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Implementation of thermal processes for feedstock recycling of bromine and antimony, with energy recovery, from plastics waste of electrical and electronic equipment (WEEE)

Phase 1: Literature survey/status update, evaluation, and ranking of combustion, gasification, and pyrolysis based technologies

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#### Account

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# Keywords

European Brominated Flame retardants Industry Panel (EBFRIP), Waste of Electronical and Electronic Equipment (WEEE), Automotive Shredder Residue (ASR), Brominated Flame retardants (BFR), bromine, antimony, energy recovery, thermal processes, combustion, gasification, pyrolysis, Austrian Energy, ALSTOM TwinRec Ebara, Von Roll RCP, Gibros PEC, Sustainable Bromine Production.

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#### **SUMMARY**

The bromine industry wants to process plastics Waste from Electrical and Electronic Equipment (WEEE), containing brominated flame retardants (BFRs), to recycle bromine, antimony, and other valuable metals, with energy recovery for sustainable production and closing the bromine loop at the End-of-Life. The motivation to perform the underlying study was that insufficient detailed information was available regarding the technical and economical feasibility of processing of WEEE with thermal processes (*i.e.* combustion, gasification, and pyrolysis-based technologies). The objective of the study was to make an inventory of available processes and evaluate them based on the following six criteria:

- Energy efficiency,
- Bromine and Antimony recovery,
- Emissions (legislation permits),
- General quality of residues/secondary products,
- Operational experience, and
- Profit expectations (ROI).

The consumption of electrical and electronic equipment (EEE) within the European Union is expected to be 12.8 million tons in 2000. The total waste electrical electronic equipment (WEEE) is amounted to 4.4 millions tons in 1995. Approximately 273,000 tons of plastics used in EEE contained flame retardants (FRs), which represents 13% of the total plastics consumption (2.1 million tons in Europe).

In a mixed WEEE stream the plastics fraction accounts for 15 to 20%. This fraction contains some minor amounts of metals and other components due to the non-optimal separation. The composition of the plastics fraction of WEEE has comparable aspects as that of Automotive Shredder Residue (ASR), except for the high metal content. Therefore, in identifying processes for WEEE treatment, operational experiences with ASR give a good indication of the possible performance.

ECO-impact studies in the Netherlands and Germany have demonstrated that there is a limit of 15-18% of the plastics waste that can be mechanically recycled with environmental gain. Therefore, it is economically and environmentally more advantageous to use the plastics waste for energy recovery. With energy recovery a reduction is achieved in the use of fossil fuels and corresponding CO<sub>2</sub> emissions. Additional recycling of bromine and antimony from BFRs in the plastics, with the energy, recovery increases the percentage recycling.

The major applications of BFRs comprise brown goods, office equipment, and printed circuit boards. The element antimony (Sb) is generally added as a synergist to improve the flame retardant behaviour of EEE plastics. A (mass) ratio of 3-4 Br to 1 Sb is typical for flame retardant. The annual masses of bromine and antimony contained in the WEEE that could potentially be recovered are 9200 and 2100 tons, respectively.

The expected fate of bromine and antimony was discussed based available literature and knowledge. To quantify bromine and antimony distribution in a selected process experiments are necessary.

In thermal processes bromine (Br) will form radicals that will then generally react to form HBr and  $Br_2$ . The isolation of the bromine from the gas stream in a suitable form for re-use is based on absorption into an aqueous (hydroxide) solution.

In combustion the bromine distribution depends in general on the amount of bromine; an indicative distribution is in the grate ash (2%), fly ash (20%), and in the flue gas (78%). Under these conditions the distribution of antimony between the grate ash and the fly ash is approximately 2:1, accounting for ~95% of the mass. Under gasification conditions a similar partitioning of the bromine and the antimony is expected as for combustion conditions.

Eight processes were selected that are potentially suitable for the conversion of plastics WEEE, based on available literature, reports, information from the companies, and ECN knowledge from the market. *The information is updated to March 2000*. In the Table below the selected processes are summarised. Furthermore, six processes that are in development or on pilot-scale were briefly mentioned.

Company / Process	Technique	
Austrian Energy	combustion (& smelting)	
TPS	gasification & combustion	
ALSTOM/Ebara	gasification & combustion & smelting	
Siemens Schwel-Brenn	pyrolysis & combustion	
Von Roll RCP	pyrolysis & combustion & smelter	
Noell	pyrolysis & gasification	
Gibros PEC	pyrolysis & gasification & smelter	
Thermoselect	pyrolysis & gasification & smelter	
Processes in development or on pilot-scale.		

From the inventory based on the criteria followed that four processes are possibly suitable for treatment of WEEE: Austrian Energy, ALSTOM/Ebara, Von Roll RCP, and Gibros PEC. Other companies have indicated that they are not capable of processing WEEE (Thermoselect) or stopped their waste processing activities (Siemens, Noell).

The evaluation is based on the data collected in the report and an enquiry with a 'case' that was sent to the companies. In the discussion and evaluation the processes are ranked for each criterion mentioned above. The best process for each item is (are) ranked "1". Other processes are ranked 2 through 4, depending on the mutual differences (*i.e.* 'almost equal' *vs.* 'much better').

The ranking of the processes for each criterion are summarised in the Table below. In the last row of the Table the points are added up (the lowest total ranks 'best'). *N.B.* The added up totals are calculated without weighting factors.

In the discussion and evaluation of the processes care should be taken to keep in mind that nearly all experimental data of processing WEEE is available from the TAMARA studies that comprised (co-)processing of WEEE with household waste in a municipal waste combustion installation. Except for the Austrian Energy process, the thermal processes discussed in this report function according different principles (*e.g.* gasification and smelters). Therefore, the TAMARA results are not representative for these processes!

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Energy efficiency	3 / 2	2	2	1 / 2
Bromine recovery	4 / 1	3 / 1	1	1
Antimony recovery	1	1 / 2	2	2
Emissions	1	1	1	1
Quality of secondary products	4/3	2	2	1 / 2
Operational Experience	1 / 2	2 / 1	3 / 4	4/3
Profit expectations and ROI	1	2	3	2
Summarised	15 - 10	14 - 10	15 - 14	14 - 11

In this report thermal processes were evaluated for their suitability for the feedstock recycling of bromine and antimony, with energy recovery, from WEEE. The processes of Austrian Energy, ALSTOM/Ebara, Von Roll RCP, and Gibros PEC were evaluated in detail with the defined criteria. All four processes are in principle able to process WEEE and as no weighting factors were defined for the criteria no 'best' process was determined.

When weighting factors are added to the criteria, the evaluation and ranking is differentiated based on the following considerations:

- When the main purpose of the process is decided to be "best available technology", the Gibros processes would be the 'best' process.
- However, when "operational experience" together with low running costs is considered as the most important issue and given a high weighting factor, the Austrian Energy processes would rank the highest.
- If "proven technology" is the criterion for the decision than ALSTOM/Ebara will be the process of choice because of the plant in Japan that is running with ASR

As continuation, first has to be resolved with experimental tests if the bromine salts produced in the thermal processes are suitable as feedstock for the bromine industry. Furthermore, aiming the implementation of WEEE processing in 2004 co-operation with other industries and co-process the WEEE plastics with their wastes is essential.

The report was presented and discussed on the meetings of the European Brominated Flame retardant Industry Panel (EBFRIP) held on March 17, 2000 in Brussels, Belgium and of the Bromine Science and Environmental Forum (BSEF) meeting held on March 22, 2000 in Brussels.

#### 1. INTRODUCTION

# 1.1 Background

In a sustainable society the need for virgin materials and fossil fuels should be minimised. To realise this, waste must be optimally converted into energy and products. In case of waste from electrical and electronic equipment (WEEE), containing brominated flame retardants (BFRs), this means recycling of bromine, antimony, and other valuable metals, and recovery of the energy content. ECO-impact studies in the Netherlands and Germany have demonstrated that there is a limit to the amount of household plastics waste that can be mechanically recycled with environmental benefits. Further mechanical recycling requires relatively many efforts that annul the environmental gain. The maximal amount is 18% [1], or even as low as 15% according to a more recent study [2]. This means that the majority of the remaining waste must be recycled by other techniques. Combustion, gasification, and pyrolysis-based technologies have in principal the possibility to realise this.

The bromine industry needs to find accepted processing routes and have them implemented before the year 2004. As by the year 2004 there will be an EU ban on landfilling plastics of WEEE. They have concluded that for the realisation of this the above-mentioned thermal processes (combustion, gasification, and pyrolysis) are promising. Aiming at implementation in 2004 the following phased approach is necessary:

- Phase 1: Literature survey/status update, evaluation and ranking of available (existing on full-scale) combustion, gasification, and pyrolysis-based technologies.
- Phase 2: Feasibility study on three selected technologies. An economic assessment will be performed based on lab/bench-scale tests detailed mass and energy balance calculations and.
- Phase 3: Implementation scenario's. Existing facilities and implementation initiatives will be summarised and checked with respect to availability for WEEE, also in relation with optimal logistics. A new consortium will be formed in the case of insufficient treatment capacity in order to fulfil the implementation objective.
- Phase 4: Design and construction.
- Phase 5: Operation.

#### 1.2 Definition of the Issue

At this moment there is insufficient detailed information available regarding the technical and economical feasibility of processing of WEEE with <u>thermal processes</u>.

# 1.3 Objectives

The objective was to make an inventory of available combustion, gasification, and pyrolysis-based technologies and processes. Subsequently, a description of the operating experiences with these processes is made and feasibility for the recycling from plastics WEEE of bromine, antimony, and other metals, with energy recovery, is evaluated (Phase 1).

The criteria for evaluation are:

- Energy efficiency.
- Bromine recovery.
- Antimony recovery.
- Emissions (legislation permits).
- General quality of residues/secondary products.
- Operational experience.
- Profit expectations (ROI).

#### Restriction

The restriction is made that the presented results will be indicative as gasification and pyrolysis tests with a representative mixture of WEEE are not included in this phase. The accuracy of the presented results will depend on available data and experience of technology suppliers. ECN will evaluate these data based on a technology assessment and their experience with combustion, gasification, and pyrolysis. The presented results will be suitable to make a ranking and a first selection for further implementation phases.

# 1.4 Approach

The following activities were performed:

- 1. Literature survey/status update. Literature survey, in combination with interviews, were performed in order to update available information regarding available (existing on full-scale) combustion, gasification, and pyrolysis-based technologies. The mentioned criteria were quantified where necessary. The technologies were reviewed against these criteria.
- 2. Fate of bromine and antimony were discussed based on literature data. The resulting information were used for the evaluation of the in activity 1 described technologies.
- 3. Evaluation and ranking. Based on the results of activity 1 and 2 a final ranking were made and evaluated.
- 4. Reporting. All results and discussion were presented in a report. In the report advice for possible funding request on national and European level were given.
- 5. Meetings with EBFRIP. Two meetings with EBFRIP were held: one for discussion of the intermediate results and the second for discussion of the final results.

#### 1.5 Intended results

The result of the project is underlying report that describes the basic criteria for evaluation and ranking, the results of the literature survey/status update, the evaluation and ranking, and the conclusions of gasification and pyrolysis-based technologies for processing electronic scrap.

#### 2. WEEE CHARACTERISTICS

In this Chapter the origin and properties of the waste electrical and electronic equipment (WEEE) are briefly discussed.

#### 2.1 Volumes of WEEE

The consumption of electrical and electronic equipment (EEE) within the European Union has increased from 5.76 million tons per year in 1980 to 11.15 million tons in 1995 [3]. It is expected that this value will rise to 12.75 million by the year 2000, which relates to an increase of 121% over the twenty-year period. The total waste electrical electronic equipment (WEEE) is amounting to 4.4 millions tons in 1995, which represents approximately 40% of the total EEE consumption. The percentage of plastics used in the design of electrical equipment has also significantly increased. The contribution of plastics is anticipated to reach 20% (over 1,000,000 tons) by 2000 from a value of 15.3% (1995) [3].

According to a review on plastics consumption and recovery, published in 1995, approximately 273,000 tons of plastics used in EEE contained flame retardants (FRs). This represents 13% of the total plastics consumption which amounted to 2,129 millions tons in 1995 [3]. The type of flame retardant added to the polymer varies with the electronic or electrical product. The main FRs used in the plastics industry comprises brominated flame retardants (BFRs). Four sectors of the EEE represent almost 95% of the total consumption of (B)FRs: brown goods, data processing and office equipment, electrical equipment materials, and printed circuit boards [3].

