



Thermal Treatment as a Chance for Material Recovery

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Abstract The recovery of materials in the course of thermal waste treatment may sound contradictory at first glance because thermal treatment is supposed to destroy materials. However, this is only the case for organic materials. But waste consists of more: Metals and minerals are part of the trash, and there are options to get them back afterward or better **by** thermal treatment.

This chapter addresses the possibilities for recovering resources for material applications by thermal waste treatment. Two thermal routes are considered: Waste-to-energy (WtE) plants and pyrolytic disintegration approaches.

WtE enables the recovery of iron, nonferrous metals, and also minerals from bottom ash. Another opportunity for material recovery is flue gas utilization. The recovery and material utilization of HCl and sulfur (in the form of gypsum) has been industrially practiced for decades. In the last few years, the first approaches to recover metals from the filter dust were also industrially implemented.

Pyrolytic processes offer the chance to recover valuables from composite material parts, like carbon fiber-reinforced plastics (CFRP), or from metal-enriched fractions of other waste treatment processes like shredder residues. The containing plastics can be volatilized at high temperatures and the emerging pyrolysis gases can be utilized to supply the thermal energy for the process. The absence of oxygen and relatively low temperatures prevents the valuables in the composite matrix from damage.

Keywords Bottom ash, Dry ash discharge, Flue gas cleaning, Material recovery, Pyrolysis

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Abbreviations

CFB	Circulating fluidized bed (reactor)
CFRP	Carbon fiber-reinforced plastics
DM	Dry matter
DOC	Dissolved organic carbon
FGC	Flue gas cleaning
KEZO	Kehrichtverwertung Zürcher Oberland (WtE plant in the Zurich region)
MSW	Municipal solid waste
MSWI	Municipal solid waste incineration
RDF	Refuse-derived fuel
SCR	Selective catalytic reduction (of nitrogen oxides)
SNCR	Selective non-catalytic reduction (of nitrogen oxides)
SRF	Solid recovered fuel
WEEE	Waste electrical and electronic equipment
WtE	Waste-to-energy

1 Introduction

The subject of this section is the recovery of materials in the course of the thermal treatment of waste. This may sound contradictory at first glance. Thermal treatment is supposed to destroy materials. However, this is only the case for organic material. But waste consists of more. Metals and minerals are part of the garbage, and there are options to get them back afterward or better **by** thermal treatment.

Figure 1 gives an overview of the basic process concepts for the thermal treatment of waste. The state of the art for the treatment of municipal solid waste (MSW) is combustion, directly in waste-to-energy (WtE) plants, or after a pretreatment process, as so-called refuse-derived fuels (RDF), by mono-incineration in RDF power plants, or by co-combustion in cement kilns or coal power plants.

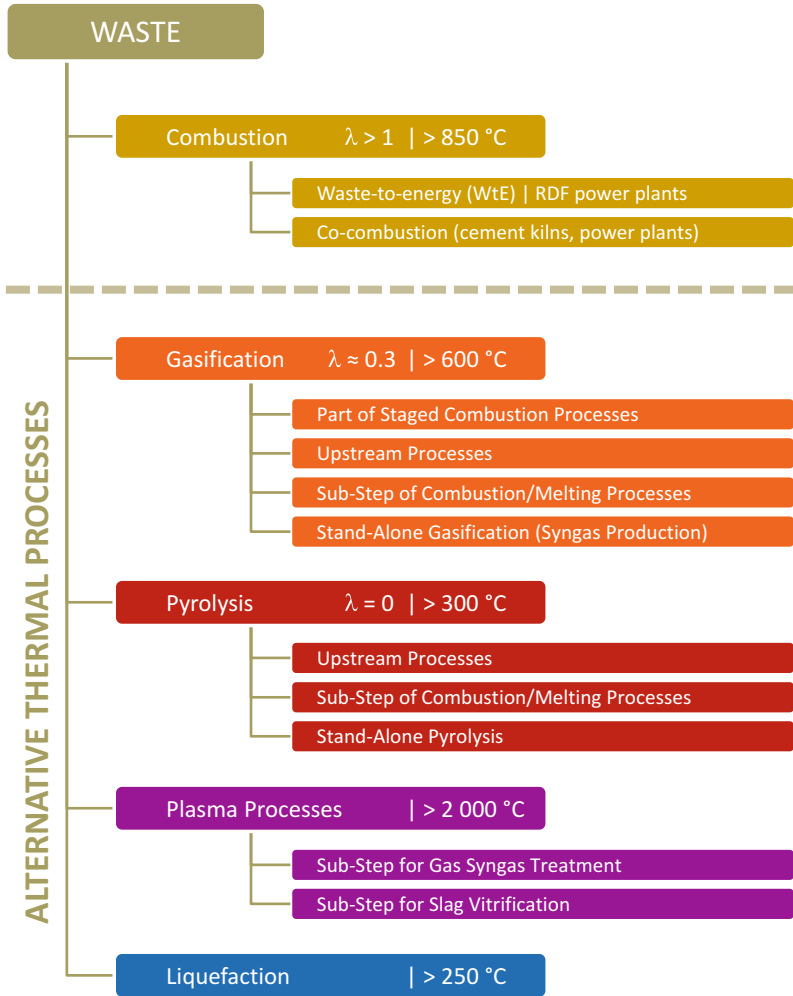


Fig. 1 Overview of thermal processes for waste treatment [1]

The possibilities for material recovery from WtE plants are presented in Sect. 2. The options are the utilization of fractions from bottom ash (especially metals) and the generation of valuables (HCl, gypsum, and zinc) from the flue gas.

Material recovery by the co-combustion of waste is not discussed deeper in this chapter. In such facilities, the material utilization of the co-combusted waste (e.g., RDF, sewage sludge, animal meal, waste oil, etc.) is restricted to its ash content, which ends in both co-incineration processes as an additive in cement production. In cement kilns, (preprocessed) waste is used as a solid recovered fuel (SRF) to supply the heat for the energy consuming burning of the clinker. The containing ash remains in the kiln and becomes a part of the product. The fly ash from coal power

plants is normally used as an aggregate for mixing with the cement clinker. In both cases, the share of the mineral fraction in the final cement product originating from the waste is marginal. Therefore, the composition of the waste does not normally influence the product properties if reasonable quality management for SRF and RDF is provided.

In Fig. 1, the so-called alternative thermal waste treatment technologies are also listed. The adjective “alternative” in this context refers to incineration. That means all thermal processes which are not combustion (i.e., which are not operated with excess oxygen) are labeled with this term. Gasification and pyrolysis are the most popular “alternatives.” More novel approaches are plasma processes, operated at very high temperatures, which are generated by the use of electricity, or liquefaction procedures, which are supposed to supply high quality liquid fuels from solid waste by conversion in an oily liquid medium. Due to their limited technical relevance for waste treatment in Germany and Europe (which is supposed to also be the case in the future), the options to recover material products from gasification, plasma processes, and liquefaction are not discussed further here. A detailed evaluation of those processes can be found elsewhere [1].

In contrast to the aforementioned alternative thermal waste treatment technologies, processes on the basis of pyrolysis offer interesting options for material recovery from special fractions (but not from MSW). This thermochemical approach is an efficient tool for the treatment of composite structures with valuables in a matrix of other components (normally plastics or resins). The plastics can be volatilized at higher temperatures and the emerging pyrolysis gases can be combusted to generate the heat for the process (condensation and material utilization of the pyrolysis liquids is not recommended due to the difficult processing and poor properties of such liquids). The remaining valuables, e.g., metals, carbon fibers, etc., may be of high quality due to the inert atmosphere during treatment in the absence of oxygen. Examples for the recovery of valuables from composite waste fractions by pyrolysis are given in Sect. 3.

2 Material Recovery from Waste-to-Energy Plants

The principle structure of all state-of-the-art WtE plants is similar (cf. Fig. 2): MSW and commercial waste is delivered by truck (sometimes also by train or ship) and dumped into an underground bunker. The waste is mixed and fed via crane and feed hopper into the furnace. The vast majority of the WtE plants are equipped with grate furnaces. Forward and backward pushing configurations as well as roller grate systems are in use. Bottom ash and raw flue gas, including solids and gaseous pollutants, are the two product streams out of the incinerator, which offer the potential for material recovery. Typical compositions of residues from MSWI are given in Table 1.

