrete with characteristic compressive strength 25–35 MPa.
– Prefab concrete self supporting system floors with characteristic compressive strength 45–60 MPa.

The hydrothermal produced Aardelite was only used in the Netherlands for non-reinforced concrete, mainly building blocks due to its limited compressive strength.

Properties of LWAC based on Lytag and other LWA are presented in several reports and articles (CUR, 1994), (Haque and Kayali, 2004) and within the EU-project EuroLightCon (Eurolightcon, 1998).

### 1.4. Production on lightweight aggregates based on fly ash

#### 1.4.1. Energy and raw materials

Table 3 gives some specifications of the use of energy and raw materials for the Lytag process in the Netherlands, which is based on sintering using a sinter strand. Electricity is used for conveyors, mixers, granulation discs etc. Heating of the pellets is mainly obtained by oxidation of the fuel in the pellet itself, namely unburned matter and some additional coal. Some heavy oil is used for the burners, which have to ignite the fuel in the pellets.

According to the Dutch experience some clay (bentonite, dry) had to be added to obtain a green pellet, with sufficient strength to be handled in the Lytag process (whereby a sinter strand is used). If a rotary kiln is used, higher strength of the green pellet is needed. Instead of primary materials, waste materials like dredged sludge can be used. Ramamurthy and Harikrishnan concluded that the use of bentonite as additive up to 20% to improve the ballability of fly ash on a granulation disc, also improved pellet strength and water absorption (Ramamurthy and Harikrishnan, 2006). Scanning electron microscope studies showed that pore size distribution (finer and closed pores) changed due to addition of bentonite.

Lytag was produced from fly ash from dry bottom boilers. Some trials were performed with addition of 5–8% m/m fly ash from coal gasification (IGCC). The use of these fly ashes caused problems during the sintering process of the pellets. The sintering process became unstable: lower sintering temperatures, hard surface and incomplete firing inside the pellet (KEMA, 1998). These phenomena were related to the lower melting point and the smaller particle size of these ashes ($D_{50} \pm 7$ μm) and the influence on the grain size distribution of the ash mix of the green pellet.

**Table 3**
Use of energy and raw materials for the production of 1 ton of sintered LWA (Lytag process in the Netherlands) (KEMA, 1996).

| Raw material | Unit | Amount |
|---|---|---|
| Fly ash | kg/ton | 1047[1] |
| Additive | kg/ton | 8 |
| Pulverised coal | kg/ton | 7.5 |
| Heavy oil | kg/ton | 6.8 |
| Water | $m^3$/ton | 0.8 |
| Energy consumption | kWh/ton | 32.2 |

[1] Including 6% m/m loss on ignition.

Shuguang et al. tried to improve the mechanical properties of LWA from sintered fly ash by modifying the mineralogical composition (Shuguang et al., 2010). The hypothesis was that if cordierite ($2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$) is formed in LWA, less micro cracks will be formed during cooling of the LWA as this mineral has an excellent thermal shock resistance. However, Mg was added as a carbonate/hydroxide, which decomposes during heating and sintering. The generated $CO_2$ and $H_2O$ influenced the porosity of the pellet. Therefore it is difficult to draw unequivocal conclusions about the influence on strength. Some authors focussed their research on LWA based on other raw materials like Wang et al., who investigated mixtures of dewatered sewage sludge with 0–32% coal fly ash addition (Wang et al., 2009). The mixtures were pressed into cylindrical specimen and sintered at 1050–1100 °C for 30 min. Mixtures with 18–25% coal fly ash that was preheated at 420 °C for 20 min and then sintered at 1100 °Cfor 30 min obtained the highest quality regarding strength, water absorption and density.

#### 1.4.2. Environmental aspects of LWA production

Thermal processes lead to direct emission by flue gasses. A very relevant aspect is the emission of greenhouse gases, caused by the total production process. A carbon foot print can be calculated for different processes, taking also the emissions related to the additives into account. In Table 4 an indication is given of the $CO_2$ emissions for production of LWA per $m^3$ of LWA. Process data are based on (KEMA, 1996; Aarding, 2008; Sarabèr, 2001); the conversion factors (c.f.) are derived from official carbon foot print calculations (ProRail, 2010; Strukton, 2010) and some other studies (Ecofys et al., 2009a,b).