# 2.2 Composition of WEEE

A detailed literature review of the composition of the above products has been carried out with the objective to derive a typical elemental composition for each sector [3]. Table 2.1 gives a typical material breakdown by category:

Table 2.1 WEEE Material Breakdown by Sector [3]

	<b>Brown Goods</b>	Data Processing & Office Equip.	White Goods	
	%	%	%	%
Plastics	26	13	7	30
Ferrous	35	40	44	
Non-Ferrous	26	30	32	30
Glass	4	5	4.5	30
Wood	1	1	1	
Other	8	11	11.5	10
Total	100	100	100	100

Based on the material (Table 2.1) and plastics (not shown) breakdowns a typical elemental composition was derived for each type of product:

Table 2.2 Typical WEEE Elemental Composition by Sector [3]

	Brown (	Goods	Data Pr	ocessing &	White (	Goods	Printed Ci	rcuit Boards	ASR
			Office E	Equipment					
Weight [%]	Plastics	Total	Plastics	Total	Plastics	Total	Plastics	Total	
Calorific value	*		*		*		*		~18 MJ/kg
C	79	20.6	65.8	8.6	46.4	3.2	27	8.1	37
O	3.4	1	7.7	1	20	1.4	30	9	13
Н	6.8	1.8	5.9	0.8	5.6	0.4	2.3	0.7	4.5
N	0.8	0.2	3.7	0.5	5.3	0.4	0.55	0.17	1.9
S	0	0	0	0	0	0	0	0	0.11
P	0.01	0	0.14	0.02	0	0	0	0	
Br	4.4	1	5.3	0.6	5.1	0.36	9	2.7	0.1
Cl	1	0.3	1.2	0.2	0	0	0	0	1.6
Sb	1.3	0.3	1.9	0.19	0.4	0.03	0	0	
В	0.02	0.01	0.14	0.02	0.6	0.04	1	0.3	0.03
Ferrous Metals	0	35	0	40	0	44	0	0	9.7
Non-Ferrous	1.3	26.4	4.2	31	14.2	33	30	40	2.0
Glass	0.7	4.8	1.6	4.6	2.2	4.6	0	30	
Others	0.35	8.6	2.2	11.6	0.2	11.5	0	10	30
Total	100	100	100	100	100	100	100	100	100

<sup>\*</sup> The calorific value of mixed WEEE plastics is approximately 22-24 MJ/kg.

In practice, the plastics fraction that need to be recycled is not free from metals and inerts, as suggested by the data in Table 2.1. In the subsequent steps in the (manual) recycle process of WEEE equipment, these constituents remain in the plastics fraction. A detailed analytical characterisation of the plastics waste has shown that the major portion of the heavy metals in WEEE is not due to the plastics, but comes from a poor level of metal separation [4, p.2]. The composition of the plastics fraction of WEEE is comparable to that of Automotive Shredder Residue (ASR). Therefore, in identifying processes for WEEE treatment, operational experiences with ASR give a good indication of the possible performance. For comparison, the composition of ASR is included in Table 2.2 [5].

# 2.3 Recycling of WEEE

In the Netherlands, currently Mirec and Coolrec recycle WEEE. In this process ~75% of WEEE is recovered to be used as recycled feedstock (see Figure 2.1). This fraction comprises glass, wood, and mainly the Ferro and non-Ferro metals. The non-Ferro metals are aluminium and copper and smaller amounts of the valuable noble-metals gold, silver, and palladium. In a mixed WEEE stream the plastics fraction accounts for ~20% and contains some minor amounts of metals and other components due to the non-optimal separation (Table 2.2).

ECO-impact studies in the Netherlands and Germany have demonstrated that there is a limit of 15-18% of the plastics waste that can be mechanically recycled with environmental gain [1,2]. It should be noted that 15% recycling of the 20% plastics in the WEEE only result in an additional 3% feedstock recycling. Therefore, it is economically and environmentally more advantageous to use the plastics waste for feedstock and energy recovery. With energy recovery a reduction is achieved in the use of fossil fuels. Additional recycling of bromine and antimony from BFRs in

the plastics, with the energy recovery increases the percentage recycling. Approximately 5% of the WEEE cannot be recycled or used for energy recovery and has to be disposed of.

The recycling of cars and white goods, as currently existing **in the Netherlands**, is comparable to the intended recycling of WEEE (compare in Figure 2.1). However, due to the different nature of the ASR a higher percentage mechanical feedstock recycling can be achieved.

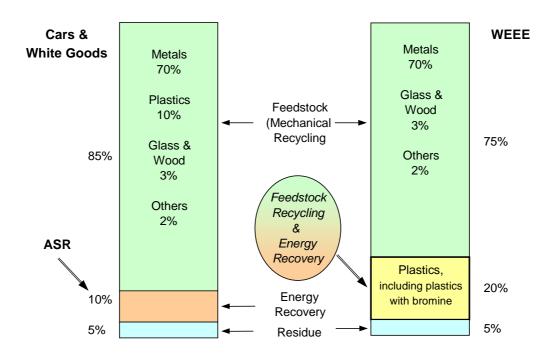


Figure 2.1 Breakdown of Cars & White Goods and WEEE into fractions for mechanical recycling, feedstock recycling, energy recovery, and a residue

# 2.4 Bromine and Antimony

As mentioned before, the major applications of BFRs in EEE comprise brown goods, office equipment, and printing circuit boards. The more frequently used polymer for brown goods is HIPS (High Impact Polystyrene) and this is generally associated with decabromodiphenyl ether (DECA). Housing of office equipment is generally made up of ABS associated with tetrabromobisphenolA (TBBA) and octabromodiphenyl ether (OCTA) or brominated epoxy oligomers (produced with TBBA). The element antimony (Sb) is generally added as a synergist to improve the flame retardant behaviour of EEE plastics. A (mass) relation of 3-4 to 1 for Br and Sb is typical for flame retardant. In the printed circuit boards industry, the dominant fire retardant used is TBBA. It should be noted that antimony is generally not used in printed circuit board applications when transparency is required. Table 2.3 gives a typical resin-FR breakdown for brominated plastics with the average bromine content by sector [3].

Based on the available information for 1995, the annual masses of bromine and antimony contained in the WEEE that could potentially be recovered can be estimated. The masses of bromine and antimony for the four sectors are 9200 and 2100 tons per year, respectively [3].

Table 2.3 Typical FR-Resin Breakdown by Sector [3]

Weight [%]	<b>Brown Goods</b>	Data Processing &	<b>Printed Circuit Boards</b>
		Office Equipment	
Resin (HIPS, ABS, Epoxy)	81.5	77.5	40
DECA	11.5		
TBBA		14	15
Antimony Trioxide	3.5	4.5	
Others	3.5	4	45
<b>Average Bromine Content</b>	8	8	10

#### 3. BROMINE AND ANTIMONY

In this Chapter the properties of bromine and antimony are discussed and their expected fate in thermal processes discussed.

# 3.1 Chemistry in thermal processes

#### 3.1.1 Bromine (and Chlorine)

Besides the bromine, also chlorine will be present in the plastics fraction of the WEEE. The chemistry of both elements in thermal processes is comparable and, therefore, they are discussed together. Unless otherwise stated all remarks about bromine are also valid for chlorine. Under combustion conditions, bromine (Br) will form radicals that will then generally react to form HBr and Br<sub>2</sub>, although the formation of other species such as metal bromides is possible [6]. For example, in case of gasification of PVC with calcium containing filler material, the HCl was nearly quantitatively (100%) captured by calcium, forming CaCl<sub>2</sub> [7].

The isolation of the bromine from the gas stream in a suitable form for re-use is based on absorption into an aqueous solution. This might be water to produce aqueous hydrobromic acid or an alkaline sodium or calcium (hydroxide) solution. Information from the hazardous waste combustion industry indicates that scrubbing into sodium hydroxide is the preferred method. Recovery of the bromine as either the hydrogen bromide or metal bromide will impact on the materials of construction for the absorption system and its configuration [3]. For bromine recovery, the temperature of the gas entering the absorption system will be relatively low.

#### 3.1.2 Antimony

In the combustion process two chemical forms of antimony (Sb) have been considered as potential products. In oxidative conditions the antimony remains in its trioxide form Sb<sub>2</sub>O<sub>3</sub>. The trioxide melts at 656°C and sublimates at 1550°C [8] [NB. In the PBKD report (ref. 3) it was incorrectly stated that SbO<sub>3</sub> sublimes at 650°C]. In a thermal process antimony may be found in either the grate or fly ash or in the gas phase depending on the operating conditions. To recover antimony from a flue gas additional procedures are required such as downstream high temperature filtration. In reductive conditions it will be released (volatilised) in the form of the halide salt SbBr<sub>3</sub> (due to the excess bromine). The antimony bromide boils at 280°C [8] and thus volatilises in the thermal processes. In contact with water or other products of thermal treatment (e.g. CO<sub>2</sub>) and at lower temperatures the SbBr<sub>3</sub> hydrolyses to the trioxide. Again, the antimony may then be found in the grate ash, fly ash, or in the vapour phase [3].

# 3.2 Fate of Bromine and Antimony

#### 3.2.1 Combustion of WEEE

APME has performed a study in which mixed plastics waste (MPW) is used as a single fuel to recover energy. Two techniques were evaluated: the CFB of Ahlstrom in Finland and the ICFB of Ebara in Japan [1]. From these experiments no data on the partitioning of bromine and antimony is available. From studies of co-combustion of WEEE with MSW in a (grate) combustion pilot-plant TAMARA at the Forschungszentrum Karlsruhe in Germany partitioning data is available [4,9].

The distribution depends in general on the amount of bromine (and chlorine, or more general halides) in the feed. An indicative distribution is: in the grate ash (2%), fly ash (20%), and in the flue gas (78%). The partitioning of the bromine (halides) between the fly ash and the gas is dependent on the geometry of the combustor, as this does influence the residence time. At higher residence times at high temperatures more HBr and HCl are 'captured' in the fly ash and in other conditions the fly ash content can be as small as a few percent [4].

In these experiments the distribution of antimony between the grate ash and the fly ash is approximately 2:1, accounting for ~95% of the mass. Up to 60% of the antimony mass was found in the fly ash in experiments with a large content of chloride demonstrating the well-known influence of increased Cl (or Br) levels upon volatilisation of antimony [4].

### 3.2.2 Gasification of WEEE

Under gasification conditions a similar partitioning of the bromine is expected as for combustion conditions. For antimony also comparable partitioning is expected. However, as a gasifier operates under reducing conditions (formation of  $SbBr_3$ , and mixed compounds with Cl:  $SbBr_2Cl$ ,  $SbBrCl_2$ , and  $SbCl_3$ ), the residence time might be not sufficient to convert all the antimony into  $Sb_2O_3$ . Furthermore, when metals are present that form stable and high boiling salts with bromine, the HBr and HCl are captured and remain in the ash (see also the next section).

#### 3.2.3 Limestone

When limestone (CaCO<sub>3</sub>) is used in a combustion or gasification process to capture the formed SO<sub>2</sub>, also (part of) the HBr and HCl are captured. The bromine-containing fraction of the process output is then a slag with parts of CaBr<sub>2</sub> (and CaBrCl and CaCl<sub>2</sub>) salt. In this form it will be less economical to regain the bromine.

## 3.2.4 Pyrolysis of WEEE

In pyrolysis experiments with printed circuit boards ~75% was found in the product gas and the remainder in the char [10]. For a large-scale facility, the process is preferably operated using a wet discharge system for the coke and with water as the quench medium. Using the wet discharge the most of the inorganically bound halogens are leached out of the pyrolysis coke. In this situation the bromide present in metal halide salts are recovered. The halogens in the gaseous phase (flue gas) are also accumulated in the aqueous phase as they dissolve in water of the quench and the scrubbers [11].

As the pyrolysis atmosphere is reducing, the antimony is present as SbBr<sub>3</sub> which volatilises at the (normal) operational temperatures (550-850°C). In contact with water or CO<sub>2</sub> at lowered temperatures, the SbBr<sub>3</sub> hydrolyses to the trioxide and can be recovered from the flue gas by filtration. Similar to the combustion process, part of the antimony will remain in the pyrolysis coke and part will be volatilised, the partitioning depending on the pyrolysis temperature. Mixed halide antimony compounds, which possibly are present, have a similar chemical behaviour as SbBr<sub>3</sub> (*e.g.* boiling point of SbCl<sub>3</sub> is 283°C [8]).

#### 3.2.5 Restriction

In the previous paragraphs the fate of bromine and antimony is evaluated based specific literature examples and general knowledge of the chemistry of the elements. To quantify the fate and distribution of bromine and antimony in a selected process, experiments are necessary. In this context, the presence of metals in the feed is especially important (see remarks in sections 3.1 and 3.2.3 [12,13]).

#### 4. THERMAL PROCESSES

The first step in the evaluation of thermal processes that are potentially suitable for the conversion of WEEE was a literature survey and a status update. In Table 4.1 the selected processes are summarised. The selection was made based on available literature, reports, information from the companies, and ECN knowledge from the market [14]. In the following sections these processes are discussed, based on their 'basic' configuration (as presented in brochures, etc.). Specific extensions to optimise the processes for WEEE/ASR treatment, when applicable, are discussed in Chapter 5.