The bottom ash can be discharged from the furnace by wet or dry operated systems (cf. Sect. 2.1.1) and the subsequent processing can be operated wet or dry

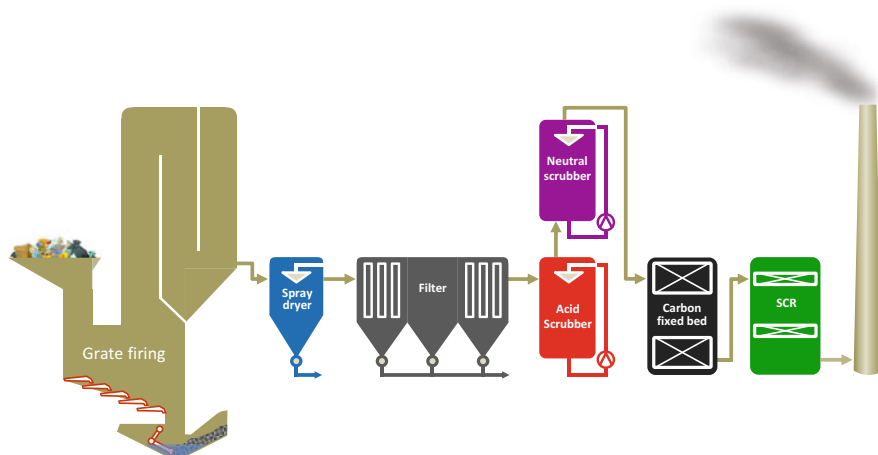


Fig. 2 Diagram of a typical waste-to-energy (WtE) plant with wet flue gas treatment system and selective catalytic reduction (SCR)

Table 1 Composition (main components) of residues from MSWI in wt.-% (dust collector, wet scrubber, milk-of-lime neutralization, and scrubber effluent evaporation [3])

Components	Bottom ash	Ash and dust from boiler and flue gas cleaning system	Reaction salts from scrubber effluent evaporation
Al ₂ O ₃	5.7–8.1	5.1–18.0	2.1–3.1
CaO	8.7–21.3	6.5–30.0	29.4–46
Fe ₂ O ₃	3.0–14.2	1.6–6.5	1.1–1.3
SiO ₂	45.7–60.1	12.5–54.7	5.0–5.1
Chloride	0.2–0.3	6.5–8.2	17–32 (26–50% CaCl ₂)
Sulfate	0.1–2.7	2–4	4.3–15.0 (18–64% CaSO ₄)

also. The processing goals are the recovery of iron and nonferrous metals, as well as at the production of mineral fractions, which can be used as construction materials. The processing of bottom ash and the subsequent recovery of valuables are discussed in Sect. 2.1.2.

The heat from the flue gas is recovered by a water tube boiler and the gas is thereby cooled down to temperatures of about 180–230°C. Subsequently, the pollutants – heavy metals, organic substances, acidic gases, nitrogen oxides (may also be reduced by selective non-catalytic reduction (SNCR) already in the boiler), and dust – are eliminated from the flue gas before they are released into the atmosphere through a chimney. The installations for flue gas cleaning can be classified as dry, conditioned dry, and wet systems, as defined in Fig. 12. The prerequisite for the recovery of products from the flue gas – hydrochloric acid, gypsum, or metals from the flue dust – is the application of a wet cleaning system, as it is depicted in Fig. 2. The possibilities for material recovery from the flue gas of MSWI are described in Sect. 2.2.

RDF power plants are technically configured in a very similar manner as WtE plants. Regarding the furnace, some fluidized bed incineration systems are in operation, but most installations are equipped with grate firing. The character of the ash from both furnace types and the methods for processing them are similar to those of WtE plants. Usually the metal and mineral content in RDF is lower compared to MSW because of the preprocessing of the RDF (which implies the separation of metals), but the quality of the products is comparable to WtE plants. Recovery of products from the flue gas cleaning systems is not common in RDF plants because these plants are normally equipped with (economically advantageous) dry flue gas cleaning systems.

2.1 Material Recovery from Bottom Ash

The material utilization of bottom ash from waste incineration is almost as old as waste incineration itself. The first German waste incineration plant, put into operation in Hamburg Bullerdeich in 1896, was already equipped with a magnet to separate the iron (which was sold for 15 Reichsmarks per ton) from the bottom ash. But not only the metal was recovered in these old times, the mineral fraction was utilized also. This was common practice in plants over the whole of Europe (cf. Fig. 3). The city council of Brno, for example, decreed in the beginning of the twentieth century that the mineral fraction from the local waste incineration plant had to be used mandatorily as a substitute for sand in public buildings [4].

The inorganic fraction in the waste amounts to about 25–35 wt.-%. Hence, bottom ash is the most relevant product stream from WtE plants [5]. In Germany

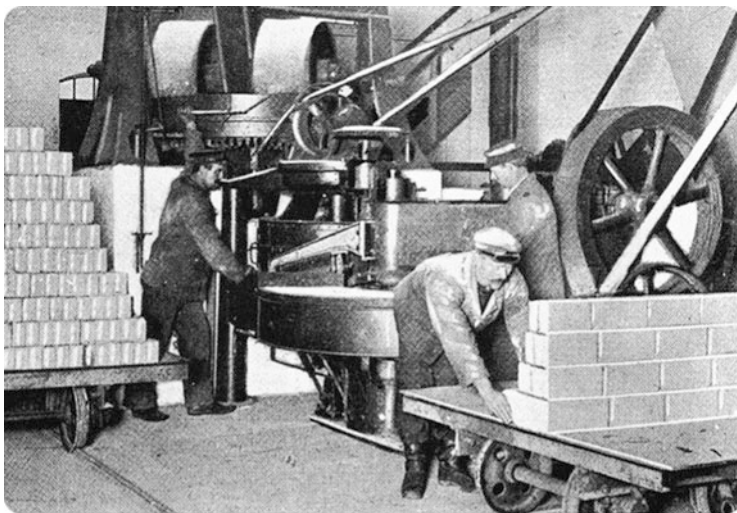


Fig. 3 Fabrication of bricks from bottom ash at the beginning of the twentieth century [4]

in 2013, about 5.35 million tons of bottom ashes were generated [6]. The average composition of this ash can be specified as follows [7]:

- 45 wt.-% ash and slag <2 mm
- 40 wt.-% course melted aggregates (oxides and silicates)
- 10 wt.-% original mineral materials (fragments from glass, ceramics, and stone)
- 6 wt.-% metals
- 1–2 wt.-% unburned components

Recent studies showed that the metal content in MSW decreased in Germany within the past three decades. Analyses carried out in the last 10–15 years resulted in metal contents of between 1 and 3.5 wt.-% of the residual waste in Germany. In commercial and bulky waste, metal content between 3 and 7 wt.-% was found. A survey among the operators of bottom ash processing plants in Germany, referring to 2014, which represented 4.4 million tons and therefore 80% of the bottom ash generated in Germany, showed that 1.3 wt.-% of nonferrous metals and 7.7 wt.-% iron scrap could be recovered on average. The amount of unburned material amounted to 0.9 wt.-% [8].

Despite the decreasing metal content, an extensive treatment of the bottom ash, with a focus on the recovery of the metals, is nowadays state of the art. Especially in the last decade, the technologies for the recovery of metals have made great progress. The processes became more and more sophisticated and the treated grain sizes smaller (down to 0.25 mm). Some new processes were developed, which enhanced the metal recovery by applying crushing steps to destroy the agglomerates in the ash. This results in the disintegration of the mineral fraction, which on the other hand impairs the building properties of this material.

The following sections give an overview of technologies for bottom ash discharge and processing. The current state is described below and some interesting new approaches are presented.

2.1.1 Bottom Ash Removal

The state of the art and therefore applied in the vast majority of the WtE plants installed worldwide is the wet discharge of the bottom ash. In Switzerland and also in Japan, some plants are operated with dry discharge systems, which are supposed to enhance the quality and quantity of metal recovery.

Wet Bottom Ash Discharge

Wet bottom ash discharge means that the ash falls from the grate directly into a water bath (cf. Fig. 4). The water bath has two functions: it cools the hot ash and seals the furnace from the ambience to keep the desired underpressure within the combustion chamber. Further advantages of the wet operation are the prevention of dust during ash handling and the destruction of sintered agglomerates by the rapid

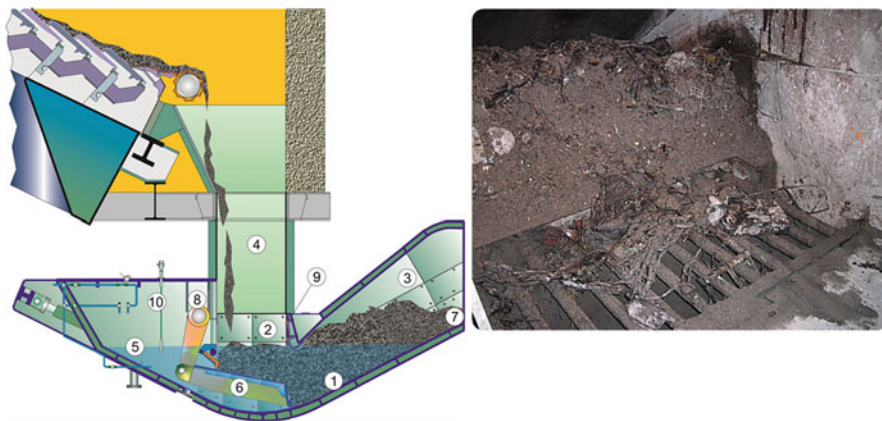


Fig. 4 *Left:* scheme of a wet type bottom ash ram discharger. 1 – Discharger tub, 2 – Inlet section, 3 – Outlet chute, 4 – Connecting piece, 5 – Water level, 6 – Discharge ram, 7 – Drop-off edge, 8 – Drive shaft, 9 – Air sealing wall, and 10 – Electrically controlled level metering system (Copyright Martin GmbH für Umwelt- und Energietechnik, Munich). *Right:* outlet of a wet type bottom ash ram discharger with screen for the separation of coarse material (Photo Peter Quicker)

quenching of the material (result of thermal tensions between cool water bath and hot agglomerates). Disadvantageous is the initiation of hydration, sulfatization, salt building, and solution reactions from the contact with water. This results in a solidification (hydraulic reactions) of the ash, which hampers the following processing.