As seen in Table 4, the greenhouse emissions of cold bond products are lower than sintered products despite the relatively high $CO_2$ emissions due to the production of cement and lime itself.

It is assumed that part of the residual organic matter in the fly ash which originates from biomass is equal to the co-combustion percentage (=33%) in the power plant, where the fly ash was generated. This part of the organic matter does not contribute to the $CO_2$ emission in relation to the greenhouse effect.

Starting point is that the application of LWA meets the leaching requirements of the Dutch Decree on Soil Quality, which is the relevant regulation for application of building materials in civil engineering and the outside of buildings (walls, roofs, foundations, etc.). Leaching tests on LWA concrete (cold bond and sintered), with closed and open structure, meet the requirements for utilization without restrictions (Intron, 1994).

#### 1.4.3. Use of primary materials

Sintered LWA requires no binder but clay and some oil or coal as secondary fuel. The clay can be replaced by secondary materials like contaminated dredged sludge. A positive effect is that for every $m^3$ LWA one $m^3$ of natural gravel can be saved.

### 1.5. Proposed production process

The process scheme of the proposed LWA production process for this study is presented in Fig. 1.

After mixing/blending the raw material has to be formed into pellets. The shape and strength of the pellets depends on the granulation process. Techniques suitable for fly ash are agitation, granulation and compacting. Based on the experience with Lytag,

**Table 4**
Indication of $CO_2$ emissions for production of LWA [kg/m$^3$] in the Netherlands.

| Per ton LWA | Unit | c.f. | Sinter process | | Hydrothermal based | | Cold bonding | |
|---|---|---|---|---|---|---|---|---|
| | | | Amount | kg $CO_2$ | Amount | kg $CO_2$ | Amount | kg $CO_2$ |
| Fly ash (exl. 6% m/m LOI) | kg | 0 | 992 | 0 | 865 | 0 | 779 | 0 |
| 6% carbon in fly ash | kg | 2.85 | 60 | 114 | 60 | 0 | 60 | 0 |
| Additive | kg | 0 | 8 | 0 | - | - | - | - |
| Pulverised coal | kg | 2.42 | 7.5 | 18 | - | - | - | - |
| Heavy oil | kg | 3.185 | 6.8 | 22 | - | - | - | – |
| Natural gas | m3 | 1.825 | 0 | 0 | 13 | 24 | - | - |
| Water | kg | 0.0003 | 800 | 0 | 150 | 0 | - | 0 |
| Energy consumption | kWh | 0.615 | 32.2 | 20 | 15 | 9 | 11.4 | 7 |
| Lime | kg | 1.04 | 2.8 | 2.9 | 55 | 57 | - | - |
| Cement | kg | 0.76 | - | - | - | - | 115 | 87 |
| Transport raw materials[1] | ton km | 0.13 | 161 | 21 | 139 | 18 | 6 | 20 |
| Transport LWA[2] | ton km | 0.13 | 180 | 23 | 188 | 24 | | 23 |
| Total per ton | | | | 221 | | 133 | | 137 |
| Total per m$^3$ | | | | 321 | | 225 | | 206 |

[1] Transport distance 150 km.
[2] Including 20% moisture.