Based on the evaluation four processes were selected as most suitable for WEEE treatment with respect to the defined criteria (see Chapter 5; Evaluation and ranking). Anticipating the evaluation, these four processes will be discussed in more detail in this Chapter. In Section 4.9 these processes will be briefly discussed, which are not operated on commercial scale or are in stage of development.

Standard combustion processes are not taken into account, as the existing ovens cannot handle 100% WEEE or ASR. Co-combustion of the streams is very well possible and this was studied in depth in the TAMARA studies [4,9].

 Table 4.1
 Overview of the discussed thermal processes

Company / Process	Location / Plant	Technique	Section		
Austrian Energy	Niedergösgen (CH)	combustion (& smelting)	4.1		
TPS	Grève-en-Chianti (I), Nyköping (S), Selby (UK)	gasification & combustion	4.2		
ALSTOM/Ebara	Sodegaura (JP), Fujisawa (JP)	gasification & combustion & smelting	4.3		
Siemens Schwel-Brenn	Goldshöfe (D), Ulm-Wiblingen (D), Fürth (D)	pyrolysis & combustion	4.4		
Von Roll RCP	Bremerhaven (D)	pyrolysis & combustion & smelter	4.5		
Noell	Lausitz (D), Northeim (D)	pyrolysis & gasification	4.6		
Gibros PEC	Freiberg (D), Aalen (D)	pyrolysis & gasification & smelter	4.7		
Thermoselect	Fondotoce (I), Karlsruhe (D)	pyrolysis & gasification & smelter	4.8		
Processes in development or on pilot-scale.					

# 4.1 Austrian Energy

In January 2000 Austrian Energy was in process of merging into the Babcock Borsig Power Group. Austrian Energy offers a selection of three processes in the fluidised bed firing sector: (1) internally circulated fluidised bed, (2) externally circulated fluidised bed, and (3) bubbling bed units. The internally circulating fluidised (or *Turbo fluidised*) beds are utilised for outputs of up to approximately 100 MW<sub>th</sub>. These are mostly waste residues with problematic characteristics and calorific values of around 5-35 MJ/kg (*e.g.* shredder waste, used tyres, and municipal waste fractions) [15].

#### 4.1.1 Process description

Basically, in the Fast Internally Circulating Beds (FICB) combustion process a major amount of bed material is circulated (see Figure 1 in Appendix A for a schematic process overview). The core of the process is the fast-fluidised bed in which most of the combustion takes place. The height of this fluidised bed combustion chamber is limited to a few meters. This results in a particle loading at the outlet of the chamber that is substantially higher than in the case of conventional circulation fluidised beds. The particles are removed from the flue gas by a baffle-type separator located at the outlet of the chamber. The solids precipitate on the inclined wall in the fuel feed zone and slide into the 'slow bed' that serves as a storage for bed material. The slow bed is connected to the fast fluidisation section by a gap of defined geometry. The flue gas passing the particle separator enters at high velocity the post-combustion chamber located above the feeding zone. A freeboard burner ensures that the required temperature of minimal 850°C is maintained [16], however, in the case of waste treatment a minimal temperature of at least 1100°C can be employed [17]. To clean the flue gases fabric filters or electrostatic precipitators can be employed. Depending on other factors (*e.g.* legislation, types of feed) additional flue gas cleaning systems may be employed.

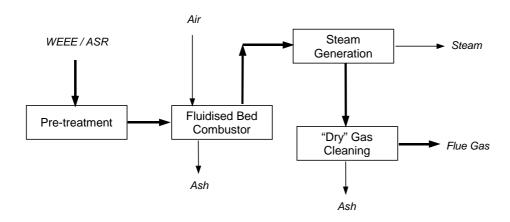


Figure 4.1 Schematic Representation of the Austrian Energy Process

#### 4.1.2 Operational experience

In 1991 Austrian Energy commissioned a Fluidised-bed-Boiler to an Austrian company producing fibre panels and chipboards. Although this was the first industrial application of FICB-boiler, the technological expectations were exceeded [17]. Municipalities and private companies established the Regionale Ensorgungsanlage Nierdegösgen AG (RENI) at the beginning of 1994 in order to dispose the production wastes and sludges from regional sewage treatment plants and the neighbouring cardboard mill. The saturated steam generated by the combustion process is used as process steam in the cardboard mill. The (FICB) plant started commercial operation in September 1995. Due to good operational results, in 1996 an application for an increase in plant capacity by 20% was made [16].

In 1991-1993 Austrian Energy has done a lot of test work on the energy recovery from ASR. In these years Austrian Energy has performed about 1000 operational hours at the testrig and they claim to have now a lot of operational experience with this material (possible issues around fuel sizing, fuel feeding, ash sintering, and combustion behaviour). A correct design of the fuel feeding system to avoid plugging and a proper combustion temperature control to avoid sintering were main results of these tests. Further focal points of their investigations were ash

amounts, ash distribution, ash behaviour and ash composition as well as flue gas analysis, ash behaviour on the deposit, ash treatment and flue gas treatment [18].

After having reached stable operating conditions in the process Austrian Energy started to design the industrial units. The results of the tests gave all necessary data and know-how to design and build such units with capacities of about 30,000 to 40,000 tons of ASR per year. Considering the experience with their latest and largest FICB unit, which started up last year, Austrian Energy is now able to burn up to 80,000 tons per year in one furnace. Unfortunately both projects have been cancelled due to legal reasons (not emission related) and financial reasons, respectively [18].

#### 4.1.3 Emission aspects

The  $SO_x$  formed by the combustion of sulphurous fuels is retained in the fluidised bed by the addition of limestone. The HCl produced by burning chlorine-containing fuels is also retained in the ashes with good efficiency. Fuels with high nitrogen content have potential for forming nitrogen oxides. In FICB plants ammonia or urea can be injected in the post-combustion chamber for the selective non-catalytic reduction (SNCR) of these nitrogen oxides [16]. Due to the presence of a de-NO<sub>x</sub> facility, possible present traces of dioxins and furans (DIF) are also converted. Austrian Energy collected much knowledge about bromine recovery and materials selection for the flue gas cleaning systems from their experience with their hazardous waste installation.

#### 4.2 TPS

TPS (Terminska Processor) is an offshoot of Studsvik, a Swedish public sector company. Between 1991 and 1995 TPS has focused on process development for small to medium scale electricity production plants using biomass and RDF as feedstocks. The process is a fluidised gasification process with direct combustion of the syngas to produce electrical power [19].

#### 4.2.1 Process description

The TPS gasifier comprises a bubbling fluidised bed (BFB) into which pre-heated air and shredded MSW or RDF pellets are fed. The BFB operates at about atmospheric pressure. Solid feed material is distributed across a lower 'dense bed' and volatilises. The temperature in this zone is approximately 700-800°C. Steam can be added besides the primary air to facilitate gasification. Secondary air is injected above the dense bed zone and the temperature rises to about 850-900°C. Gas and elutriated solids leaving the BFB pass to a hot cyclone where the solids are removed and returned to the bed. The syngas then passes to a CFB containing dolomite which 'cracks' the tars and higher hydrocarbons. The hot gas is cleaned and combusted in a boiler to raise steam or in a gas engine or turbine to generate electricity [19].

#### 4.2.2 Operational experience

TPS is involved in three significant commercial demonstration BIGCC projects. A twin CFB gasification plant (15 MW each) was constructed in Grève-en-Chianti in Italy and commissioned in 1992 for RDF. During 1995-96 the plant has predominantly processed biomass. In late 1994, a joint venture company, Arbre Energy Limited, was set up for the construction of a 8 MW<sub>e</sub> BIGCC plant in Selby in the United Kingdom. For the construction of

this plant Arbre received financial support from the EU. The plant also became the recipient of an UK Non Fossil Fuel Obligation (NFFO) contract that provides a guarantied preferential price for a period of 15 years. The plant is due for start-up later in 2000 and shall use short rotation coppice as feed.

TPS was contracted for the realisation of a CFB gasifier with combined cycle power generation (30 MW<sub>e</sub>) in Brazil for eucalyptus feed. Plant start up is expected in the year 2003, depending on financial support from the World Bank and United Nations Programme's [20].

TPS operates a 2 MW<sub>th</sub> pilot plant in Nyköping (Sweden). This pilot plant is used for testing only. Many different feedstocks have been tested, including different types of industrial and domestic wastes. TPS has not performed any tests with ASR or WEEE. TPS is reluctant to give a definitive answer as to whether the TPS process is suitable for ASR, without the analysis of the composition of ASR. Important items in such evaluation would be items like the ash and chloride content. Furthermore, tests at pilot scale would probably be necessary if TPS were to provide performance guarantees [21].

### 4.3 ALSTOM/Ebara

The TwinRec process is designed and built by ALSTOM Power (Switzerland) Ltd (former ALSTOM) under the license of the Japanese Ebara (Ebara's brand name TIFG). The core components of TwinRec are the fluidised bed gasifier and the cyclonic combustion chamber (see Figure 2 in Appendix A for a schematic process overview). TwinRec has officially been approved for commercial operation by the Japanese Authorities, based on long time continuous operation of a 1 ton/h pilot plant [22,23].

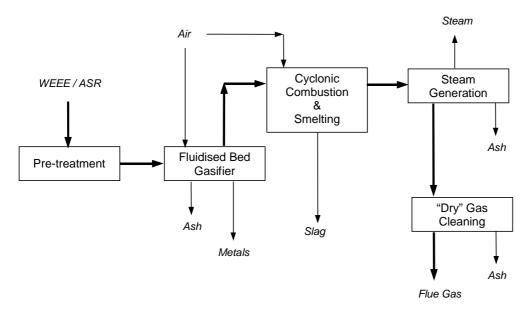


Figure 4.2 Schematic Representation of the ALSTOM TwinRec Process

#### 4.3.1 Process description

The feed (waste) is gasified in an air blown internally circulating fluidised bed at temperatures between 500-600°C. The high specific thermal load enables a small gasifier cross-section. The combustible portion of the waste is gasified as well as partly suspended as particles by the produced fuel gas. The low gasification temperature in the fluidised bed leads to slow, easily

controllable process conditions. Metals like aluminium, copper, and iron can be recycled as valuable products as they are neither oxidised nor sintered with other ash components [22].

Produced fuel gas and carbonaceous particles are burned together in the cyclonic combustion chamber at temperatures between 1350-1400°C by the addition of secondary air. Melting of fly ash is achieved without auxiliary fuel and the molten slag is quenched with water and obtained as granulate [22]. The technology produces a reduced mass-flow of the flue gas, allowing for a reduction in the size of the steam boiler and air pollution control units, located after the combustor. The energy content of the waste is converted into electricity and/or district heating with high net efficiency, via steam generation [22].

#### 4.3.2 Operational experience

Ebara operated a 1.5 MW<sub>th</sub> pilot plant Sodeguara (Japan) from 1995 to 1997 and has a 1 ton/h (4.8 MW<sub>th</sub>) pilot plant in operation in Fujisawa (Japan). In both plants tests with ASR were performed. Four commercial plants are currently under construction, all scheduled to be commissioned in 2000. The largest plant has a planned waste capacity of two times 10 ton/h (twice 40 MW) and has been started-up in the beginning of 2000 [24].

#### 4.3.3 Emission aspects

The standard slag granulate meets all common leachability regulations. The high combustion temperature, in combination with the gas cleaning (*i.e.* de-NO<sub>x</sub>), enables the fulfilment of most stringent dioxin emissions regulations up to 0.1 ng TEQ/Nm<sup>3</sup> with minimal additional measures. From a 1000 kg feed mixture ASR and sewage sludge (ratio 7:3), 47 kg of boiler ash and Air Pollution Control residue have to be landfilled [22]. It was expected that, when processing WEEE, the dry gas cleaning with ASR would not fulfil the HBr, HCl, and Br<sub>2</sub> emissions regulations [25]. At installations in Europe they will install wet scrubbing systems to completely fulfil the emission regulations.

#### 4.4 Siemens Schwel-Brenn

The Schwel-Brenn process is a combination of pyrolysis and combustion. The pyrolysis step is used to homogenise the fuel and for separation with sieves of the major part of the mineral and metallic fraction. In the combustion step energy is generated [14].

#### 4.4.1 Process description

The process is build up from units with established technologies. The pyrolysis is carried out in an externally heated rotary kiln to which the fuel is fed with a conical screw. The pyrolysis char is sieved and the smallest char particles are powdered. The pyrolysis gases and the powdered char are combusted in a vertical combustion reactor, comparable to those commonly used in coal combustion installations. The cleaning of the gases is comparable to the technique used for combustion plants. The hot product gases are used to produce steam in heat exchangers and the steam is used to generate electricity with a turbine in a steam bottoming cycle [14].