The installed bottom ash discharge systems differ in the way they remove the ash from the water bath. Plate and chain conveyors are possible solutions, but predominantly ram dischargers are applied (cf. Fig. 4). The ram (no. 6) periodically pushes the ash through the outlet chute (no. 3) out of the water bath into a container, or onto a conveyor belt (the photo shows a rod screen to separate coarse material). During its residence time at the “drop-off-edge” of the discharger (no. 7), the material is dewatered. The water level in the tub has to reach the air sealing wall (no. 9) to ensure the tightening of the furnace.

Dry Bottom Ash Discharge

The first trials with dry bottom ash discharge were carried out in the 1990s by the companies ABB and Martin GmbH. In Japan and also in Switzerland (Hinwil, Monthey, Zurich, Horgen, and in Zuchwil, in combination with a wet discharge), dry discharge systems are in industrial operation. The companies have implemented different approaches to cool down the hot ash and to handle the strong dust formation.

Figure 5 shows the dry discharging system applied in the KEZO Hinwil WtE plant in the region of Zurich. A central element of the ash removal system is a vibrating channel [9, 10]. After burnout, the ash falls down from the grate into the channel. The dropping impact results in a crushing of agglomerates. The ash is

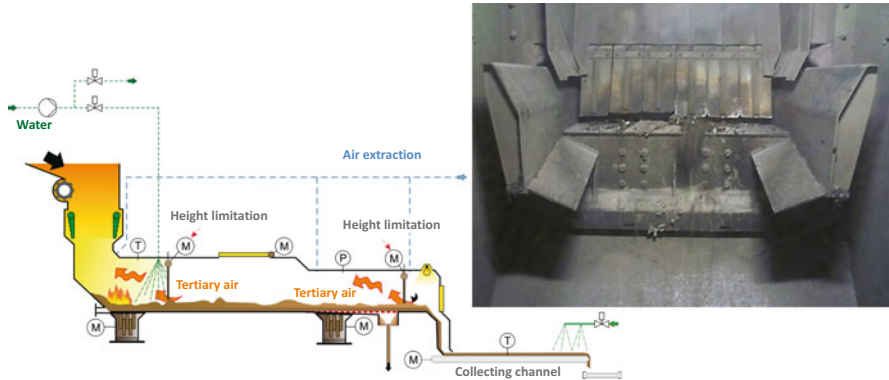


Fig. 5 *Left:* scheme of the dry bottom ash discharger in Hinwil, Switzerland (Copyright KEZO). *Right:* outlet of the dry bottom ash discharger (Photo Peter Quicker)

transported through the channel onto an integrated 5 mm screen by vibration and is cooled down by a countercurrent airstream [9, 11]. After passing the vibrating channel, the air enters the furnace and substitutes about 10% of the combustion air [12]. This so-called tertiary air has multiple functions [9, 12]:

- The bottom ash is cooled and the air concurrently warmed up. The sensible heat of the bottom ash is hereby recovered and reverted to the furnace. In comparison to the wet discharge process, where the heat is lost through the cooling of the ash in the water bath, the thermal efficiency of the process is (slightly) increased.
- The air oxidizes unburned material in the bottom ash. Typical TOC values are below 0.3%.
- The air generates a screening effect and transports fine particulates back into the furnace.

Advantages named by the operator are the saving of approximately 70–100 L of water per ton of waste, the reduction of the total mass of the bottom ash of about 20 wt.-%, because of the absence of water, and – as the main point for the following ash processing and metal recovery – the creation of better bulk material characteristics (no hydraulic solidification reactions) [9].

Other systems for the dry bottom ash discharge were developed by the companies Martin GmbH and Hitachi Zosen Inova (HZI). Whereas HZI also uses a channel for discharge, the Martin system combines an air classifier with a dry operated ram discharger, like the one depicted in Fig. 4 [13–17].

2.1.2 Bottom Ash Processing

The following sections (sections “[State of the Art](#)” to “[Optimization of Mineral Fraction](#)”) refer to the processing of wet discharged bottom ash. The first section gives an overview of state-of-the-art technology for the dry and wet processing of wet discharged ash. The two following sections highlight the possibilities to optimize the recovery of metals and minerals. Finally, section “[Processing of Dry Discharged Bottom Ash](#)” deals with the processing of dry discharged ash.

State of the Art

Bottom ash processing in Germany is undertaken by some of the WtE plant operators by themselves, but to a greater extent by independent, external bottom ash processing companies. State of the art and applied in most of the processing facilities are dry operated systems. The main focus of the operators is metal recovery. Therefore, a multitude of separation steps is applied, e.g., up to 12 separators for nonferrous metals in one facility (maximum value). In Fig. 6, an exemplary process

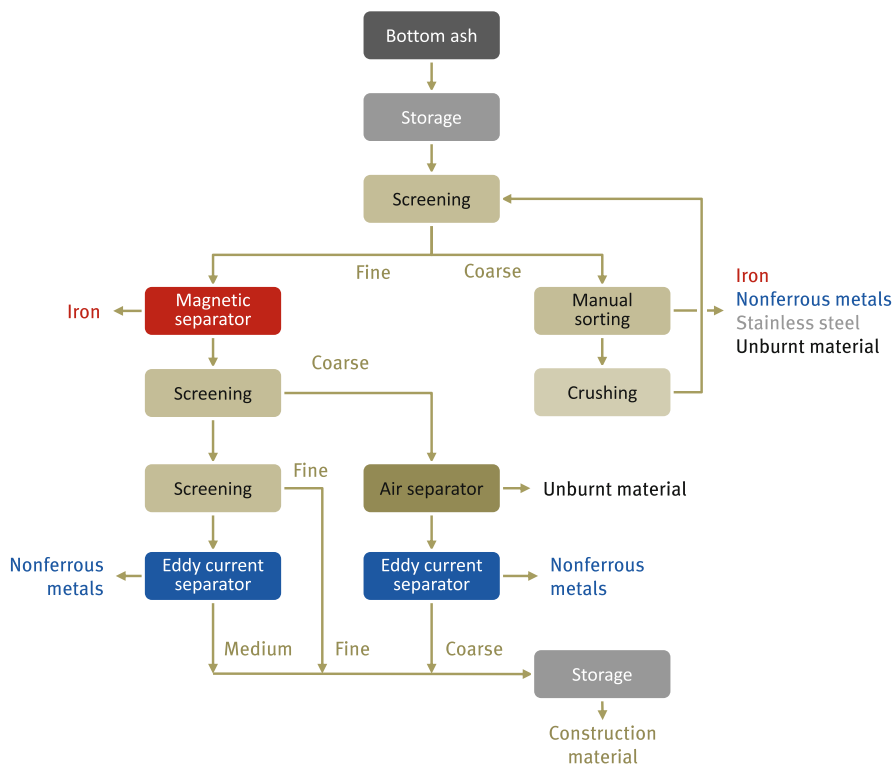


Fig. 6 Exemplary plant setup for the processing of wet discharged bottom ash from municipal solid waste incineration (MSWI) on the basis of [2, 11, 18–21]

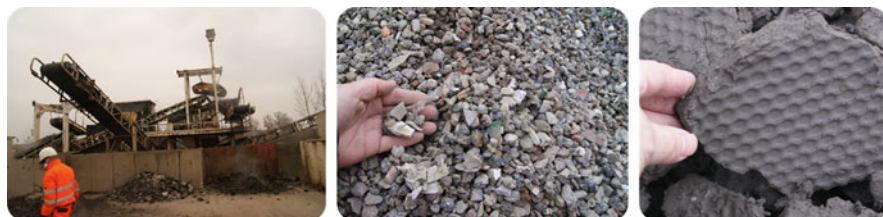


Fig. 7 Bottom ash processing plant of the company Scherer und Kohl in Ludwigshafen. *Left*: processing machinery, *middle*: produced gravel fraction, and *right*: sludge filter cake (Photos Peter Quicker)

scheme for dry bottom ash processing is given. Modern plants create more grain-size fractions because the efficiency of metal recovery can be increased by narrow grain-size corridors. Further information about details of bottom ash processing technologies can be found elsewhere [18, 22].