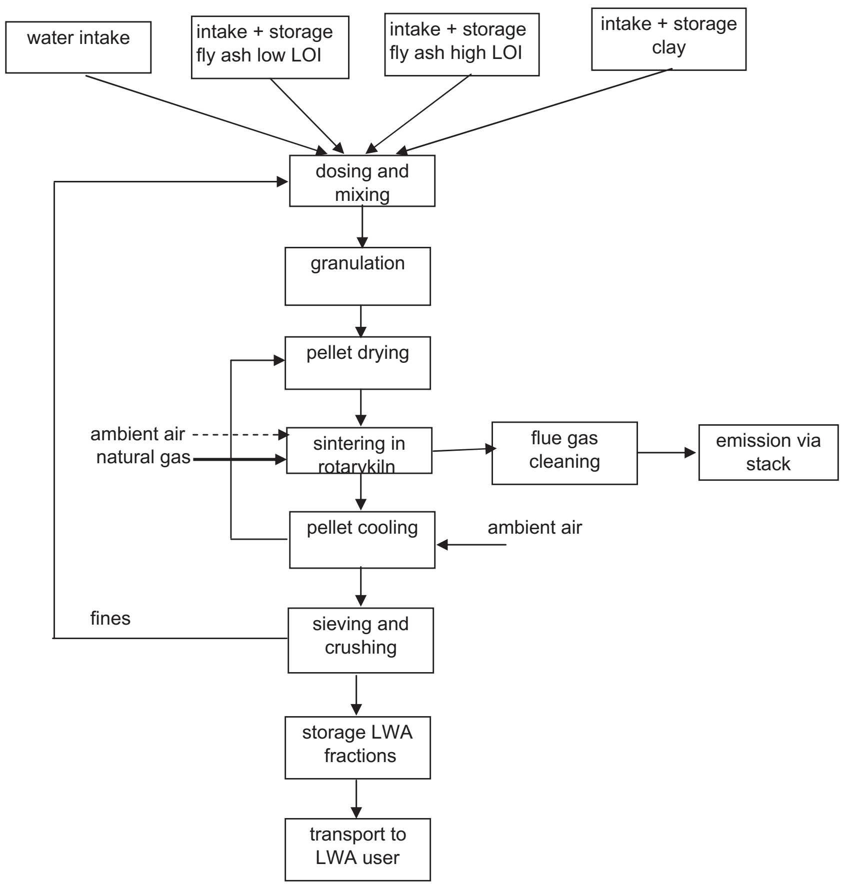


**Fig. 1.** Process scheme as proposed in this study.

Aardelite and Trefoil, it is concluded that pellets can be produced with a granulation disc with sufficient green strength.

Before sintering, pellets are dried using preheated air from the pellet cooler. The water content of the pellets is maximum 5% m/m when they leave the pellet dryer with a temperature of about 500 °C.

There are three basic types of sintering equipment, namely rotating kilns, sinter strands and shafts. In this study a rotary kiln has been chosen. The kiln is normally made of a steel cylinder with an insulating lining of thermal and chemical resistant material (general thickness lining in the range 80–300 mm). The kiln has a burner at the input side to ignite the organic matter in the green pellet. The kiln gases are captured in a gastight exhaust hood for further processing (cleaning, energy recovery). Advantages of a rotary kiln above a sinter strand is better heat recovery from flue gases, better control of sintering process and consistent shape of pellets. An important disadvantage is the higher mechanical impact on the green pellet. Wainwright developed an improved rotary kiln, the so called Trefoil kiln. This kiln uses a kiln bed with a typical trefoil. Several advantages of this type of rotary kiln are mentioned like better thermal efficiency, faster heat-up and cool-down and higher load-factors (Wainwright and Cresswell, 2001). The kilns that Wainwright used at Leeds University were designed and supplied by Trefoil on a loan basis. When the university closed this facility these kilns were taken by RTAL/Trefoil to Chong Qing University, China and were used in the trials of this study.

Before sieving and crushing, the pellets have to be cooled with ambient air in a fluidized bed cooler. After the pellet cooler, the air is used for pellet drying and then used as secondary air in the rotary kiln.

The last step in the production process is size grading of the pellets. Fines are recycled in the process.

## 2. Experimental production of lightweight aggregates

### 2.1. Introduction

As part of the EOS project experiments were carried out to investigate the sintering behaviour of fly ash, especially the trace elements, and further to assess the physical, chemical and mineralogical properties of the produced LWA and the basic properties of concrete whereby the coarse aggregate is replaced by the produced LWA.

**Table 5**
Overview of test methods.

| Test | Method |
|---|---|
| Particle density | NEN-EN 1097-6 |
| Loose bulk density | NEN-EN 1097-3 |
| Water absorption | NEN-EN 1097-6 Annex C |
| Crushing resistance | Pin crusher test |
| Compressive strength concrete | NEN-EN 12390-7 |
| Elasticity modulus | ISO 6784 |

**Table 6**
Mix composition of concrete.

| Parameter | Unit | Mix with gravel | Mix with LWA |
|---|---|---|---|
| Cement | $kg/m^3$ | 300 | 300 |
| Water free | $kg/m^3$ | 150 | 150 |
| Sand | $kg/m^3$ | 985 | 985 |
| Gravel | $kg/m^3$ | 985 | 0 |
| LWA | $kg/m^3$ | 0 | 598 |
| Air | $l/m^3$ | 10 | 10 |
| Density theoretically | $kg/m^3$ | 2420 | 2033 |

### 2.2. Materials

Fly ash was used from coal fired power plant Hemweg 8, which is a dry bottom boiler (opposed wall burner configuration). This fly ash is representative for Dutch pure coal fly ash. The content of macro and trace elements is representative for the Dutch situation based on the Database Trace elements (KEMA, 2009b).