#### 4.4.2 Operational experience

The basis for the Siemens Schwel-Brenn process is the Kiener pyrolyseproces (KPA). In 1984 Siemens took part in KPA and since then they have tried to commercialise the process. To avoid the difficult step of cleaning of the pyrolysis gas, the combination of pyrolysis and combustion was chosen. This concept was tested in an installation in Ulm-Wiblingen and, subsequently, larger scale tests were performed in an installation in Goldshöfe. At the end of 1997 three commercial installations were being constructed with capacities between 100,000 and 150,000 ton waste per year (Germany). The first commercial installation in Fürth was in operation from 1997. This installation exists from two lines with each a combustion reactor and a gas cleaning systems and each two pyrolysis kilns with a capacity of 5 ton/h [14]. Currently, the installation is stopped and withdrawn from the contract.

A large number of possible fuels were tested in several test installations for the process. The process was designed for domestic waste and similar waste streams. To domestic waste 30-35% (dry basis) sewage sludge can be added without significant problems [26]. In the installation in Ulm-Wiblingen tests with automotive shredder residue (ASR) have been performed. Besides some minor problems these tests were successful, showing that the Schwel-Brenn process is very suitable for the thermal treatment of shredder waste (based on their own judgement) [27,28].

In contacts with Siemens they reported that they had stopped all thermal waste treatment activities. The reason was that the process is not economically feasible [28]. Therefore, the *Siemens Schwel-Brenn process cannot be used for the treatment of WEEE*.

### 4.5 Von Roll RCP

# 4.5.1 Process description

The Recycling Clean Product (RCP) process employs a forward reciprocating grate furnace as a pyrolysis (degassing) chamber where fuel is heated to convert it into pyrolysis gas and cokes. The energy required for this stage is released by partial combustion of the gas with oxygen. Pyrolysis takes place at a bed temperature of about 500°C. The gas leaves the chamber at 900°C. The pyrolysis coke flows into a smelting furnace where oxygen injection creates the high temperatures (1400°) necessary to smelt all solid material. The use of oxygen reduces the volume of gases formed (compared to air). Heavy metals are concentrated and separated from the molten slag. The residual slag is pelletised and can be used as a cement additive or construction material. The hot gases from the smelting furnace and slag treatment (smelter) are combusted in a circulating fluidised bed (CFB) boiler at a temperature well below 1000°C. Oxygen is again added to ensure complete oxidation of organic compounds. The flue gas and the circulating sand are separated in a hot cyclone. The gases are used to generate steam for electricity and heat [19,29].

### 4.5.2 Operational experience

The process is a combination of two processes (pyrolysis and smelting) brought together by Von Roll using their own in-house grate technology and a bought-in smelting technology. The Von Roll process was developed for improved recycling of non-combustible fractions of waste by thermal treatment under total smelting conditions. The first RCP plant for the combustion of MSW has started operation in 1997 at Bremerhaven, Germany, and it has been further

optimised in 1999. During four operation periods between June 1997 and October 1998 the plant was for about 1000 hours operating under melting conditions. It performance reached up to 75% of its nominal thermal capacity (17.5 MW in 24 h shift operation) [30].

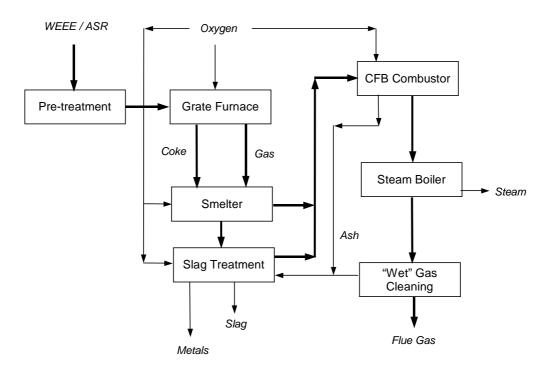


Figure 4.3 Schematic Representation of the Von Roll RCP Process

Only a test with 100 tons of German ASR was performed in January 1999. It worked well and very smoothly, although higher dust loads as expected were experienced. A later test with 400 tons Swiss ASR was abandoned due to operational equipment problems. Von Roll has been several months online last year (especially from September to November 1999), reaching a constant throughput of 50% nominal capacity and higher loads over several hours. However, there were still problems left causing interruptions. Currently, Von Roll is modifying their plant again (smelting oven: simpler design with improved cooling) [31,32].

Von Roll expects to go on stream with waste in March 2000 and with ASR in April 2000. They state: "We underestimated the necessary development time for such a system". However, they still believe that the basic design data is correct and that RCP is a suitable solution for ASR and similar waste materials as well as MSW [31].

# 4.6 Noell

#### 4.6.1 Process description

The Noell Conversion Technology ('Konversionsverfahren') is a combination of pyrolysis and gasification and based on an entrained flow ('Flugstrom') gasifier. The gasifier requires a fuel that exists of small solid or liquid particles. Therefore, the products of the preceding pyrolysis step have to be condensed (gases, tars, and dust) or grinned (char). The pyrolysis pre-treatment was developed to make the process suitable for inhomogeneous wastes and wastes that are difficult to grind [14].

The entrained flow reactor consists of a vertical steal tube with a water-cooled ceramic inner tube. Gasification of the particles is performed with oxygen at 2-50 bar pressure and 1400-1700°C. On the inner wall of the tube a layer is built up of solidified slag and over this layer the liquid slag streams downwards. These liquid slags pass constriction in the inner tube and drop through a water-cooled second chamber into a water bath [33]. The advantage of this reactor design is that the layer of solidified slag prevents damaging of the reactor wall by the aggressive molten slag. The lifespan of the reactor can be longer (up to ten years) than of other reactor types (sometimes only one year).

An important parameter for this process is the melting temperature and viscosity of the slag, as the slag smelt flowing down the reactor wall should be low viscous. Domestic waste usually produces a low viscous slag whereas sewage sludge (especially when stabilised with CaO) affords a slag that smelts at high temperatures. The smelting temperature of a slag can be lowered by addition of SiO<sub>2</sub>. To minimise the smelt temperature a CaO/SiO<sub>2</sub> ratio of 0.7-1.1 is aimed at [33].

## 4.6.2 Operational experience

The basis for the Noell Conversion technology ('Konversionsverfahren') is the pressurised entrained flow ('Flugstrom') gasification, which was developed in the mid 1970s. The process was initially developed for high calorific and salt-rich fuels like brown coal and coal. However, with the preceding pyrolysis step the process can also be used for domestic waste [33,34,35]. At the end of the 1970's a 3 MW demonstration installation was built and in 1982-1984 the 130 MW 'Schwarze Pumpe' were constructed in Lausitz (near Cottbus, Germany). Initially with one, and later with two, Flugstrom reactors the installation was tested from 1984-88 and taken into operation in 1988. Since 1991 the Noell technology has been developed further for the large-scale conversion of wastes, resulting in the introduction of the preceding pyrolysis step [36,37]. In 1996 a 10,000 ton/y (16 MW) installation was constructed in Northeim for the conversion of domestic waste and sewage sludge [14].

At this moment we have no information about the operational experiences of the plant in Northeim. So far Noell has not replied to repeated requests for more information [38]. At a seminar on pyrolysis and gasification (Freiberg, March 2000) the Noell representative indicated that Noell has stopped all integrated waste treatment activities [39]. Therefore, *their process is not further evaluated*.

### 4.7 Gibros PEC

The pyrolysis technology of the Gibros PEC process is developed by Pyrolyse Kraftanlage (PKA) GmbH. Gibros PEC is shareholder of PKA and world-wide licence-holder of the technology. An other partner of Gibros PEC is Metalgiesserei ("Foundry") Velmede GmbH, which is the owner and developer of the smelt technology that is an essential part of the Gibros PEC process. The complete process, including pyrolysis, gas treatment, and smelt technology, is covered by more than 40 patents [14].

# 4.7.1 Process description

The process is a combination of pyrolysis, gasification (thermal cracking), and smelting (Figure 4.4). The pyrolysis takes place in a rotating kiln (500°C; residence time of approximately one hour) that is externally heated with part of the gas produced in the process.

The pyrolysis gases containing tars are subsequently gasified (cracked) in a high temperature (1200-1300°C) gasifier by the addition of oxygen [14].

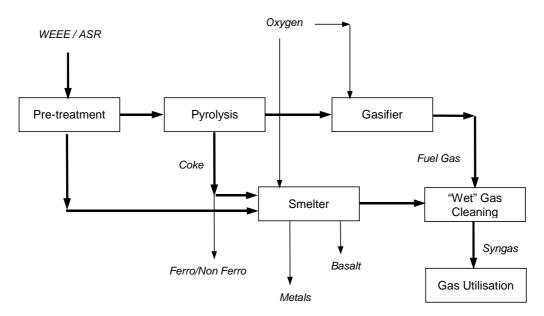


Figure 4.4 Schematic Representation of the Gibros PEC Process

Larger pieces of Ferro and non-Ferro metals are removed from the solid product from the kiln. The remaining solids (char) are smelted (at 1400-1500°C). The heat is generated by partial oxidation of the char with oxygen (under reducing conditions). In the smelter a layer can be formed of molten metals under the molten slag, allowing separation of both fractions. The offgas is combined with the gas from the gasifier. The mineral slag can be used as construction material (synthetic basalt) [14]. Due to the high temperatures in the gasifier and the smelter no organic compounds are present in the syngas.

The syngas contains sulphur and nitrogen compounds like  $H_2S$ ,  $CS_2$ , COS, and HCN ( $NH_3$  is decomposed at the high temperatures). In a combustion step these compounds would be converted to  $SO_x$  and  $NO_x$ . This means that for the syngas different gas cleaning has to be utilised than for a flue gas. Compared to combustion, cleaning of the syngas is advantageous, as the gas volume is much smaller.

Gas cleaning is performed prior to the gas utilisation and it exists of a washer to remove HCl, HBr, HCN, NH<sub>3</sub> (if present) and dust, followed by a bag filter to remove the remaining dust, and a basic and a biological washer to remove H<sub>2</sub>S. Finally, an active coal filter to absorb dioxins, furans, and Hg (Hg concentration is reduced from ppm to ppb levels) [14]. The syngas is used to generate electricity (and heat) in a gas engine. Part of the gas is needed to provide the heat for the pyrolysis and the production of oxygen on site will consume part of the electricity.

#### 4.7.2 Operational experience

In 1983 a pyrolysis test installation was build with a capacity of 10,000 t/y in Aalen-Unterkochen (D) and different wastes were tested. The installation was developed further in the late 1980s and moved to Freiberg (D) in 1993. In this installation up to fifteen waste streams were tested (like shredder residue and aluminium-containing package waste). Based on those experiences a 30,000 t/y installation was built to treat aluminium-containing package waste and to recover the aluminium [40,41]. In 1998 the first commercial installation was constructed in

Aalen (South Germany) for the treatment of the 'Fluff'-fraction of Domestic Waste. The capacity of this installation is 25,000 ton/y. The product gas will is in gas engines [42]. In 1999 a test with Dutch ASR was performed in Freiberg, but the results are not yet public available. All steps in the process are proven technology, however, the combination and integration is still in pilot stage.

#### 4.7.3 Emission aspects

Gibros PEC claims that their process is almost zero-emission (less than 0.5%): all residual streams (product gas, metals, and minerals) can be used, either to be sold or to produce heat and electricity [14].

#### 4.8 Thermoselect

#### 4.8.1 Process description

The process is a combination of pyrolysis and gasification. With a screw waste is fed into a degasification 'channel' where it is heated from 50 to 600°C and pyrolyses. From the channel the solids drop into a high-temperature gasification reactor. The bottom of the reactor is connected with a smelt oven. The smelter is operated with oxygen and the high temperature of the smelt (1600-2000°C) is necessary to complete all chemical reactions. The smelt is cooled quickly to keep the metals and minerals separated (the metals congeal quicker than the mineral compounds) and the metals are separated with a magnet separator. The pyrolysis/gasification gases are cooled with a water quench and subsequently cleaned [43,44].

#### 4.8.2 Operational experience

The development of the Thermoselect process started in 1989 with a small test installation (10 kg/h). The technology was developed further in 1990 and 1991 with several other test installations (250-1000 kg/h) and a demo installation (4200 kg/h) or 30,000 ton/y). In 1991 the construction started of a installation in Fondotoce (Italy), which was taken into use in 1992 [45]. With this installation in Fondotoce experience was gained and in 1998 a commercial installation of 225,000 t/y was constructed in Karlsruhe [46]. The mentioned capacities are valid for fuel (waste) with a heating value of 10-12 MJ/kg, with higher heating values the capacities of the installations decrease [14].

At this moment we have no information about the operational experiences of the plant in Karlsruhe. No attempts were made to collect information as <u>Thermoselect had indicated to EBFRIP that their process in Karlsruhe is not suitable for treatment of WEEE</u> [47].

#### 4.8.3 Emission aspects

In the Thermoselect process the gasses are quenched to prevent formation of organic compounds like dioxins and dibenzofurans. Comparison of the measured gas compositions with thermodynamic equilibria showed that this concept works in practice [43]. The measured concentrations of  $CH_4$  and dioxins are low  $(0.002 \text{ ng/Nm}^3)$  compared to the emission limit of  $0.1 \text{ ng/Nm}^3)$  and in agreement with the thermodynamics. In several descriptions of the Thermoselect process [43,44,46,48] the very low PCDD and PCDF emissions of the process are

emphasised. They claim that per kilogram waste approximately 500 times less dioxins are emitted than present in the waste [44], and 800 times less than a German waste combustion plant [46]. The quality of the syngas is comparable with natural gas [43,46].