Only few facilities in Germany are applying wet processes for bottom ash treatment. The company Scherer und Kohl in Ludwigshafen has a long tradition in this field. The company's process comprises a dry preconditioning and a wet processing of the fine fraction smaller than 22 mm. Firstly, iron scrap and coarse components (>56 mm) are separated. Afterward, the remaining material (predominantly minerals) is crushed by an impact mill to grain sizes below 22 mm. This material is further treated in the wet processing part of the plant. Products – besides iron and nonferrous metals – are substitute construction materials with defined grain sizes (2–5, 5–8, and 8–16 mm) and high quality (cf. eluate parameters “S&K” in Table 2). About 70% of the whole material can be used as construction material. Only about 6–7% of the inputs have to be landfilled as sludge [28] (Fig. 7).

Optimization of Metal Recovery

As already mentioned, metal recovery is the main target for bottom ash processing, because the metals offer an additional income for the operators besides the tipping fee for accepting the ash. Against this background, it is not surprising that some new processes with enhanced metal yields have been developed within the last few years.

The Advanced Dry Recovery (ADR) process was developed in a cooperation between the Technical University of Delft and the company Inashco BV. The innovation of this process is a dry mechanical fractionation step by application of a rotating drum. It could be demonstrated that the aluminum recovery rate of the process is significantly higher compared to conventional technology. In the meantime, several industrial ADR plants are in operation, treating 2.5 million tons of bottom ash altogether per year [29–31].

Table 2 Eluate parameters of bottom ash dependent on discharging type, state of aging, and way of (further) processing

State of aging	Fresh			Aged			Processed			Rules	
	Type of discharging	Wet	Dry	Wet	Dry	Wet	Wet	S&K	Wet	LAGA	NRW
Processing		n.s.	2008	2005	2008	2005	2004	2004		2003	2001
Parameter	Unit	[23]	[23]	[24]	[23]	[24]	[26]	[26]		[27]	[24]
pH value		11.6	11.9	11	10.3	10.3	10.3–11.9	bid	n.s.	7–13	7–13
Conductivity	mS/cm	5	6	1.5	2.2	2.2	0.7–3.4	bid	0.27	6	2
DOC	mg/l	14	26	16	16	16	3.5–140	bid	n.s.		
Chloride	mg/l	150	650	120	260	260	68–335	29	22	250	50
Sulfate	mg/l	330	460	250	370	370	95–609	43	n.s.	600	200
Cyanide tot.	µg/l	60	20	5	12	12	k.A.	bid	n.s.		
Arsenic	µg/l	4	4.5	10	13	13	<10	1.3	<10		
Lead	µg/l	1,500	450	2,000	167–1,030	12	5–286	bid	<10	50	50
Cadmium	µg/l	2	n.s.	13	<0.2–1.5	10	0.3–1	bid	<1	5	5
Chrome tot.	µg/l	23	30	25	41– 123	43	5–14	bid	<10	200	50
Copper	µg/l	150	450	500	26.4–337	250	10–236	bid	30	300	
Nickel	µg/l	30	10	15	3.3–11.1	10	4–8	bid	<20	40	
Mercury	µg/l	0.6	1.3	0.85	< 2	0.15	0.1–0.2	bid	0.2	1	1
Zinc	µg/l	250	500	60	106–200	20	10–40	bid	40	300	300

bid below limit of detection, *n.s.* not specified, *S + K* wet bottom ash processing according to the process of the Scherer + Kohl company, Ludwigshafen, *SYN+* processing according to the SYNCOM-Plus process of the Martin GmbH company, Munich, *LAGA* LAGA technical bulletin (Merkblatt) M20, *NRW* recovery decree of North Rhine-Westphalia

Another product of corporative research is the ATR¹ process, developed by several industrial, public, and academic partners. This concept is based on the application of a high-velocity impact crushing device (impact velocities of 800 km/h), which allows the disintegration of the agglomerates and thereby the recovery of the embedded metals. More than 50% of the material is crushed to get sizes smaller than 2 mm by this procedure [32–35].

Other approaches to enhance the metal recovery from MSWI bottom ash include the VeMRec-process, developed by the Institute for Recycling at RWTH Aachen University [36] or the ReNe-process, designed by the Technical University of Clausthal [37, 38].

Optimization of Mineral Fraction

The mineral fraction in the bottom ash shows parameters which hamper the utilization of the material for construction purposes. Besides structural properties, influencing the construction stability, particularly the elution behavior of the material, is of high importance. Only if the legal requirements are fulfilled and the elution values of heavy metals fall below the limits, an application of the material in construction is possible.

Table 2 shows typical eluate values for different types of bottom ash: wet and dry discharged, fresh and aged as well as processed by a wet system (Scherer und Kohl, cf. section “State of the Art”) and “pretreated” by the SYNCOM-Plus process (see below). For comparison, the limits from two German regulations are also given.

All values exceeding the limits of at least one of those regulations are marked in bold. Wet and dry discharged, as well as fresh and aged bottom ashes without processing, show several values higher than prescribed in the regulations. It is clearly visible that only the processed mineral fractions can fulfill the rules. Therefore, influencing the elution behavior of bottom ash from MSWI was and is a topic of research and development. There are several possibilities for enhancing the elution behavior of bottom ash besides the already discussed wet processing. Solidification processes use binder materials to immobilize the heavy metals. Sintering and melting processes apply higher temperatures to induce structural modifications in the mineral matrix to reach the same goal.

A thermal treatment subsequent to the incineration process, with the already cooled bottom ash, is very energy demanding. It is more reasonable from an energetic point of view, to design the incineration process in a way that allows the melting or sintering of the bottom ash directly in the furnace. An example of such an approach is the SYNCOM process, developed by the company Martin GmbH and realized in the MSWI facility in Arnoldstein, Austria (Figure 8 shows the scheme of the SYNCOM-Plus process, which consists of an integrated washing step for bottom ash in addition to the sintering step). The necessary sintering temperature of 1,150°C is reached by

¹ATR for German: Aufschluss (disintegration), Trennung (separation) and Recycling.

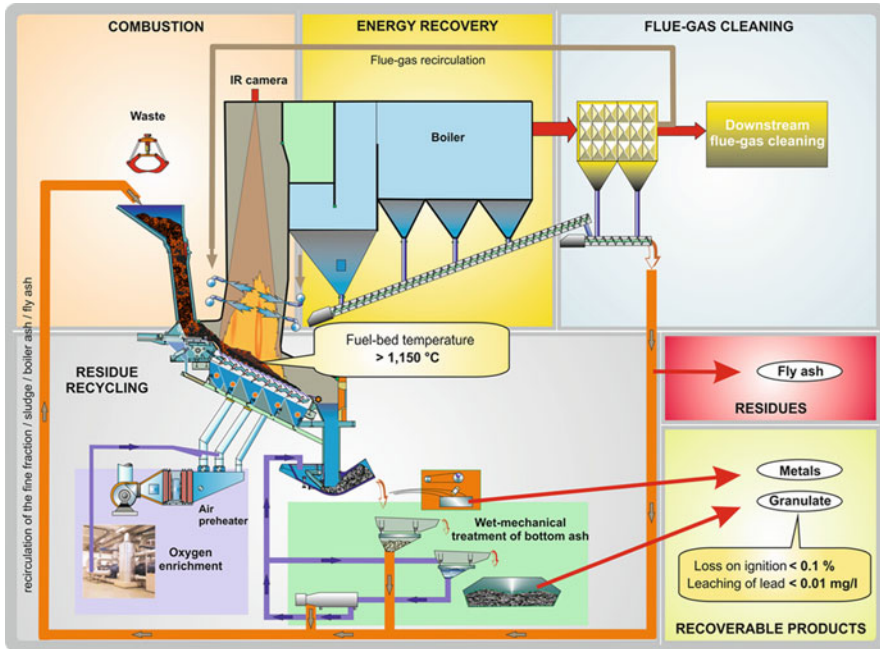


Fig. 8 Scheme of the SYNCOM-Plus process, designed by MARTIN GmbH für Umwelt- und Energietechnik, Munich (Copyright Martin GmbH)

enrichment of the combustion air with oxygen. The sintering process results in a reduction in the fine fraction in the bottom ash and decreases the extractability of the heavy metals and anions from the mineral fraction (cf. Table 2) [39, 40].

Processes with integrated washing steps focus on the leaching of the heavy metals instead of a demobilization. An example is the already discussed process of the Scherer und Kohl company (cf. section “State of the Art” and Table 2). An elution of the heavy metals can also be carried out directly into the wet operated bottom ash discharger if it is operated with increased water throughput. It is thereby possible to reduce the chloride and sulfate eluate concentrations by about 50% [41].

A washing step is also included in the SYNCOM-Plus process (cf. Fig. 8) in combination with the already explained sintering step (SYNCOM process). The partly sintered bottom ash is washed, screened, and a granulate with a high leaching stability is produced (cf. Table 2). During screening and washing, a fine fraction and a sludge are generated. Both are recycled to the bunker and fed into the incineration process again to form agglomerates, and thereby reduce the share of fine particles in the bottom ash [40]. The granulate product as well as the fine fraction and the sludge are pictured in Fig. 9.