### 2.3. Experimental

#### 2.3.1. Sinter experiments

This coal fly ash was enriched with calcium and to a small extent with potassium and phosphorus. This enrichment is based on the expected future co-combustion situation, which will be mainly woody biomass. The main influence of co-firing woody biomass is the increase of free calcium oxide. Calcium was added as $Ca(OH)_2$, which will decompose to CaO during heating, while potassium and phosphorus were added as KOH and $P_2O_5$ (all analytical quality). The chemical composition of the raw mixture is presented in Tables 5 and 6. All compounds are expressed as oxides (remark: it does not mean that these are present as oxides in the fly ash).

Bench scale experiments are conducted at the Institute of Resources & Environment Science at the University of Chong Qing. At first some trials were performed to establish the minimum amount of clay to get green pellets with sufficient strength. Based on these trials addition of 8% m/m primary clay is chosen for these experiments (minimum was 5% m/m). The clay used in the experiments is coming from a local field in the Shapingba district. The fly ash is mixed with the clay and granulated with a drum granulator. The moisture content of the green pellets was 24.1% m/m (on dry). The pellets are fired in several batches (3–4 kg per batch) in a rotary kiln. The length and diameter of the kiln are about 0.8 m and 0.20 m respectively. The lining of the kiln is made of incaloy. The residence time is about 25 min combined with a rotation speed of 7–8 rpm.

Firing of the batches was almost continuous, the kiln is emptied at 1000 °C and recharged at 500 °C and then fired at sintering temperature of 1166–1206 °C. In Fig. 2 temperature profile is given during firing of the green pellets.

#### 2.3.2. Content of macro elements X-ray fluorescence

The macro-elements Al, Ca, Fe, K, Na, P, S, Si and Ti were measured with X-ray fluorescence. The sample is melted together with

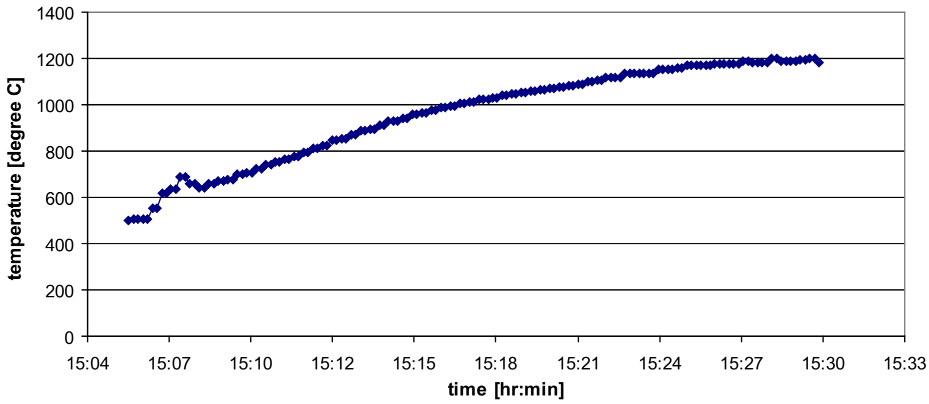


**Fig. 2.** Temperature development during firing the green pellets in the rotary kiln.

lithium tetra borate. S was measured after cold pressing of the sample.

*2.3.3. Content of trace elements*

The trace elements were analysed by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) after total digestion. Hg was measured with CV-AFS (cold vapour atomic fluorescence spectroscopy). The anions Br, Cl and F were measured with IC (ion chromatography).

*2.3.4. Mineralogical properties of raw materials and produced LWA: X-ray diffraction*

The qualitative X-ray diffraction measurements (XRD) were carried out with a Philips diffracto meter (CuKa-radiation). The scan velocity was 0.01 2$\theta$/s.

*2.3.5. Mineralogical properties of raw materials and produced LWA: free lime*

Free lime was analysed in accordance with NEN-EN 450-1.

*2.3.6. Loss on ignition*

Loss on ignition is analysed gravimetrically according to NEN-EN 450-1.

*2.3.7. Basic properties LWA*

Several basic properties of the LWA were tested, which are summarised in Table 5.