# 4.9 Processes in development or on pilot-scale

In this section processes are mentioned and briefly discussed that do not operate on commercial scale or are in stage of development (in alphabetical order). The thermal processes of Lurgi and Pyrovac were brought in at the end of the project and, therefore, not evaluated.

# 4.9.1 Aarding Energo

**Process.** In a furnace the feed waste is gasified in two stages at temperatures between 650-900°C. The residence time in the furnace is constant and the temperatures are carefully controlled. Ash is removed at the end of the furnace. After the gasification the flue gases are combusted and cleaned. Together with sludge the ash is fed to a smelter and smelted at 1400°C. The gases that arise from the smelt are cooled with air in a secondary reactor in which all the residual products are combusted. The solids residue from the smelter is a granulate [49].

**Experience**. Currently, the Aarding Energo installation in Amsterdam is tested without smelter. For February 2000 trials are scheduled for the treatment of ASR for Auto Recycling Nederland (ARN).

#### 4.9.2 Battelle

**Process description**. Battelle Memorial Institute in Columbus (USA) operates a commercial installation in Vermont (USA). The Battelle reactor exists from two circulating fluidised bed (CFB) risers. The feed enters the first circulating bed that is heated with sand from the second fluidised bed. The CFB is blown with steam as fluidisation gas. The temperature in the gasifier is typically in the range of 800-850 °C. At these temperatures the feed is partly pyrolysed and gasified with the steam. At the top of the riser the gas is separated from the solids (sand and char) and the solids are fed into the bottom of the second CFB. This riser is blown with air to combust all the char. In this way the sand is burned 'clean' and its is heated up to maintain the endothermic pyrolysis and gasification reactions [50].

**Operational experience**. In a larger test unit (250-500 ton/h) mainly refuse derived fuel (RDF) and wood were tested (1993) [50]. In a 10 kg/h pilot-scale reactor experiments has been performed with mixed plastics waste (MPW), including the chlorine containing PVC. The released chlorine was captured as HCl. Akzo-Nobel [as member of the European Council of Vinyl Manufactures (ECVM)] has considered processing PVC with the Battelle process (the Linde process was the other option; see section 4.9.4).

In the pilot reactor a 10 h test with PVC containing plastics mixture was performed. The reactor was 'old' and due to operational problems the test had to be stopped frequently. HCl was released in good yield from the feed and no dioxins were formed but problems due to significant soot formation were encountered (expected). As the test periods were short no information was obtained on the effect of the inorganic additives on the process (*e.g.* agglomeration). Akzo-Nobel has stopped all activities on HCl recycling from PVC. In the opinion of Akzo-Nobel the Battelle process seems to be suitable for HCl recycling from PVC. However, due to the lack of experimental information on pilot-scale from a prolonged test-period this cannot be reasoned [51].

### 4.9.3 BP Grangemouth

BP Chemicals operates a pilot installation for pyrolysis in low pressure fluidised bed in Grangemouth (United Kingdom). In this large pilot plant they have not carried out tests with ASR, but limited tests have been carried out in a smaller fluidised bed. These tests were essentially successful to assess process chemistry. To determine if the BP process is suitable for ASR treatment on industrial scale they need to characterise the plastics. Furthermore, as the process is developed to process mixtures of packaging plastic, the differences in terms of engineering aspects for ASR should be fully explored [52].

#### 4.9.4 Linde smelt-bath

Linde has designed (although not constructed and put into operation) a process based on gasification in a smelt-bath. The temperature in the reactor is kept above the melting temperature of the ash to create a liquid slag. The slag is kept in turbulent motion to ensure good transfer of heat and oxygen (from air) [53]. Akzo-Nobel (as member of the ECVM) has considered processing PVC with the Linde smelt-bath (the Battelle process was the other option; see section 4.9.2). The process was developed specially for the recycling of the HCl from PVC. Currently, the project is stopped [38].

#### 4.9.5 RESHMENT

RESHMENT is a process for maximised recycling, reuse and recovery of ASR and developed by CT Environmental (Umwelttechnik) in Winterthur [54]. CT Environmental is a subsidiary of Austrian Energy (AE) and is also very active in the field of various recycling processes. Furthermore, they are involved in AE activities concerning recycling of WEEE [55].

**Process description**. The first step is a pre-treatment of ASR in order to separate ferrous scrap, copper, and aluminium. In the second stage ASR and fly ash are mixed together and treated in a melting cyclone. There the organics decompose, the mineral fraction is melted, and the low melting metals are volatised. Downstream of the melting cyclone the melt is separated from the gas and can be vitrified by shock cooling, while the gasses are completely combusted [56].

**Status**. At present Austrian Energy is going to favour the fluidised bed technology for recycling of WEEE. As AE has many references with fluidised bed incinerators on industrial scale (for different other fuel materials) and AE believes that it will be easier to adapt their process to ASR or WEEE than to upscale RESHMENT [55].

#### 4.9.6 Texaco

Texaco constructs high-temperature gasifiers for coal and refinery residues. Solid waste materials are ground with water to produce a slurry with a solid concentration > 60%. The water serves as a transport medium, temperature moderator, and moderator as it promotes the formation of hydrogen within the process. The slurry is injected with air or oxygen into a burner at the top of the gasifier (cf. Noell; section 4.6). The reactor is refractory lined pressure vessel that operates at a pressure of around 24 bar and above the ash melting temperature of the waste material (typically > 1315°C). The process affords a syngas while a molten slag exits the bottom of the gasifier into the quench chamber for cooling and metals present are encapsulated in the

cooled slag [19]. Texaco has no experience with co-gasification of plastics and in general their process can only handle feeds with up to six percent inerts [57].

**Status**. Texaco has also designed an installation especially for the treatment of mixed plastics waste. The performance of the designed installation was tested on parts in the Texaco pilotinstallation in California. First, making the plastics fluid and pumpable and secondly, the feeding of the slurry to the gasifier. These tests were successful. The planned location of the installation was Rotterdam (The Netherlands), however, they stopped the project as they encountered problems in contracting the plastics for treatment [57].

# 5. EVALUATION AND RANKING

In Chapter 4 the thermal conversion processes that are possibly suitable for treatment of WEEE are discussed. Some of the companies that operate these processes have already indicated to EBFRIP that they are not capable of processing WEEE (Thermoselect). Other companies have stopped their waste processing activities (Siemens, Noell). In Table 5.1 an overview is given of the remaining processes. *The report is based on available data up to February/March 2000. Information received in a later stage is added in notes.* 

Table 5.1 Overview of the remaining processes

Company / Process	Technique
Austrian Energy	combustion (& smelting)
ALSTOM/Ebara	gasification & combustion & smelting
Von Roll RCP	pyrolysis & combustion & smelter
Gibros PEC	pyrolysis & gasification & smelter

#### 5.1 Definition of Criteria

The ranking is based on the suitability for the (economical) treatment of WEEE. Six criteria were used. In the next sections the criteria are specified and quantified when possible (in random order).

#### 5.1.1 Energy efficiency

The percentage of the energy content of the feed converted into energy (including energy consumption of the installation). This includes electricity and heat generated in a steam cycle or with the product gas in a gasturbine, and the energy value of the char, minus the energy consumption of the process (*e.g.* heating of ovens and smelters and oxygen production). The numbers for electricity and heat are important indicators because of the different economic values of these types of energy.

The possibility to make use of rest heat streams of the process might have a significant influence on the overall efficiency. The energy efficiency might be improved when this 'rest' heat can be used for district heating, production of demineralised water, or in other applications.

## 5.1.2 Bromine and Antimony recovery

The distribution of both elements over the streams of the process are important, *e.g.* grate ash, fly ash, bottom ash, pyrolysis char, or scrubber. Furthermore, in what chemical form the elements are present and with what technique they can be recovered (isolated). For the possible recovery a quantitative indication should be given.

#### 5.1.3 General quality of residues/secondary products

What are the secondary products of the process and what amounts will be produced. Secondary products comprise fly ash, grate ash, smelter slag, metals, etc. Important is whether these products can be used as construction material or sold as raw material (*e.g.* metals) or have to be disposed of as hazardous waste? This depends on mainly on whether the residues fulfil regulations on leaching.

### 5.1.4 Emissions (legislation)

The expected emissions of the process are mentioned and quantified when possible (in ppm or kg/h). This includes the gaseous effluents with possibly bromine, chlorine,  $SO_2$ ,  $NO_x$ , dioxins, and furans. Also the grate, fly, and bottom ashes and smelter residues that are not suitable as construction material are considered as an emission. The current maximum emissions, according to European Union legislation, for bromine, HBr, HCl, and  $NO_x$  are 0.7 (proposed), 15 (proposed), 10, and 70 mg/Nm<sup>3</sup>, respectively.

## 5.1.5 Operational experience

The current experience with the installation gives an indication of the performance, especially experience with processing ASR. At least three years in operation is given as indication. Furthermore, important indicators are throughput, operational hours, and flow per hour. If a process is already in operation on full scale with (co-feeding of) WEEE and/or ASR it will be advantageous for implementation

### 5.1.6 Profit expectations and ROI

If possible, a rough estimate of the profit expectations of a (industrial) scale process for the recycling of WEEE is made. Important indicators are the expected investment costs and economic value of the produced energy and the secondary products that determine the final gate fee that has to be charged. The values of the energy and products are determined by the (availability of a) market.

### 5.2 Evaluation and Discussion

The evaluation is based on literature and reports available to ECN, questions to the companies, and ECN knowledge of the market. ECN also sent an enquiry with a 'case' to the companies with the question to evaluate their process with the mentioned criteria. In Appendix C the enquiry that has been sent to the companies is added and in Appendices D-G the reactions are added.

In the discussion and evaluation the processes are ranked for each criterion. The best process for each item is (are) ranked "1". Other processes are ranked 2 through 4, depending on the mutual differences (*i.e.* 'almost equal' vs. 'much better').

#### 5.2.1 Energy efficiency

In the Austrian Energy process electricity and heat is produced from the steam generated in the combustor, while in the ALSTOM and Von Roll processes steam is used that was generated with the flue gas. The output of the Gibros process is a syngas (a gas mixture mainly consisting of H<sub>2</sub> and CO), which can be used in a gas engine or turbine to generate also electricity and heat. However, the syngas can also be applied as feedstock for the chemical industry. Although currently there is limited experience in using waste-based syngas as chemical feedstock, this might be an issue for consideration with respect to the recycling targets.

Based on general 'rules-of-the-thumb' the overall (brut) electrical efficiencies, excluding the installations own consumption, are < 30% for direct combustion (Austrian Energy), 20-30% for staged combustion (ALSTOM and Von Roll), and maximum 35% for a gas engine with an integrated heat recovery (Gibros). These numbers are in agreement with the data provided by the companies. The data provided by the companies are *net* electrical efficiencies, it is therefore assumed the own energy consumption is subtracted. The total energy efficiencies including heat recovery can vary between 70 and 85%. The ranking in the Table is based on the provided data on the *net* electrical efficiencies.

-	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Ranking:	3/2	2	2	1/2

In all processes the energy efficiency might be improved when 'rest' heat can be used for district heating, production of demineralised water, or in other applications.

#### 5.2.2 Bromine recovery

In the ALSTOM, Von Roll, and Gibros processes the gasification or pyrolysis results in a reducing atmosphere, which may result in the formation of both metal halides in the bed, and formation of HBr. However, in the smelters the bromine will largely be released again as HBr and as the smelter gases are combined with the flue gas the HBr will be present in those streams. The subsequent combustion in the ALSTOM and Von Roll processes will also generate some  $Br_2$ , part of which will be converted into HBr with the  $SO_2$  present (when no limestone is used in the bed).

Effectively, most of the bromine (> 90%) will be present in the gas phases, either as HBr or as  $Br_2$ . When a suitable 'wet' gas cleaning is present both bromine compounds can be removed from the gas (*i.e.* two scrubbers: one alkaline to remove the HBr and the second with bisulphite to remove the  $Br_2$ ).

The installations of Von Roll and Gibros have a "wet" gas cleaning in which the HBr (and Br<sub>2</sub>) is captured (*e.g.* ranking "1").

The 'standard' ALSTOM has a "dry" scrubber resulting in HBr emissions, as part of the acid is not captured (e.g. ranking "3"). However, ALSTOM has experience with bromine removal in their Cleanaway (UK) hazardous waste treatment plant, where similar high bromine concentrations are handled. Furthermore, they have a pilot plant running with wet APS systems aimed at recovering halogens, which recovered HCl from waste flue gases. A similar facility can be included in the process to recover all bromine in the flue gas. In that case the ranking will be "1".