Fig. 9 Granulate product from SYNCOM-Plus process (*right*) and sludge, as well as fine fraction for recycling in the incineration process (Photos Peter Quicker)

Processing of Dry Discharged Bottom Ash

The development and implementation of new dry operated bottom ash discharge systems also necessitates new processing approaches for the ash. As for the dry discharge systems, in Europe, these facilities can only be found in Switzerland.

The dry discharged bottom ash in the MSWI plant in Monthey is processed in the same facility formerly used for the wet discharged bottom ash. Only the eddy current separators had been optimized by the operator [14]. It is obvious that a further adjustment of the processing technology would result in an increase in the metal yield. The operator intends to realize this potential [14].

In the incineration plant in Hinwil (KEZO), in contrast, a totally newly developed processing system for the dry discharged ash was installed. Through the application of conventional but optimized processing steps, it was possible to treat bottom ash fractions down to 0.2 mm. It was stated by the operator that a 90% recovery of nonferrous metals could be reached [9, 42, 43].

The question currently discussed, whether dry bottom ash discharge can enhance the yield and quality of the recovered metals from bottom ash in comparison to wet discharge systems, was investigated in a project funded by the German Federal Environmental Agency [2]: In the WtE plant in Mainz, the wet and dry (the normally wet operated ram discharger was run without water filling) discharge of bottom ash was realized on two subsequent days at the same line, and 10 tons of bottom ash were extracted each day. Both materials were processed according to the state of the art (top belt magnetic separator, magnetic drum separator, and eddy current separator), with special focus on a deep fractioning of the material

(fractions: 0–2 | 2–4 | 4–10 | 10–30 | 30–80 | >80 mm), to enhance the metal recovery (Fig. 10).

It could be shown that by application of the same processing and separation steps, the share of nonferrous metals that could be recovered from the bottom ash was significantly higher for the dry discharged material. Furthermore, the qualities of the metals were better because of the absence of products from oxidation processes and hydraulic reactions (cf. Fig. 11).

Nevertheless, it has to be mentioned that similar yields may also be extracted from wet discharged material with special adapted technologies. This is at the moment being investigated in a project in Switzerland [44].

2.2 Material Recovery from Flue Gas

The second product stream of waste incineration, besides the bottom ash, is the flue gas. It is possible, and has already been realized, to recover resources from this output stream as well. Valuable materials can be recovered from the particles (dust) as well as from the gaseous components. Table 3 shows the concentration ranges of the main components in the raw gas of waste incineration facilities.

Dominant components in the flue gas are particles (dust), HCl, and sulfur dioxide. The recovery and material utilization of HCl and sulfur (in the form of gypsum) has been practiced industrially for decades (cf. Sects. 2.2.2 and 2.2.3).

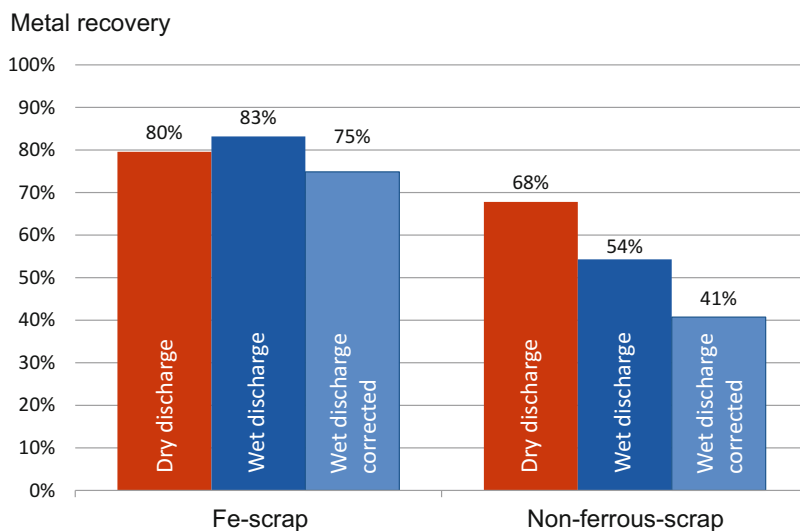


Fig. 10 A comparison of metal yield through the mechanical processing of dry and wet discharged bottom ash. According to the lower product quality, the yields of metals from wet discharge were corrected by an empirical factor (Fe: 10%, NE: 25%), according to the experience of the authors [2]



Fig. 11 Iron product from dry (*left*) and wet (*right*) discharge of MSWI bottom ash

Table 3 Main components in raw and clean gases of waste incineration plants (daily average values)

Component	Raw gas [mg/m ³ _{i.N., 11% O₂, dry}]	Clean off gas [mg/m ³ _{i.N., 11% O₂, dry}]
Dust	600–5,000 (15,000)	0.1–2
Total organic carbon	10–40	0.1–2
Carbon monoxide (CO) ^a	2–30	
Hydrochloric acid (HCl)	400–2,000 (5,000)	0.02–7
Hydrofluoric acid (HF)	2–30	0.02–0.5
Sulfur dioxide (SO ₂)	100–1,500	0.1–30
Nitrogen oxides (as NO ₂)	200–500	30–190
Mercury (Hg)	0.1–1	0.0002–0.01
PCDD/PCDF [ng/m ³ _{i.N., 11% O₂, dry}]	3–6 (15)	0.001–0.05

^aCarbon monoxide is not reduced within the flue gas treatment and therefore raw and clean gas values do not differ

New approaches focus on the recovery of metals from the dust in the flue gas. The dust is a variable mixture of different particulate components and contains noticeable amounts of metals that can be recovered (cf. Sect. 2.2.4) (Table 4).

2.2.1 Flue Gas Cleaning Systems

As already mentioned, a variety of different flue gas cleaning systems for WtE plants exist regarding the separation of the acidic gases. Figure 12 gives an overview of typical solutions (according to VDI 3460 [3]).

Dry and conditioned dry flue gas cleaning processes have the common disadvantage that dust and flue gas cleaning residues are collected at the same point (dust filter). This complicates an effective recovery of resources.

Wet flue gas treatment systems normally consist of an acid and a neutral washing stage. This allows the stepwise and more or less selective extraction of chlorine and

Table 4 Composition of the metal fraction in filter dust from Swiss MSWI plants [45]

Component	Mass concentration [mg/kg DM]			Mass concentration [mg/kg DM]	
	Minimum	Maximum		Minimum	Maximum
Aluminum	25,000	45,000	Magnesium	6,000	18,000
Antimony	700	5,000	Manganese	400	900
Arsenic	20	120	Sodium	25,000	70,000
Barium	50	200	Nickel	60	300
Lead	7,000	25,000	Phosphor	1,000	8,000
Cadmium	150	1,000	Mercury	0	3
Calcium	100,000	250,000	Sulfur	30,000	150,000
Chlorine	40,000	150,000	Silver	30	60
Chrome	100	500	Silicon	50,000	100,000
Cobalt	20	100	Titanium	2,500	5,000
Iron	18,000	55,000	Zinc	20,000	120,000
Potassium	30,000	60,000	Tin	800	3,000
Copper	1,500	5,000			

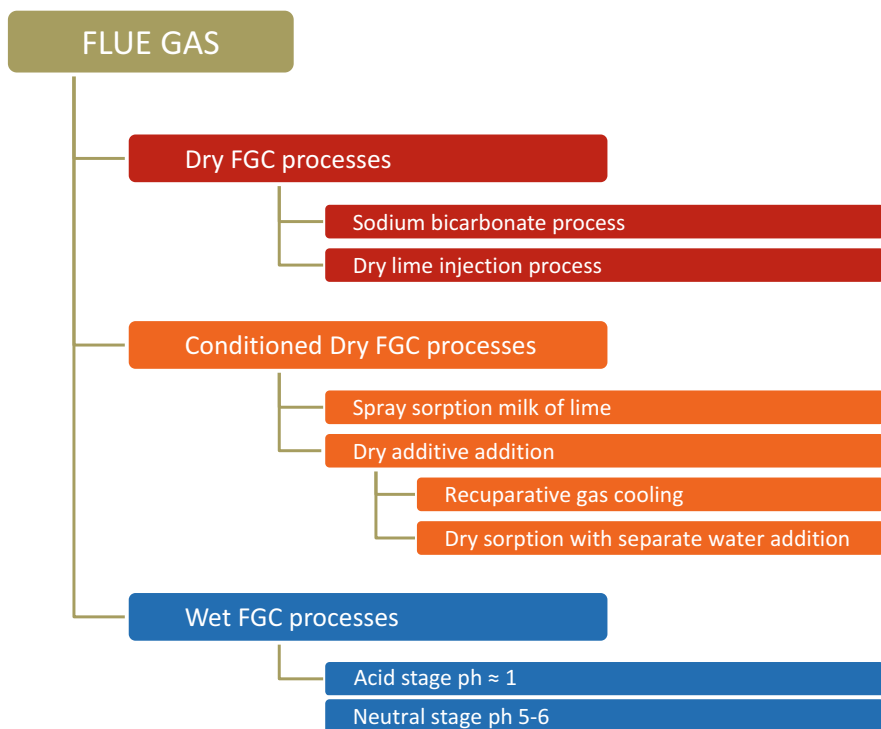


Fig. 12 Classification of flue gas cleaning systems in waste incineration plants according to VDI guideline 3460 Emission control – Thermal waste treatment [3, 46]

sulfur from the flue gas as a basis for the further material utilization of these components.