*2.3.8. Properties of concrete made with produced LWA*

The behaviour of LWA in concrete was tested using a concrete composition, with a moderate water/cement ratio of 0.50 and 300 kg/m$^3$ Portland cement (CEM I 42.5R) as binder. The mix composition of the concrete is presented in Table 6.

Concrete samples with gravel or LWA have been produced using the following procedure:

- Before mixing, LWA was saturated with water during more than one day.
- Cement, water and sand were mixed together during 4 min in a forced action mixer.
- LWA was added to the mortar and mixed till it was homogeneous (about 3 min).
- after moulding samples were stored at > 95% RH/20 °C for 24 h
- 24 h after moulding the samples were demoulded and stored in water at 20 °C for 27 days.

The E-modulus and compressive strength were determined after 28 days according to ISO 6784 and NEN-EN 12390–7 respectively.

The leaching behaviour was measured using the Dutch Tank Test according to NEN 7375, which is a standardised diffusion test for bound materials, like concrete.

*2.4. Results*

*2.4.1. Chemical properties of raw materials and produced LWA*

The aggregate tests and the concrete tests were performed with LWA material smaller than 16 mm. The aggregate was brown with a spherical shape.

The measured concentration of the main compounds is presented in Table 7. There was a good match between the calculated and the measured concentrations in LWA for the macro compounds. The maximum relative deviation was <15%, with exception of Cl, S which decreased due to its volatile behaviour. Sulphur is not only volatilized, but also combusted as far as it is organically bound in residual organic matter.

**Table 7**
Concentration of main compounds in fly ash (without enrichment with Ca, K and P) clay and produced LWA (% m/m).

| Component | Fly ash | Clay | LWA |
|---|---|---|---|
| $SiO_2$ | 57.72 | 63.9 | 53.1 |
| $Al_2O_3$ | 23.90 | 17.58 | 22.15 |
| $Fe_2O_3$ | 7.86 | 7 | 8.08 |
| CaO total | 3.51 | 0.37 | 8.54 |
| MgO | 1.58 | 0.95 | 1.34 |
| $Na_2O$ | 1.55 | 0.2 | 1.42 |
| $K_2O$ | 1.56 | 2.36 | 2.10 |
| $P_2O_5$ | 0.30 | 0.1 | 1.28 |
| $TiO_2$ | 0.99 | 1.13 | 1.00 |
| $Mn_2O_3$ | 0.055 | 0.1 | 0.10 |
| S | 1.45 | 0.03 | 0.03 |
| $Cl^-$ | 0.00 | 0.01 | 0.01 |
| Total | 100.5 | 93.7 | 99.2 |
| LOI | 1.99 | 5.94 | 0.28 |

The measured concentrations of the trace elements are presented in Table 8. The concentration of the class III elements As, Cd, Hg are clearly decreased and removed by the flue gasses. In contrary to these Cr, Ni and W showed an increased concentration, which can be explained by contamination, due to firing in the rotary kiln and/or sample preparation. The same phenomenon was found during sintering experiments with dredged sludge (Van der Gaag and Wevers, 2003).

*2.4.2. Mineralogical properties of raw materials and produced LWA*

The raw materials used in the production of LWA are analyzed on their mineral composition with X-ray Diffraction (see Fig. 3). The diffraction pattern of fly ash is typical for class F fly ashes. The fly ash consists of amorphous phases, quartz ($SiO_2$), mullite ($3Al_2O_3{\cdot}2SiO_2$ or $2Al_2O_3{\cdot}SiO_2$) and (added) portlandite ($Ca(OH)_2$). The dry clay contains quartz ($SiO_2$), some clay minerals and feldspar ($KAlSi_3O_8$–$NaAlSi_3O_8$–$CaAl_2Si_2O_8$).

The produced LWA contains quartz, feldspar, mullite and amorphous phases. Compared with the pattern of the fly ash, the main peak of quartz is of the same order of magnitude (600–700 counts), indicating that the quartz content has not really been changed. Apparently, sintering reactions between quartz and the alumino silicates in the LWA sintering process were not significant. The main peak of feldspar is strongly increased compared to the fly ash. The peaks of Portlandite have completely disappeared. Also no peaks of lime appeared anymore. The amount of free lime was also chemically analyzed. Free lime content was <0.1% m/m. This means that all free lime has been involved in the sintering reactions. The low free lime content implies that this will not cause unsoundness when used in concrete.