In the 'standard' Austrian Energy process limestone is used in bed to capture the SO<sub>x</sub> and this will also capture the HBr. All the bromine is then contained in the ash as CaBr<sub>2</sub> (and CaBrCl and CaCl<sub>2</sub>) salt. The resulting bromine concentration in the ash might be too low to allow

economic recovery (*e.g.* ranking "4"). Austrian Energy has indicated that no limestone will be used in case of WEEE treatment. In that case the bromine distribution will be comparable to that found in the TAMARA studies: 2% in the grate ash, 20% in the fly ash, and 78% in the flue gas (see also section 3.2.1) [4,9]. When the FLUAPUR process (see section 5.2.5) is incorporated in the installation, the bromine in the ashes will also end up in the flue gas, so the bromine will be fully recovered.

The Austrian Energy subsidiary CT Environmental (see section 4.9.5) sells scrubbers especially designed for bromine recovery. When such a unit is included in the Austrian Energy installation the bromine recovery will also be ranked "1".

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Ranking:	4/1	3/1	1	1

#### 5.2.3 Antimony recovery

In the oxidative conditions of the Austrian Energy combustion, part of the Sb will remain in the combustor (grate ash) and part will volatilise, especially with a high halide content in the feed. This will be captured (precipitate as dust) in the gas cleaning, and thus finally be present in that residual fly ash stream, *e.g.* comparable to the results from the TAMARA tests [4].

In the other three processes the thermal conversion (gasification or pyrolysis) affords a reducing atmosphere that results in volatising of part of the antimony as  $SbBr_3$ . In the ALSTOM process this will be oxidised again in the cyclonic combustion chamber and in the Von Roll process the same will happen in the CFB combustor. In the Von Roll processes the  $Sb_2O_3$  will end up in the slag treatment and in the ALSTOM process part will bind in the molten slag and part will end up in the ash.

In the Gibros process the volatilised Sb will not oxidise as the pyrolysis is followed by a (reducing) gasifier, therefore, all the Sb in the gas phase ends up in the gas cleaning. In the scrubber it dissolves in the water or afterwards it will be captured in the bag filter. A maximum of about 20% of the antimony dissolves in the mineral slag and is not recovered but locked into the mineral structure of the construction material (synthetic basalt) in a non-leachable form (the remainder 80% is potentially recoverable).

For the specific situation of processing WEEE no experimental data is available for the four processes. Furthermore, it is not possible to predict the partitioning of antimony, as the chemistry of Sb is relatively complicated under the conditions and temperatures of the discussed thermal processes. Therefore, no order of preference for the processes regarding the Sb recovery can be given. However, it is expected that the yield of antimony recovery for combustion processes will be higher than for pyrolysis and gasification processes. Due to the low concentration in the ashes economic recovery will not be feasible.

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Ranking:	1	1 /2	2	2

In spite of the technical possibilities of the processes, the concentrations antimony in the processed material are most likely too low for economic recovery.

## 5.2.4 Emissions (legislation permits)

All companies claim that the secondary (residual) products of their processes meet the current legislation limits (see also the next section). Detailed experimental information and testing with WEEE is necessary to determine exact emission data specifically related to using WEEE as (co-)feedstock. All processes rank "1".

## 5.2.5 General quality of residues/secondary products

In the discussion on the quality of residues and secondary products the bromine that is recovered is excluded as it is considered as the primary product.

The Gibros process affords Ferrous, non-Ferro, and a mixed metal stream as secondary products that can be recycled as raw material. From the ash (crystalline) synthetic basalt is produced in the smelter that fulfils all standard legislation and can be used as construction material. The process produces a syngas that is in the 'standard' situation used to generate electricity and heat. However, it can also be used as feedstock for the chemical industry or to produce liquid transportation fuels. This comprises methanol, like commercially in the industrial plant at SVZ Schwarze Pumpe, or gasoline and diesel with the Fischer-Tropsch process. This might be a potential additional advantage of the process. The process ranks "1" for this item, however, if no advantage is taken of the syngas but it is used for electricity generation the process ranks "2".

The ALSTOM process recovers non-oxidised metals from the fluidised bed that can be recycled as raw material. From the combustion ashes slag is made in the smelter. In this (non-crystalline) slag also volatile (heavy) metals are present, but the slag is applicable as construction material as it fulfils all leaching legislation. The Von Roll process affords a metal smelt and also a slag that fulfils all leaching legislation. Both processes rank "2". A minor advantage of Von Roll over ALSTOM is that the first applies to all ashes in the slag and thus have no waste ash streams, while the ALSTOM process has additional (small) waste ash streams.

In the 'standard' Austrian Energy process only the different ash streams are residual outputs. There is no facility to recover metals from the ash, *i.e.* a smelter. These ashes have to be landfilled, which results in additional costs, whereas with proper treatment construction materials can be made form the ashes like in the other processes (*e.g.* ranking "4").

The Austrian Energy subsidiary CT Environmental (see section 4.9.5) developed the FLUAPUR process to treat ashes and to recover heavy metals. The aim is to reach a fraction of reprocessable heavy metals and a mineral fraction, which contains real low amounts of heavy metals (almost down to average earth crust). The process concept is to treat the ash in a fluidised bed under a HCl containing atmosphere. Temperatures are below melting of the ash (approx. 950°C). At this temperature the heavy metals are converted to halides and become volatile. The metals are then recovered as metal chlorides form the flue gas for further treatment. The ash residue is almost free off heavy metals.

When slag is properly produced from ashes, it fulfils all leaching tests also when heavy metals are present (*cf.* the other three processes). The only benefit from the FLUAPUR process would be the recycling of the heavy metals. As long as there is a market as construction material for the slag, it is more beneficial to produce slag than to produce 'clean' ashes. Therefore, the ranking would be "3" with the FLUAPUR process integrated.

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Ranking:	4/3	2	2	1/2

#### 5.2.6 Operational experience

Austrian Energy has a lot of experience with the fluidised bed combustion technology (e.g. ranking "1"), however, relatively limited experience with ASR (e.g. ranking "2"). On the other hands, ALSTOM has less experience in general, but more with processing ASR (e.g. ranking "2" and "1"). Of the other two processes has Von Roll more experience than Gibros with their currently operational processes (e.g. ranking "3" vs. "4"). However, Von Roll has very limited experience with ASR (e.g. ranking "4"). Gibros has done more experimental work with ASR, but as these results are not public yet, a ranking "3" is given.

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Ranking:	1/2	2/1	3/4	4/3

#### 5.2.7 Profit expectations (ROI)

The expected profit (and the ROI) is based on the investment costs, operational costs, and the economical value of the secondary products. These aspects will determine the ROI of the installations in relation to the gate fee. When the gate fee is below current fees for landfilling and waste incineration will become feasible to process WEEE.

Due to the specific processing aspects for WEEE and the limited experimental experience with respect to the secondary products it is difficult to indicate the profit expectations. Only ALSTOM and Von Roll provided *estimates* for the investment costs and resulting gate fees. Some elements that determine the economics are briefly assessed.

**Operational costs**. Based on the complexity of the installations (*i.e.* number of units) the Austrian Energy process is assumed to be simplest and cheapest to operated and the Von Roll process the most expensive. Additional (major) costs in the Von Roll and Gibros processes are the use of oxygen; 790 and 270 kg oxygen per ton feed, respectively.

**Equipment lifetime**. The presence of large concentrations of HBr will effect the lifetime of the installations (see also: Investment costs). That means that the part of the installation between the thermal conversion and the gas cleaning can be subjected to corrosion due to the HBr (and HCl).

## Economic value of secondary products.

This cannot be quantified in this stage. Detailed information concerning chemical and physical characteristics is not available. Therefore, experimental information and testing with WEEE is necessary.

An indicative ranking is shown in the Table:

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Ranking:	1	2	3	2

## 5.3 Ranking

The ranking for each criterion of the processes in the previous sections are summarised in Table 5.2. As explained before, the best process(es) for each item is (are) ranked "1". Other

processes are ranked 2 through 4, depending on the mutual differences (*i.e.* 'almost equal' vs. 'much better'). In the last row of the Table the points are added up (the lowest total ranks 'best').

Table 5.2 Summarised ranking of the processes for each item

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Energy efficiency	3 / 2	2	2	1 / 2
Bromine recovery	4 / 1	3 / 1	1	1
Antimony recovery	1	1 / 2	2	2
Emissions	1	1	1	1
Quality of secondary products	4/3	2	2	1 / 2
Operational Experience	1 / 2	2 / 1	3 / 4	4/3
Profit expectations and ROI	1	2	3	2
Summarised	15 - 10	14 - 10	15 - 14	14 - 11

N.B. The added up totals in the last row are calculated <u>without weighting factors for the different criteria</u>.

#### 6. DISCUSSION

In this Chapter the results described in the previous Chapters are discussed.

#### 6.1 Thermal Processes

In Chapter 3 eight thermal processes, which operate commercially are discussed and evaluated for their suitability to process WEEE. Furthermore, six other processes that are in development or operate on pilot-scale were shortly discussed. Based on this information four out of the eight processes were considered potentially suitable for WEEE processing and selected for more detailed evaluation, *i.e.* the processes of:

- Austrian Energy,
- ALSTOM/Ebara,
- Von Roll RCP, and
- Gibros PEC.

## 6.2 Bromine and Antimony

In Chapter 4 the chemistry of bromine and antimony was discussed in relation to their (expected) behaviour in the thermal processes. The behaviour, and related partitioning, of bromine is relatively predictable. The chemistry of antimony is rather complicated and only limited experimental data is available. Therefore, the antimony partitioning could only be qualitatively be predicted.

Nearly all experimental data of processing WEEE is available from the TAMARA studies that comprised (co-)processing of WEEE in a combustion installation. Except for the Austrian Energy process, the thermal processes discussed in this report function according different principles (e.g. gasification and smelters). Therefore, the TAMARA results are not representative for these processes! In the discussion and evaluation of the processes, care should be taken to keep these differences clear in mind.

## 6.3 Evaluation and Ranking

In Chapter 5 the processes of Austrian Energy, ALSTOM, Von Roll, and Gibros were evaluated and a ranking for each criterion was made. In Table 6.1 the ranges of the ranking are shown in the case that <u>no weighting factors were used</u> for the different items (the lower the number, the higher the ranking).

Table 6.1 Ranking of processes without weight factors

	7	8	9	10	11	12	13	14	15	16	17	18
Austrian Energy												
ALSTOM/Ebara												
Von Roll RCP												
Gibros PEC												

All four processes are in principle able to process WEEE. However, none of the evaluated processes is *especially designed* for processing WEEE! Therefore, all processes can be improved on one or more points.

When weighting factors are added to the criteria, the evaluation and ranking is differentiated based on the following considerations:

- When the main purpose of the process is decided to be "best available technology", the Gibros processes would be the 'best' process.
- However, when "operational experience" together with low running costs is considered as the most important issue and given a high weighting factor, the Austrian Energy processes would rank the highest.
- If "proven technology" is the criterion for the decision than ALSTOM/Ebara will be the choice because of the plant in Japan that is running with ASR

#### 6.4 Intermediate Discussions

The concept end-report was presented and discussed on the meeting of the European Brominated Flame retardant Industry Panel (EBFRIP) held on March 17, 2000 in Brussels, Belgium. Based on the discussion some points in the report are discussed in more detail and some points have been clarified. The report with the additions was presented and discussed on the meeting of the Bromine Science and Environmental Forum (BSEF) meeting held on March 22, 2000 in Brussels, Belgium.

## 6.5 Funding

In Appendix I possible funding options for the route to implementation are briefly mentioned. When more insight is available concerning the objectives and detailed activities of the next phase (e.g. feasibility studies, implementation scenarios, national vs. European level, etc.) more specific information can be collected.

#### 7. CONCLUSION AND CONTINUATION

#### 7.1 Conclusion

In this report thermal processes were evaluated for their suitability for the feedstock recycling of bromine and antimony, with energy recovery, from WEEE. The processes of Austrian Energy, ALSTOM/Ebara, Von Roll RCP, and Gibros PEC were evaluated in detail with the defined criteria. None of the evaluated processes is especially designed for processing WEEE and therefore, all processes can be improved on one or more points. All four processes are in principle able to process WEEE and as no weighting factors were defined for the criteria no 'best' process was determined.

When weighting factors are added to the criteria, the evaluation and ranking is differentiated based on the following considerations:

- When the main purpose of the process is decided to be "best available technology", the Gibros processes would be the 'best' process.
- However, when "operational experience" together with low running costs is considered as the most important issue and given a high weighting factor, the Austrian Energy processes would rank the highest.
- If "proven technology" is the criterion for the decision than ALSTOM/Ebara will be the choice because of the plant in Japan that is running with ASR

#### 7.2 Continuation

A first issue that has to be resolved is whether the bromine salts that are produced in the thermal processes are suitable to be used as feedstock in the bromine industry. Therefore, experiments under representative conditions are necessary to determine the quality of produced bromine salts and their quality as feedstock bed has to be determined.