Further information regarding the flue gas cleaning systems of solid waste incineration plants can be found elsewhere [46]. The following paragraphs give a short overview of the currently existing and applied technologies for the recovery of valuables from the flue gas of WtE plants.

2.2.2 Recovery of Hydrochloric Acid

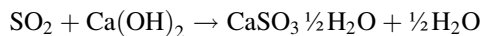
The requirement for the recovery of HCl from flue gases of municipal solid waste incineration (MSWI) is the deposition of the dust prior to the scrubber. Therefore, normally electrostatic precipitators are applied, but baghouse filters can also be used.

The absorption of HCl takes place in a wet scrubber at an acidic pH value. Figure 13 gives an impression of a three-stage HCl scrubber. In the first step, the remaining dust particles, gaseous heavy metal salts, and some HCl is absorbed. Furthermore, the flue gas is cooled down to saturation temperature.

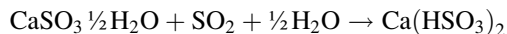
The main HCl absorption is carried out in two packed beds mounted one upon the other in the same column. Both beds are equipped with a separate circulation for the washing liquid. A countercurrent flow between the flue gas and the washing liquid is applied to realize a high concentration of HCl in the scrubbing solution. The product from the scrubber is a raw hydrochloric acid solution with about 15% HCl, which is further concentrated in downstream refining steps. The salt solution from the first quenching step has to be evaporated, e.g., together with the residues from acid refining, and the remaining solids are disposed of [47, 48].

2.2.3 Recovery of Gypsum

Because of the acidic pH value in the HCl scrubber, no absorption of sulfur dioxide takes place there. This happens in a second scrubber (cf. Fig. 14) with a higher pH value in the washing medium. To keep the pH value in the neutral region, the addition of a neutralization agent, normally limestone powder or hydrated lime, is necessary. If hydrated lime is used, the following chemical reaction takes place in the scrubber water:



The emerging calcium sulfite can react to calcium hydrogen sulfite:



And the hydrogen sulfite can be converted to gypsum by reacting with oxygen which may be present in the flue gas or is also injected into the scrubber sump (cf. Fig. 14):

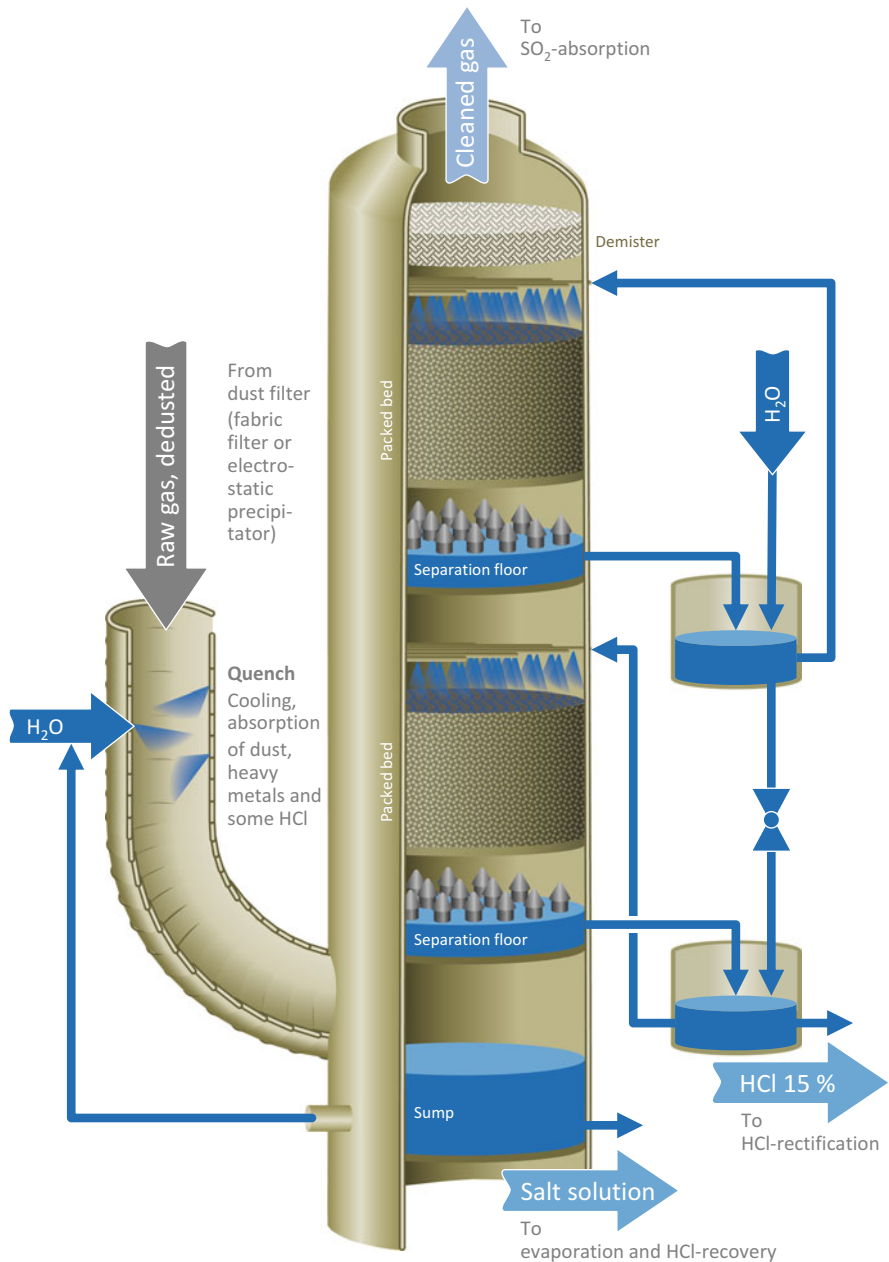


Fig. 13 Wet scrubbing system for three-step absorption of hydrochloric acid (HCl) from prior dedusted flue gas from MSWI (Graphic Peter Quicker, based on [47, 48])

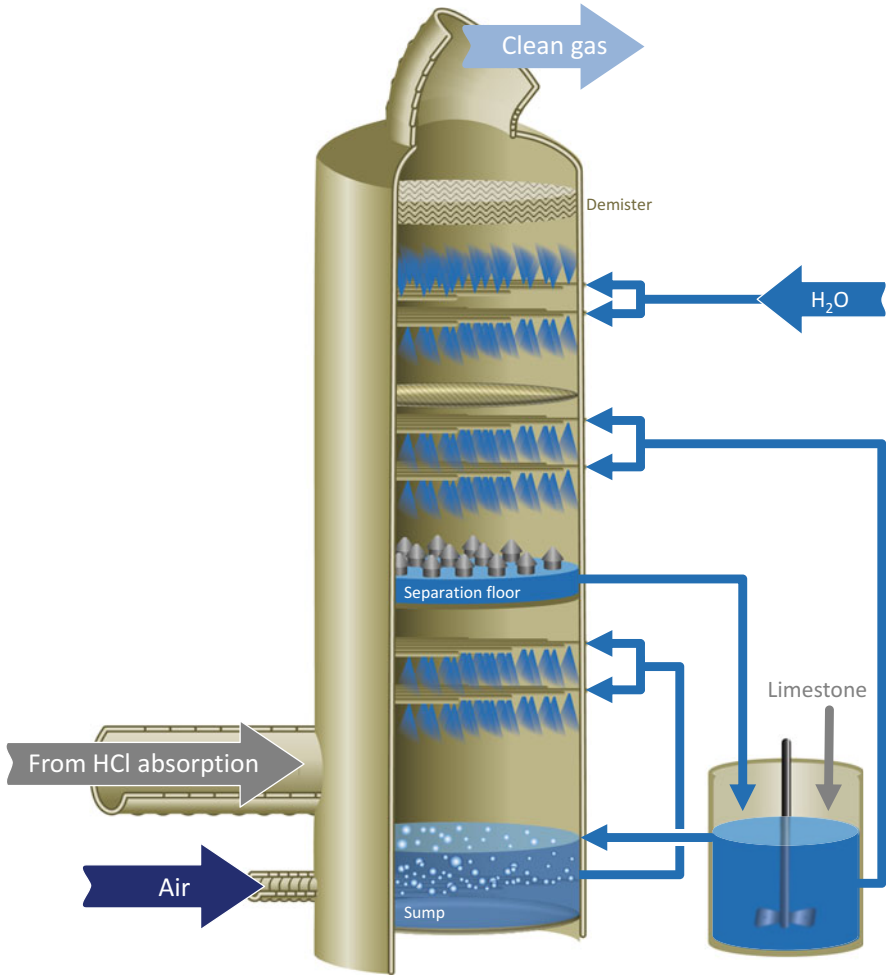
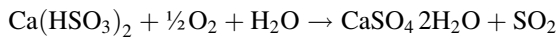
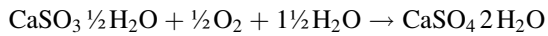


Fig. 14 Wet scrubbing system for absorption of sulfur dioxide (SO₂) from MSWI flue gas, before dedusting and cleaned from HCl (Graphic Peter Quicker, based on [47, 48])



The direct oxidation of calcium sulfite to gypsum is also possible:



2.2.4 Recovery of Zinc

An assumed typical dust concentration in the raw gas of MSWI plants of between 1 and 5 g/m³_{i.N.} (cf. Table 3) results in an annual amount of 1,100–5,500 tons of filter dust in a medium-sized MSWI plant² with a zinc content of about 50–250 tons. This potential provided motivation for the development of a new process for the recovery of zinc from the filter dust of a WtE plant that is operated at industrial scale in Solothurn, Switzerland. A scheme of this FLUREC process (German “FLUGasche RECYcling,” i.e., fly ash recycling) is depicted in Fig. 15.