The results of the physical tests (see Table 9) show that the particle density of LWA is significant higher than Lytag. This may partially explain the lower water absorption, but the cause will also be the more sealed surface of LWA due to the use of a rotary kiln instead of a sinter strand. The compressive strength and E-modulus are somewhat lower than that of gravel. These properties are sufficient to produce good quality concrete.

*2.4.3. Leaching properties*

The leaching behaviour of the concrete is measured using the Dutch Tank test. The results are presented in the second column of Table 10. The requirements according the Dutch Decree on Soil Quality are given in the third column. These requirements apply for utilization of bound materials, like concrete, in civil engineering without any restriction. The leaching of some elements is below the detection limit, like As, Hg and Se. Unfortunately the detection

**Table 8**
Concentration of trace elements in fly ash (92% m/m), clay (8% m/m) and generated LWA, both measured and calculated.

| Element | Fly ash | Clay | LWA measured | LWA calculated | Deviation | Comments |
|---|---|---|---|---|---|---|
| As | 51 | 12 | 35 | 48 | −13 | – |
| B | 220 | 37 | 250 | 205 | 45 | – |
| Ba | 1700 | 360 | 1800 | 1593 | 207 | – |
| Br | <25 | <25 | <25 | 25 | – | Volatilized |
| Cd | 2.6 | 1 | 1.7 | 2 | −0.3 | Volatilized |
| Cl | <25 | <94 | <25 | 31 | – | Volatilized |
| Co | 19 | 15 | 24 | 19 | 5 | – |
| Cr | 71 | 65 | 110 | 71 | 39 | Contamination |
| Cu | 60 | 29 | 70 | 58 | 12 | – |
| F | 69 | 460 | 95 | 100 | −5 | – |
| Hg | 0.42 | 0 | 0.026 | 0 | 0.026 | Volatilized |
| Li | 100 | 38 | 110 | 95 | 15 | – |
| Mo | 19 | 2 | 23 | 18 | 5 | – |
| Ni | 62 | 30 | 98 | 59 | 39 | Contamination |
| Pb | 21 | 22 | 19 | 21 | −2 | – |
| Sb | 11 | 1 | 11 | 10 | 1 | – |
| Se | <0.3 | <0 | <0.3 | 0 | – | – |
| Sn | 2.7 | 5 | 2.2 | 3 | −0.8 | – |
| Sr | 820 | 61 | 870 | 759 | – | – |
| V | 180 | 82 | 210 | 172 | 38 | – |
| W | 2 | 3 | 21 | 2 | 19 | Contamination |
| Zn | 100 | 96 | 120 | 100 | 20 | – |

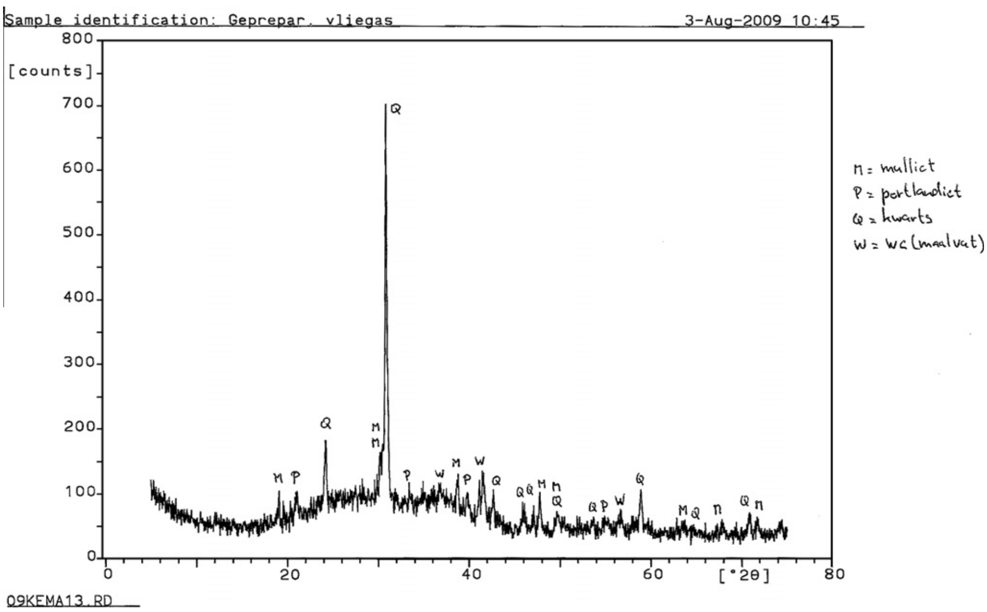


**Fig. 3.** X-ray diffraction pattern of produced LWA.

**Table 9**
Physical and chemical properties of LWA produced on laboratory scale and concrete made with LWA and gravel as reference material.