Aiming at the implementation of a thermal processes for the processing of WEEE in 2004 a phased approach was proposed in Section 1 "Introduction". For Phase 2 feasibility studies on the selected technologies were foreseen and in Phase 3 implementation scenarios would be developed. An important issue in that phase is to identify the availability of WEEE, also in relation with optimal logistics.

The minimum plant size for economically processing is estimated to be 100,000 ton/y. To minimise the costs of logistics, feed streams for a plant should be available within an approximately 150 km radius (*e.g.* within the Netherlands). However, in the Netherlands only 7000 ton WEEE plastics, containing BFRs, is annually available.

<u>It is therefore essential to co-operate with other industries and co-process the WEEE plastics with their wastes</u>. Possible waste streams for co-processing are Automotive Shredder Residue (ASR), solid and liquid bromine containing streams (from industrial processes), and other bromine containing wastes (*e.g.* package material). In the discussions with EBFRIP and BSEF it was considered as the necessary second continuation step to form consortia to contract these waste streams or to combine with initiatives for these streams.

## 8. LIST OF ABBREVIATIONS

AE Austrian Energy APC Air Pollution Control

APME Association of Plastic Manufactures in Europe

ASR Automotive Shredder Residue BFB Bubbling Fluidised Bed BFR Brominated Flame Retardant

BIGCC Biomass Integrated Gasification Combined Cycle

Br Bromine Br<sub>2</sub> Bromine; a gas

BSEF Bromine Science and Environmental Forum

Ca Calcium

CaCO<sub>3</sub> Calcium carbonate = major consistuent of limestone

CFB Circulating Fluidised Bed

Cl Chlorine

CO Carbon monoxide

DECA Decabromodiphenyl ether
DIF Dioxins and Furans

EBFRIP European Brominated Flame Retardant Industrial Panel

ECN Netherlands Energy Research Foundation (Dutch acronym for: Energieonderzoek

Centrum Nederland)

ECVM European Council of Vinyl Manufactures EEE Electrical and Electronic Equipment

EU European Union

FICB Fast Internally Circulating Bed

FR Flame Retardant
H<sub>2</sub> Hydrogen; a gas
HBr Hydrobromic acid
HCl Hydrochloric acid

 $\begin{array}{lll} HIPS & High Impact Poly Styrene \\ MPW & Mixed Plastics Waste \\ MSW & Municipal Solid Waste \\ MW & Megawatt = 10^6 Watt \\ MW_e & Megawatt electric energy \\ MW_{th} & Megawatt thermal energy \end{array}$ 

Na Sodium

 $NO_x$  Nitrogen Oxides (*i.e.* NO and  $NO_2$ )

OCTA Octabromodiphenyl ether
PBKD PB Kennedy & Donkin Limited
PCDD Polychlorinated Di-benzodioxins
PCDF Polychlorinated Di-benzofurans

PKA German acronym for: Pyrolyse Kraftanlagen

PVC Polyvinyl Chloride RDF Refuse Derived Fuel ROI Return on Investment

Sb Antimony

SNCR Selective Non-Catalytic Reduction (for NO<sub>x</sub> removal)

 $\begin{array}{c} SO_x \\ TBBA \end{array}$ Sulphur Oxides (*i.e.* SO<sub>2</sub> and SO<sub>3</sub>) Tetrabromobisphenol-A Terminska Processor Studsvik TPS

Waste of Electrical and Electronic Equipment WEEE

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## **APPENDICES**

Appendix A: Detailed Process Schemes of *Austrain Energy* and *ALSTOM/Ebara* (not available from *Von Roll RCP* and *Gibros PEC*)

Appendix B: List of Contacts

Appendix C: Enquire with Case

Appendix D: Response of 'Austrian Energy' (not received)

Appendix E: Response of 'ALSTOM Power Ltd.'

Appendix F: Response of 'Von Roll RCP'

Appendix G: Response of 'Gibros PEC'

Appendix H: Summarised Evaluation

Appendix I: Possible Funding

# APPENDIX A DETAILED PROCESS SCHEMES

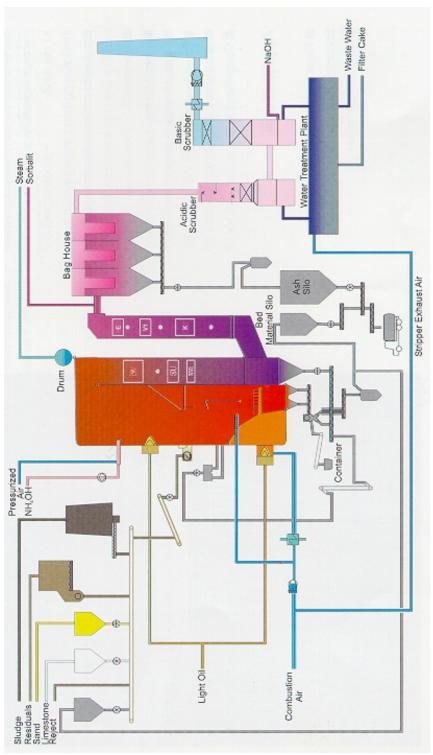


Figure 1 Schematic process overview of Austrian Energy

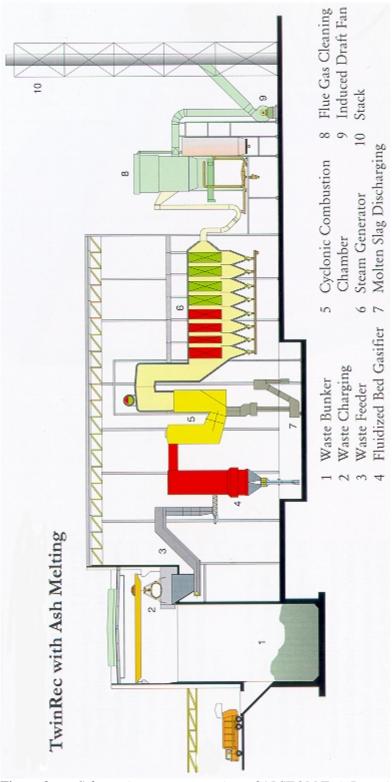


Figure 2 Schematic process overview of ALSTOM TwinRec with ash melting

#### APPENDIX B LIST OF CONTACTS

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& Dr. Christian Steiner

(Product Manager Fluidized Bed Technology)

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## APPENDIX C ENQUIRE WITH CASE

Sent by email on February 21, 2000.

Dear Mister [name],

Recently, ECN made an inventory of the processes that are suitable for processing Automotive Shredder Residue (ASR). This inventory was made for the European Brominated Flame retardant Industry Panel (EBFRIP). The EBFRIP wants to recycle Waste of Electrical and Electronic Equipment (WEEE). Due to the similar composition of ASR and WEEE, experiences with processes for the treatment of ASR give a good indication of their suitability to process WEEE.

In the previous contact we had on the above mentioned subject you indicated that the [name] process is suitability for the treatment of ASR. Based on the inventory ECN made, four processes were selected (among which the [name] process) for more detailed evaluation. Therefore, I would like your response to some more specific questions concerning the performance of the [name] process when processing ASR or WEEE on industrial scale:

#### Introduction of the 'case'

The European Brominated Flame retardant Industry Panel (EBFRIP) wants to recycle WEEE: converting it into energy and products as optimal as possible. In the case of WEEE, containing brominated flame retardants (BFRs), this means recycling of bromine, antimony, other value metals, and recovery of the energy content.

Together with the waste industry, EBFRIP wants to realise several industrial plants for the WEEE recycling all over Europe. The total WEEE fraction is estimated to be 1,000,000 tons/year, including ferrous, non-ferrous, glass, others, and plastics. An indicative composition of the WEEE is: plastics 25%, ferrous 25%, non-ferrous 25%, and others (glass, wood, and others) 25%. The annual amount of plastics in the WEEE to be processed is 270,000 tons and this fraction may contain 4-9% bromine and 1% antimony.

EBFRIP has defined six criteria based on which they will select the process(es) for a further feasibility studies and basic engineering. These criteria are *i*) in random order as the relative importance has not yet been defined and *ii*) general as the same questions are sent to all companies):

#### • Energy efficiency

Which percent of the energy content of the feed will be converted into energy? This includes electricity and heat generated in a combustor or with the product gas in a gasturbine, and the energy value of the char. Please, specify the values for electricity and heat.

#### • Bromine and Antimony recovery

Indicate the distribution of both elements over the streams of the process, *e.g.* grate ash, fly ash, bottom ash, pyrolysis char, or scrubber. In what chemical form are the elements present and how can they be recovered (isolated)? Give a quantitative indication of the possible recovery.

#### • General quality of residues/secondary products

What are the secondary products of the process and what amounts will be produced. Secondary products comprise fly ash, grate ash, smelter slag, metals, etc. Can these products be used as construction material or sold as raw material?

#### • Emissions (legislation)

Please specify the expected emissions of the process and quantify the emissions when possible (in ppm or kg/h). This includes the gaseous effluents with possibly bromine, chlorine,  $SO_2$ ,  $NO_x$ , dioxins, and furans. Also the grate, fly, and bottom ashes and smelter residues that are not suitable as construction material are considered as an emission.

#### • Operational experience

Could you indicate the expected performance of your process when recycling ASR on the (industrial) scale mentioned? Included the current experience of the installation with processing ASR (throughput, operational hours, flow per hour).

#### Profit expectations and ROI

Give a rough estimate of the profit expectations of a (industrial) scale process for the recycling of WEEE? What would be the investment costs and economical value of the secondary products?

Please, add also additional remarks that might be important for the evaluation. For more information you can contact me at below mentioned phone, fax, or email. You may also contact Mr. Lein Tange from EBFRIP at phone: +31 (0)115-689275 or email: tangel@broomchemie.dsbg.com.

Also when you answered a question in our previous contact, please respond to it now. As all companies responses will be included in the final report. For the same reason I would appreciate if you answered by email.

In order to keep pace in the project, I ask you to respond before March 14. Thank you.

Yours sincerely, Harold Boerrigter

\_\_\_\_\_\_

Dr. ir. Harold Boerrigter

ECN - Biomass

Netherlands Energy Research Foundation (ECN)

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# APPENDIX D RESPONSE OF 'AUSTRIAN ENERGY'

not received

## APPENDIX E RESPONSE OF 'ALSTOM POWER LTD.'

Received by email on March 14, 2000. ALSTOM Power (Switzerland) Ltd. Dr. Christian Steiner Product Manager Fluidized Bed Technology

ALSTOM Power provided extensive and specific process information to ECN, in more detail than the other companies. Some of the data provided by ALSTOM was omitted to have comparable data for all companies. However, the detailed data is available at EBFRIP.

#### **ENERGY EFFICIENCY**

A TwinRec plant with electrical energy production achieves a net efficiency of 17-25%. If electricity and heat (process steam or district heating) are produced, the net efficiency can be risen to over 70%.

#### **BROMINE AND ANTIMONY RECOVERY**

#### **Bromine**

In the gasification step of TwinRec, temperatures are high enough to destroy all organic bromine compounds. The subsequent combustion will result in hydrobromic acid and bromine (HBr, Br<sub>2</sub>), ca. 50% each. All HCl, HBr and some of the Br<sub>2</sub> will be stripped in the acid step of a wet scrubbing system. The remaining Br<sub>2</sub> will be stripped in a second step at higher pH, with additives as applied in our Cleanaway, UK, hazardous waste treatment plant, where similar high bromine concentrations are handled this way. The scrubber solutions are available for neutralisation and evaporation, ultimately delivering >80% of the bromine as alkali bromide.

The fly ash is converted into molten slag at ca. 1300°C and will bind only minor quantities of the bromine (<10%). This quantity is not available for leaching; the slag will pass limits for reuse in construction according to Dutch legislation.

Depending on metal availability, some bromine (10%?) will end as bromides in the gasifier bottom ash. According to Dutch legislation, leaching of bromide may thus arise as a problem, which we currently cannot quantify, as we have not treated WEEE. However, we have a number of bottom ash treatment processes available, which will enable us to meet any required legislation with appropriate steps.

#### Antimony

The chemically reducing conditions in the gasifier will volatilise a large portion of the antimony, likely >50%. Some of this will bind into the molten slag, the remainder will end up as oxide in the filter ash, from where it can be recovered if desired. However, practical experience with high concentrations of antimony is yet to be gained.

#### QUALITY OF THE SECONDARY PRODUCTS/RESIDUES

- *Metals*: recyclable to metal recovery processes.
- Glass granulate: recyclable to building industry.
- Gasifier bottom ash: recyclable to building industry; depending on country regulation an additional treatment step may be necessary to fulfil the recycling regulations.
- Filter ash and APC residues: to landfill; amount depending on selected APC process.

((Mass balance and leaching specifications of the residues are omitted)).

#### **GASEOUS EMISSIONS**

ALSTOM Power selects the best-suited type of Air Pollution Control (APC) technology according to the waste type, the country regulations and the customer requirements. The 17.BImSchV and other European Regulations can be fully met with current APC technology.