The new approach is based on the so-called FLUWA process (German FLUGasche WAesche), a fly ash scrubbing, which has been practiced for years in several Swiss WtE plants. The idea behind the FLUWA process is to extract leachable heavy metals from the filter ash by using the acid generated in the plant’s own scrubbing system. Simultaneously, the scrubbing water is neutralized by the alkaline components in the filter dust. The leached and dewatered filter dust can be landfilled. The heavy metals in the scrubbing solution are precipitated with milk of lime. The resulting hydroxide sludge can be used after dewatering for zinc recovery in a smelter due to its high zinc content (>25%) [28].

The FLUREC process goes one step further and includes all necessary processing steps – wet chemical processing and finally the electrolytic deposition of the metal – to recover metallic zinc from the hydroxide sludge directly in the WtE plant with a purity of 99.99%. As a by-product, a solid residue with a lead content of about 50% is generated that can also be used for metal recovery (in an external facility). It is planned to extend the capacity of the plant in the future to treat the filter dust from all MSWI plants in Switzerland [45].

3 Material Recovery by Pyrolysis

Thermochemical conversion in the absence of oxygen offers the chance for material recovery from special waste fractions containing valuables which are embedded in a matrix of volatile matter, like plastic. These may be composite material parts, like carbon fiber-reinforced plastics (CFRP), or metal-enriched fractions from (mechanical) upstream waste treatment processes like shredder residues. The plastics contained are volatilized at higher temperatures, and the emerging pyrolysis gases can be utilized to supply the thermal energy for the process by **direct** combustion. It is emphasized that a condensation and material utilization of the pyrolysis liquids as well as of the gases is not recommended due to their difficult processing and poor properties.

²Assumptions: 200,000 tons of waste throughput, specific flue gas amount of 5,500 m³_{i.N.}/ton of waste.

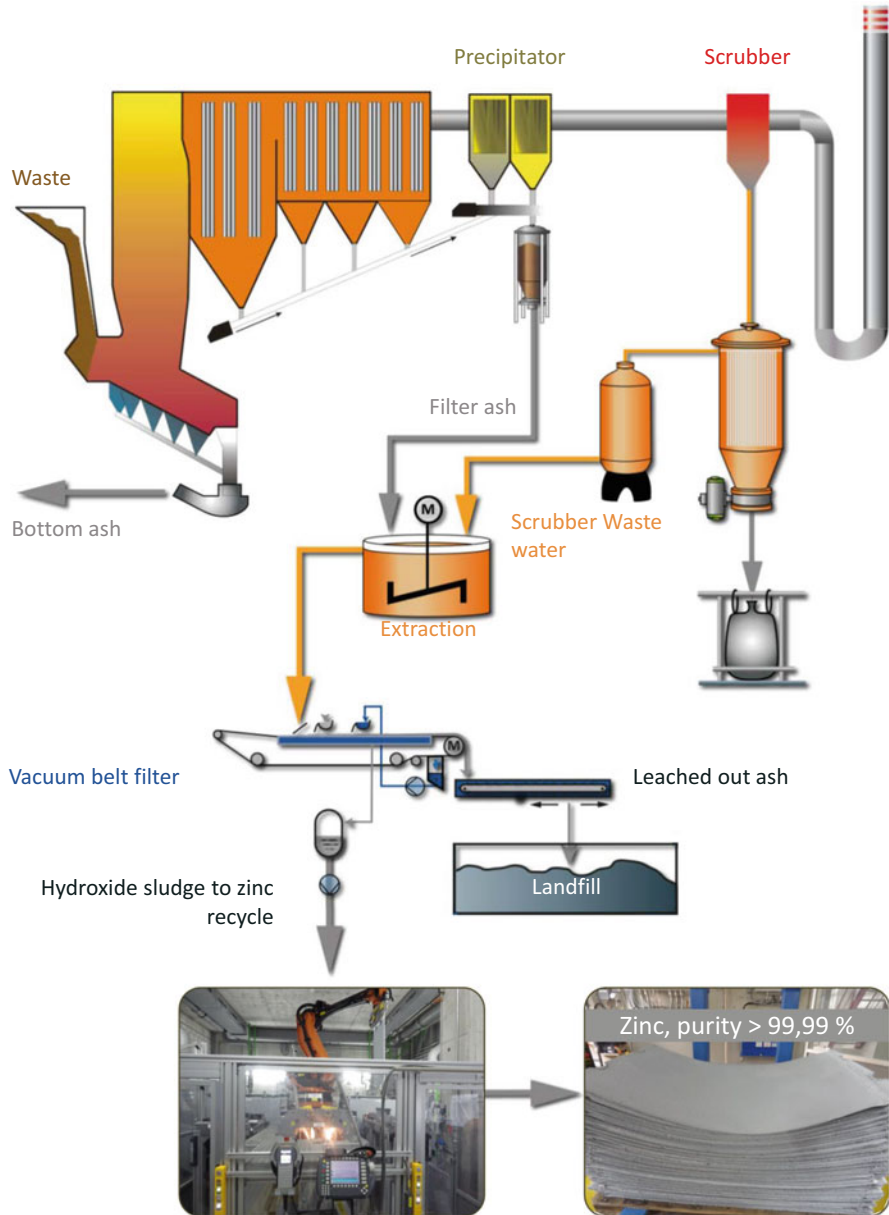


Fig. 15 FLUREC process for zinc recycling from WtE flue gas residues [28, 45]

The solid valuables remaining after pyrolytic treatment, e.g., metals, carbon fibers, etc., may have a high quality due to the inert atmosphere during treatment in the absence of oxygen. In the following, examples of the recovery of valuables from

composite waste fractions by pyrolysis are given. The investigations were carried out by the Unit of Technology of Fuels at RWTH Aachen University.

3.1 Metals from Waste of Electrical and Electronic Equipment

The objective of the investigations was the realization of high metal recovery rates from the waste of electrical and electronic equipment (WEEE) with minimized effort for additional mechanical treatment by the application of a pyrolysis step. Ferrous and nonferrous metals were the designated fractions for recovery. The composite materials were treated in a rotary kiln at 600°C in an inert atmosphere. The gaseous, liquid, and solid products were sampled and analyzed. The single steps of the material processing are depicted in Fig. 16, and detailed information can be found elsewhere [49].

The consumer electronic devices for the test runs were obtained from a local WEEE collection point in the city of Aachen. The material was shred and the composition of the material was analyzed using manual sorting:

- 12.4 wt.-% metal-plastic composites with printed circuit boards
- 23.6 wt.-% metal-plastic composites without printed circuit boards
- 47.0 wt.-% free plastics
- 10.1 wt.-% free ferrous metals
- 1.9 wt.-% free nonferrous metals
- 5.1 wt.-% fines

The high amount of plastics in the material resulted in a significant content of 49 wt.-% volatile components and an average calorific value of 24.7 MJ/kg.

After manual analysis, the material was mixed again and treated as shown in Fig. 16 by magnet and eddy current separation to enrich the metal composites prior to thermal decomposition. The pyrolysis was carried out in an electrically heated laboratory-scale rotary kiln reactor (drum diameter 162 mm, length 1,600 mm). Residence time of the solids in the reactor was between 30 and 60 min. The condensable components in the pyrolysis gas were collected in liquid form, and the volume and composition of the remaining permanent gases were measured before the gas was incinerated with a burner. The solids were screened after the thermal treatment to separate the fines, predominantly coke, from the metals.

The amount of condensates and noncondensable gases varied greatly from 5 to 10 and 1 to 10 wt.-%, respectively. The gas predominantly contained combustible components, i.e., hydrogen, carbon monoxide, methane, ethane, ethylene, and other organic hydrocarbons, but inert gases such as nitrogen and carbon dioxide were also detected.