| Measurement/test | Unit | LWA | gravel |
|---|---|---|---|
| Loose bulk density | $kg/m^3$ | 945 | – |
| Particle density | $kg/m^3$ | 1630 | – |
| Water absorption 5 min | % m/m | 5.3 | – |
| Water absorption 30 min | % m/m | 6 | – |
| Water absorption 24 h | % m/m | 8.4 | – |
| Crushing resistance | $N/mm^2$ | 3.7* | 2650 |
| Density of concrete | $kg/m^3$ | 2060 | 2360 |
| Elastic modulus of concrete | $N/mm^2$ | 22200 | 24700 |
| Compressive strength of concrete | $N/mm^2$ | 31.1 | 33.7 |

* Average of 10 tests.

limit is above the limit value. Leaching of all elements is far below these limit values.

## 3. Evaluation

In the future, ash volumes will increase in the Netherlands due to realisation of new power plants. Because of the increase of ash volumes and the increase of co-combustion of secondary fuels, other applications of fly ash have to be assessed than the successful use in concrete and cement as (pozzolanic) filler. The best quality ashes will be delivered to concrete and cement industry. The remaining ashes will be used for alternative applications. In practice this means fly ash with high CaO contents, due to co-firing of woody fuels, and/or high LOI above 5% m/m. There is long term experience with the production of sintered light weight aggregate from fly ash (e.g. the Lytag process). Besides these fly ashes, also other ashes will be generated but on a much smaller scale. E.g. fluidized bed gasification fly ash, which has very high LOI can be used to adjust the LOI content of the green mix for LWA production.

**Table 10**
Leaching of LWA-concrete and requirements for bound materials for utilization without restrictions.

| Name | Availability (mg/kg) | E64d results from Dutch tank test (mg/$m^2$) | Requirement E64d (mg/$m^2$) |
|---|---|---|---|
| Al | 204 | 697.9 | – |
| As | 0.11 | <3.82 | 260 |
| B | 9.04 | <10.9 | – |
| Ba | 56.4 | 45.5 | 1500 |
| Br | 5 | 102.7 | 670 |
| Ca | 60757 | 26,842 | – |
| Cd | 0.005 | <0.14 | 3.8 |
| Cl | 120 | 115.3 | 110,000 |
| CN– | (Total) | – | 1.87 |
| Co | 12.9 | <0.31 | 60 |
| Cr | 1.11 | 1.34 | 120 |
| Cu | 2.75 | 1.28 | 98 |
| F | 75 | 40.2 | 2500 |
| Fe | 22.6 | 2.07 | – |
| Hg | 0 | <0.007 | 1.4 |
| K | 477.5 | 5122 | – |
| Li | 2.48 | 10 | – |
| Mg | 924.6 | 38.4 | – |
| Mn | 53.2 | <0.17 | – |
| Mo | 0.11 | <0.94 | 144 |
| Na | 198.4 | 1584 | – |
| Ni | 2.36 | <0.72 | 81 |
| P | 3.15 | <5.60 | – |
| Pb | 0.021 | <1.30 | 400 |
| Sb | 0.52 | <4.99 | 8.7 |
| Se | 0.68 | <6.25 | 4.8 |
| Si | 3575 | 3288 | – |
| Sn | 0.084 | <1.28 | 50 |
| SO4 | 4600 | 1212 | 165,000 |
| Sr | 269.5 | 465.8 | – |
| Ti | 0.003 | <0.074 | – |
| V | 0.42 | 2.13 | 320 |
| W | 13 | <1.42 | – |
| Zn | 29.3 | <1.25 | 800 |

The proposed production process is as much as possible based on proven technology. Most agglomeration steps in LWA production are granulation discs. Based on own experience different fly ashes can be granulated without any problem. As in the Trefoil process and most other processes a rotary kiln will be used. There is no quantified comparison between the performance of the sinter strand and the rotary kiln. However, if green pellets can be obtained with sufficient strength, it is more advantageous to use a rotary kiln. The green pellets will be dried before sintering with preheated air from the pellet cooler.