## EXPECTED PERFORMANCE AND OPERATIONAL EXPERIENCE

Expected performance for ASR treatment at industrial scale: 7300 operating hours per year.

## Current experience:

- At the two pilot plants (Sodegaura and Fujisawa) a big number of tests were conducted with different waste types, among them ASR.
- The full size plant Aomori (2 lines, each with 40 MW<sub>th</sub>, 6.3 t/h ASR and 3.1 t/h Sewage Sludge) is under commissioning.
- ((A more detailed reference list was attached)).

## PROFIT EXPECTATIONS, INVESTMENT AND TREATMENT COSTS

These values depend greatly on the single project (plant capacity, national regulations, combinations with existing infrastructure; required standard for engineering, components and building; financial calculation; guarantees etc.). The profit expectations depend on the gate fee of the waste and subsidies (*e.g.* for bromine recycling, etc.).

## As a rough indication for ASR:

- Investment cost: 450 1100 Euro/ton per year installed capacity.
- Treatment cost: 50 230 Euro/ton.

## APPENDIX F RESPONSE OF 'VON ROLL RCP'

Received by email on March 7, 2000. Von Roll Environment Technology Dr. Marc Stammbach RCP-Marketing

For the following questions a RCP-Plant in a standard configuration (without HSR), wet flue gas treatment and heavy metal extraction is assumed:

#### **ENERGY EFFICIENCY**

Energy conversion: 84% to heat or 28.1% to electricity.

#### **BROMINE AND ANTIMONY RECOVERY**

Distribution: fly ash and scrubber together: nearly 100%

*Recovery*: with Na<sub>2</sub>SO<sub>3</sub> in wet scrubber we transform it to Br<sup>-</sup> it can be won out of the solution after heavy metal extraction (up to 100%).

Antimony: together with Zn, Cd, Pb - Hydroxides: around 60-100% (operational experience missing).

## GENERAL QUALITY OF RESIDUES / SECONDARY PRODUCTS

Secondary products:

- Vitrified slag as construction material: 25 %
- Copper iron alloy as copper raw material: 25 %
- Bromine: 4-9%
- Zn, Cd, Pb, Sb hydroxide to rotary kiln recycling process: 1-2%
- Fly ash matrix after heavy metal extraction: quality similar to grate slag and good for road construction: 2-4%

## Residue:

Left over waste water (heavy metal free, bromine free): can be discharged or concentrated to salt for underground salt mine storage.

#### **EMISSIONS (LEGISLATIONS)**

We can achieve any current required regulations with the appropriate flue gas treatment technology (or cut it down even further) the same is valid for wastewater (see residue above).

#### OPERATIONAL EXPERIENCE

We have an industrial operation in Bremerhaven (Germany) with a throughput of 6 ton/h, *i.e.* 17.5 MW waste power.

Only a test with 100 t German ASR was performed in January 1999. It worked well and very smoothly. We experienced higher dust loads (as expected).

The later test with 400 tons Swiss ASR was abandoned due to operational equipment problems. We have been several months online last year (especially from September to November 1999), reached a constant throughput of 50% nominal capacity and higher loads over several hours. However, there were still problems left, causing interruptions. We are currently modifying our plant again *i.e.* smelting oven: simpler design and improved cooling).

We expect to go on stream with waste by April 22, 2000 and with ASR in May 2000.

## PROFIT EXPECTATIONS AND ROI

Cannot be given, as they are a function of the treatment fee, which can be demanded. Values of secondary materials: zero from plant (minimal value minus transport). Indication of investment costs (for a 135,000 ton/year plant, 54 MW, with building): 75 Million Euro, resulting in approx. 131 Euro/ton treatment cost.

#### APPENDIX G RESPONSE OF 'GIBROS PEC'

Received by email on March 18, 2000. Gibros PEC B.V. Ir. Harro A. Zanting Environmental Engineer

#### **PURPOSE**

ECN inventory of possible future treatments of Waste from Electrical and Electronic Equipment (WEEE) on behalf of the European Brominated Flame retardant Industry Panel (EBFRIP).

#### **GENERAL**

General aim of the PEC<sup>®</sup> (Dutch acronym: Product en Energiecentrale) process is to produce syngas, metals and high quality construction material from different waste streams. The technology is based on staged gasification and smelting. Publicly available process description can be found in other (ECN) documents or in Environmental Impact Assessments (EIA or 'MER' in Dutch) of PEC<sup>®</sup> projects.

Based on the indicative composition of WEEE as stated by ECN, the PEC® process is very suitable for treating this material. Detailed figures on material balance, costs etc. depend on the specific aspects of the PEC project. The data below are general characteristics.

#### **ENERGY EFFICIENCY**

The energy content of the WEEE is converted into a 10 MJ/Nm<sup>3</sup> syngas with an efficiency of 70-80%. About 10% of the energy content is recovered as MP-steam from the syngas heat recuperation. Further energy conversion is project specific. The syngas and the steam can both be used for electricity production. If this is the aim, Gibros PEC generally offers the power production unit with gas engines (efficiency 35-40%) as prime movers.

In projects where syngas is supplied as chemical feedstock for *e.g.* hydrogen (H<sub>2</sub>) production, one GJ of syngas generally saves 1.3-1.4 GJ of natural gas.

#### **BROMINE AND ANTIMONY RECOVERY**

Bromine is either recovered in a salt solution or as crystallised NaBr salt. Antimony is recovered in a mixture of recyclable metaloxides.

In the pyrolysis or degasification step, antimony largely remains in the solid phase (pyrolysis residue) which is further processed in the smelter. Bromine partly goes with the (pyrolysis) gas phase. In both the smelter and the gasifier, bromine is fully converted into HBr. In the gastreatment units, the HBr is converted to a NaBr solution through scrubbing. This solution can, if requested, be concentrated by evaporation and crystallised.

In the smelters, a maximum of about 20% of the antimony (Sb) dissolves in the mineral slag and is not recovered but locked into the mineral structure of the construction material (synthetic basalt) in a non-leachable form. The main part of the antimony ends up in the gas phase from the smelter and is recovered as oxide-dust.

#### PRODUCT QUALITY

All products comply with market specification. The syngas is thus clean that it can be used as a fuel under the same (legal) conditions as primary fuels such as natural gas. The construction

material meets current Dutch legal requirements on leaching of damaging components. The halogen-salt solution is thus clean that it can be drained to, preferably salty, surface water. The metaloxide mixture (containing antimony) meets input specifications of metallurgical industries.

#### **EMISSIONS**

There are no direct air-emissions from the conversion process. Air emissions only occur in case the clean syngas is used as a fuel. Fluegas concentrations are below current Dutch legal requirements.

#### **OPERATIONAL EXPERIENCE**

Typical throughput of a single pyrolysis/gasification module is 4 tonnes per hour, which is scaled up by the installation of more modules with the same, proven capacity. Total treatment capacity also depends on the pre-separation of moisture and metals. Gibros PEC, as a supplier, guarantees an availability of 7500 hours per year on average.

The public availability of the results from a recent demonstration with ASR on behalf of Auto Recycling Nederland (ARN), is still under discussion.

## PROFIT EXPECTATIONS

A profitable operation of a PEC plant is feasible with gate fees below current Dutch tariffs on waste disposal (landfilling and incineration).

## APPENDIX H SUMMARISED EVALUATION

In Section 5.2 "Evaluation and Discussion" the criteria for the processes are discussed. In the Table in this Appendix the evaluation is summarised.

 Table 1
 Overview of the selected processes and performance

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
Energy efficiency	for a combustor is 25%	Net electrical energy production: 17-25% With heat production up to 70%.	84% to heat <i>or</i> 28% to electricity.	Cold gas efficiency 75-85% Net electrical efficiency 30-35%.
Bromine recovery	In standard plant: HBr and HCl remain in ashes. - No recovery possible With wet scrubber: captured.	In standard plant: no wet scrubber: HBr emissions Knowledge of wet scrubber is available; then captured.	and in fly ash, nearly 100%	Captured in wet scrubber.
- chemical form, stream	Salts (e.g. CaBr <sub>2</sub> ) in ashes With scrubber: NaBr < 10% in fly ash to smelt.	Flue gas Bromine emissions or captured in scrubber < 10% in fly ash to smelt.	NaBr in scrubber.	Syngas Captured in scrubber: NaBr.
Antimony recovery	In ashes.	Mainly in ashes Part will end up in slag.	Together with Zn, Cd, and Pb, 60-100%.	~80% in metal dust.
- chemical form, stream	Sb <sub>2</sub> O <sub>3</sub> in ashes.	Sb <sub>2</sub> O <sub>3</sub> in ashes.	As hydroxides.	Sb <sub>2</sub> O <sub>3</sub> in dust 20% lost: immobilised in basalt.
Emissions	by limestone SNCR for NO <sub>x</sub> removal No thermal NO <sub>x</sub> due to low temperature With	Expected Br emissions	With scrubber: according to Br legislation in Europe Heavy metal sludge (~2 wt%) Solid residues (~5 wt%).	Less than 0.5% of feed No direct air emissions According to legislation in Europe.
Quality of secondary products	No products, only several ash streams.	Non-melted and -sintered metals: Al, Cu, and Fe Slag-granulate meets leachability regulations.	Heavy metals Palletised slag as cement additive.	Metal mixture meets specs of metallurgical industry Slag affords basalt Fuel gas can substitute natural gas.
Operational Experience	Extensive with 'simple' combustion Two years with 1000 operational hours for ASR.	1995-97 a pilot plant and since 1997 a 4.8 MW <sub>th</sub> plant in Japan Several tests with ASR.		Different fuel tested ASR test performed, no results available.
- expected for WEEE	Comparable to ASR tests.	Comparable to ASR tests.	Comparable to ASR tests.	Comparable to ASR tests.
- feed size limits and	, .	For particles < 30 cm no		After pre-treatment: does
uniformity	pre-treatment Does not have to be uniform	pre-treatment Does not have to be uniform Wide range of compositions (e.g. water, ash), very fuel flexible.		not have to be uniform, 2D size < 15 cm Variable compositions can be handled.
Return on Investment		Operational hours: 7300/y (with ash smelting).	Claimed availability: 7500 h/y.	Guaranteed average availability: 7500 h/y.
- operational costs			Oxygen is required (~790 kg/ton feed).	Oxygen is required (~270 kg/ton feed).
- investment costs		Per ton per year installed capacity: 450-1100 Euro Gate fee: 50-230 Euro/ton.	For a 135,000 ton/y plant (54 MW, with building): 560 Euro/ton per annual installed capacity.	Gate fee below current Dutch tariffs on waste disposal.

	Austrian Energy	ALSTOM/Ebara	Von Roll RCP	Gibros PEC
			Resulting in 131 Euro/ton gate fee.	
- economic value of products	No secondary products.	Metal (~1 wt%) and slag (~20 wt%) can be recycled.		
- Scale-up	FICB units with capacities of 30-40,000 ton/y Maximum up to 80,000 ton/y.			Single gasification module: 4 ton/h Scale- up by installation of more modules.
Total impression	standard plants not suitable for Br and Sb recovery, however, process extensions are	Much operational experience in Japan No bromine recovery, but emissions! - With wet scrubber combined with experience from Japan this is resolved.	- One ASR test Limited	Technically versatile process No extensive operational experience.

#### APPENDIX I POSSIBLE FUNDING

#### • National (Dutch) level

Depending on the proposed content of the project possible funding from Novem or the Dutch government can be searched. Information of many Dutch programmes is provided by SENTER (a Dutch governmental foundation) and available on the Internet: <a href="http://www.senter.nl">http://www.senter.nl</a> (in Dutch).

## European Union

For the implementation of thermal processes for the feedstock recovery of bromine and antimony, with of energy recovery, from Waste Electrical and Electronic Equipment (WEEE) in principal financial support of the European Union is possible. For financial support in demonstration of the application the LIFE-Environment program is available. Originally the deadline for submission of proposals was April 1, 2000. Due to delay in the budgetary discussion of the European parliament the deadline is postponed. The new deadline is 31 October 2000.

Summarised program scope and objective of LIFE-Environment:

- (4c) The prevention, reuse, recovery, and recycling of waste of all kind, to ensure the sound management of waste streams.
- (5) Demonstration projects must set out to test an innovative solution to an environmental problem and lead to concrete and practical results of the project.
- (6) Innovative demonstration projects must be implemented at a scale that allows evaluation of technical and economic viability of large-scale introduction.
- (7) LIFE-Environment is not directed at research or investment in existing technology or infrastructure.
- (8) LIFE-Environment aims to bridge the gap between research and development results and widespread implementation.

Other programmes considered are *a/o* the 5th Frame Work and Eureka, however, these proved to be less suitable. Information about European programmes is provided by EG-Liaison (a subdivision of SENTER) and is available on the Internet: <a href="http://www.egl.nl/">http://www.egl.nl/</a>.