In this study, bench scale tests have been performed to produce LWA out of fly ash with high free lime content, simulating dry bottom fly ash with high percentages of co-combustion of wood. The produced LWA has good basic properties for use in concrete regarding water absorption and strength. The density of the produced LWA is 1630 kg/$m^3$, which is higher than e.g. Lytag (about 1400 kg/$m^3$). This means that less weight reduction of concrete can be obtained. The density of LWA can be influenced by the behaviour of unburned matter (fineness, volatiles) and the use of additives like carbonates, but this will also influence the strength of the LWA.

The applicable standards for LWA for use in concrete are the EN 13055-1 (1997) and NEN 3543 (2005). Most requirements for properties of LWA are maximum deviations from the 'declared values' by the producer. Fixed value is only given for acid soluble sulphate, namely maximum 1.0% (as $SO_3$). The measured value of the produced LWA is far below this value. The chloride content is <0.02% m/m, which is below the limit for pre-stressed and reinforced concrete according to the EN 206-1. Further, the chemistry and mineralogy of the LWA show that the product will not be unsound. The compressive strength and E-modulus of LWA concrete is somewhat lower than that of concrete with gravel, but this is still competitive in many cases. In general it can be concluded that the properties of LWA provides a good starting point for use in concrete.

The carbon foot print of 1 $m^3$ reinforced ready-mixed concrete in the Netherlands with an average composition (CEM III, C20/C25) is about 240 kg $CO_2$ equivalent (Cement en Beton, 2010), whereby aggregate is responsible for 15% of the carbon foot print, corresponding with 36 kg. Another source mentions about 230 kg $CO_2$ equivalent for concrete (Strukton, 2010). The replacement of coarse aggregate (in this case 442 l gravel per $m^3$ concrete) by LWA causes an increase of the carbon foot print with $321 * 0.442 - 36 = 106$ kg $CO_2$. However, the reduction of volume weight of the concrete causes reduction of transport emissions (less ton∗km), reduced use of concrete by reduction of the dead load and better isolation properties. Depending on the situation and the type of LWA these effects may counteract this increase of the carbon foot print of the concrete itself.

At this moment several types of LWA are accepted products on the Dutch market in different applications, like concrete, structural fill and horticulture. The focus of the proposed LWA will be on bound applications, as in non-bound applications requirements of the Decree on Soil Quality (Besluit Bodemkwaliteit) will not be met or only with restrictions and minimum quantities. The selling price for such applications is negative and therefore not interesting. This means in practice, the best option is as replacement of natural coarse aggregate in asphalt and concrete. The market volume for coarse aggregates in these applications is very high, namely about 15 Mton/year in relation to expected production volume of about 0.2 Mton/year. Such small volumes will not cause disturbance of the market if it is in competition with standard aggregates.

Own calculations of investments and operational costs confirmed the estimation of (Van der Gaag and Wevers, 2003). The expectations are that the operational costs, including sales and costs of area are about 45 euro/ton, corresponding with about 70 euro/$m^3$ as indication for the Dutch situation (some starting points: 10 years economical lifetime,7% profit and 6% interest). The selling price for natural aggregates is about 25 euro/$m^3$. This implies that, if the LWA is in competition with the standard aggregates, gate fee for fly ash will be negative, while use of certified fly ash for concrete has a positive gate fee.

## 4. Conclusions

Because of the expected increase of ash volume in the Netherlands, new utilization options were selected for fly ash from high co-combustion percentages. The option of the production of light weight aggregates has been assessed, based on a sintering process. It was shown that good quality of LWA can be produced with fly ash with high free lime content, which is the case if wood is co-fired. Also, leaching properties of concrete made with this LWA met the requirements of the Dutch Decree on Soil Quality. The carbon foot print of concrete will be negatively influenced if only the concrete itself is taken into account, but the reduction of the density has advantages regarding design, transport emissions and the better isolation properties, may counteract this. In the Dutch situation the operational costs are higher than expected potential selling price for the LWA, which implies that the gate fee for the fly ash is negative.

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