The composition of the solid products (red columns) as well as of the input material (blue columns) for the pyrolysis step is shown in Fig. 17 (Fe-concentrate

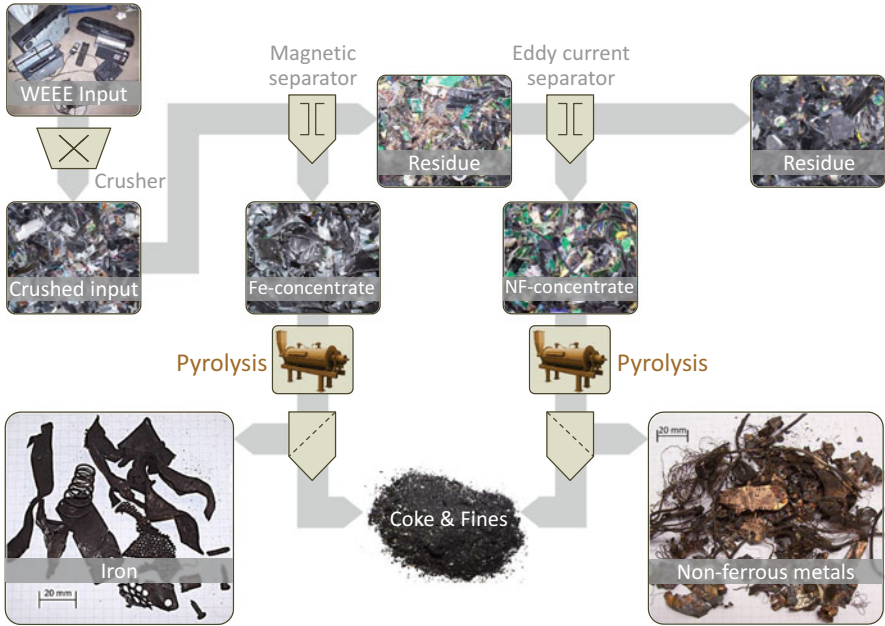


Fig. 16 Thermomechanical processing of waste of electrical and electronic equipment (WEEE) for recovery of iron and nonferrous metals (NF = nonferrous)

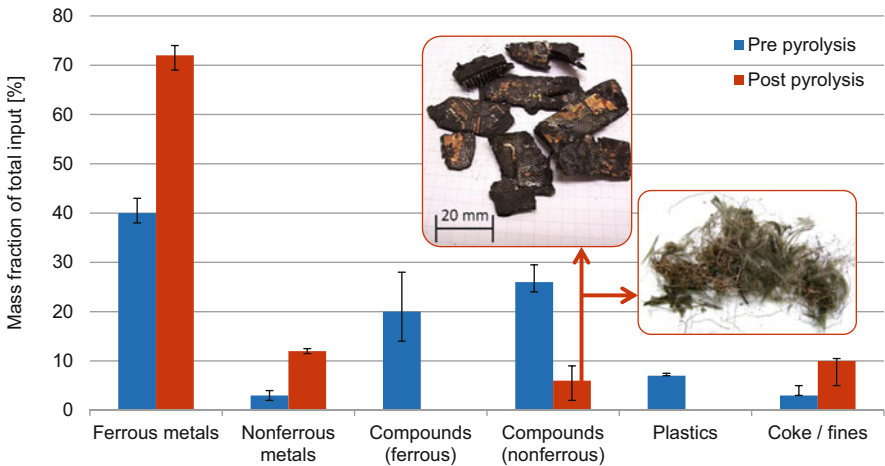


Fig. 17 Composition of solid residues after common pyrolysis of ferrous and nonferrous concentrates from mechanical processing of WEEE

and NF-concentrate together, mean values of four trials). It is clearly visible that the share of free metals (iron and nonferrous metals) could be significantly increased using the pyrolytic treatment. In the case of iron, all composites could be totally destroyed, and only free iron with no adhesions could be found in the product. Also,

the amount of nonferrous composites could be reduced. However, after all trials, nonferrous metal composites were still detected in the product. These composites primarily consisted of copper from printed circuit boards, which was interwoven with a glass fiber matrix (cf. photos in Fig. 17). The coke contained high amounts of chlorine up to 5 wt.-%, caused by the plastics (PVC) in the input material.

The concept was also successfully tested for other metal concentrates, e.g., for fractions from auto shredders or ferrous metal-enriched fractions from the processing of landfill material [49].

3.2 Carbon Fiber-Reinforced Plastics

Composite materials offer interesting properties, like high mechanical and chemical stability at a low weight. In particular, fiber-reinforced plastics are an increasing market segment and present in a multitude of different branches, like aerospace, defense, power generation (windmills), but also sports and leisure. Whereas the market is actually dominated by glass fiber-reinforced plastics, carbon fibers are a steadily growing segment. In 2016, about 46,000 tons of carbon fibers and 100,000 tons of CFRP were produced globally [50].

CFRP are composite materials with carbon fibers that are embedded in a polymer matrix. The production is very complex and energy intensive, resulting in production costs of 20–100 €/kg. This makes the recycling of these materials – that means the recovery of clean fibers to use with a new matrix for the production of new composites – economically very interesting. However, the recycling of CFRP is a challenge.

To separate the fibers from a thermoset matrix, the matrix had to be decomposed completely without any damage of the fibers. Solvolysis (chemical disintegration) and pyrolysis are possible processes for this purpose.

To carry out a solvolysis, the composition of the matrix should be known. Only on this basis can a functional and efficient process, with recovery and reuse of matrix and solvent, be developed. Unfortunately, the matrix material is normally unknown and not disclosed by the production companies because of confidentiality reasons. This presumably limits solvolytic approaches to in-house solutions for production residues.

Pyrolysis processes, on the other hand, can be operated totally independent from the knowledge of the matrix material. The arising pyrolysis gases can be incinerated and the energy be used for heating the process.

A pyrolysis-specific disadvantage is the formation of a solid carbon residue from the polymer decomposition. This char attaches to the fibers in brittle deposits (see Fig. 18, left picture). Fibers with deposits cannot be sized and reintegrated into a polymer matrix, and therefore not be reused at all. In fact, the char can be removed by an additional thermal treatment in the presence of oxygen, as depicted in the middle (5 min. of oxidation) and right picture (20 min. of oxidation) of Fig. 18. This, however, may damage the fibers and can reduce their tensile strength [51, 52].

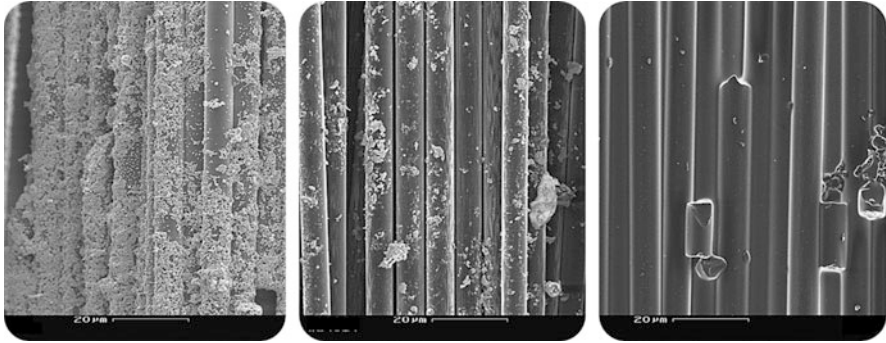


Fig. 18 SEM micrograph pictures of carbon fiber-reinforced plastics (CFRP) samples (dry fiber), treated at 670°C for 20 min in inert atmosphere (*left*). *Middle picture*: after pyrolysis an additional oxidation step (ambient air) was applied for 5 min. *Right*: oxidation step was extended to 20 min

If the fibers are exposed to an oxygen-containing atmosphere for longer, they may be totally destroyed and form small fragments of a needle shaped structure (Fig. 19). The resulting fiber dimensions and shapes can reach WHO criteria for being potentially harmful to humans.

4 Summary and Conclusion

The subject of this text is the recovery of materials in the course of the thermal treatment of waste. Even if thermal treatment is supposed to destroy materials, it offers good opportunities to recover thermostable substances, like metals and minerals.

Residues from MSWI include bottom ash and the products and deposits from flue gas cleaning.

The recovery of metals from bottom ash has a long tradition and reached a very high technical level within the last few years. Today even nonferrous metals can be recovered with high efficiency. Nevertheless, research and development is still going on in the field to optimize the metal yield and quality. Contrary to the metal components in the bottom ash, it is difficult to find reasonable recycling possibilities for the mineral fraction in most regions because natural building materials are often available, they are inexpensive, and do not have the “smell of waste.”

Also from the flue gas of the incinerators, a recovery of materials is possible. Traditionally HCl and gypsum can be recovered from the flue gas scrubbers. Unfortunately, the acceptance of these products is not very high because of the origin of the materials in the waste business. In Switzerland, new approaches to recover metals, predominantly zinc and lead from the filter dust of the plants, have been developed and are implemented on an industrial scale.

Besides waste incineration, the thermochemical conversion by pyrolysis also offers interesting technical opportunities for waste treatment and material recovery.

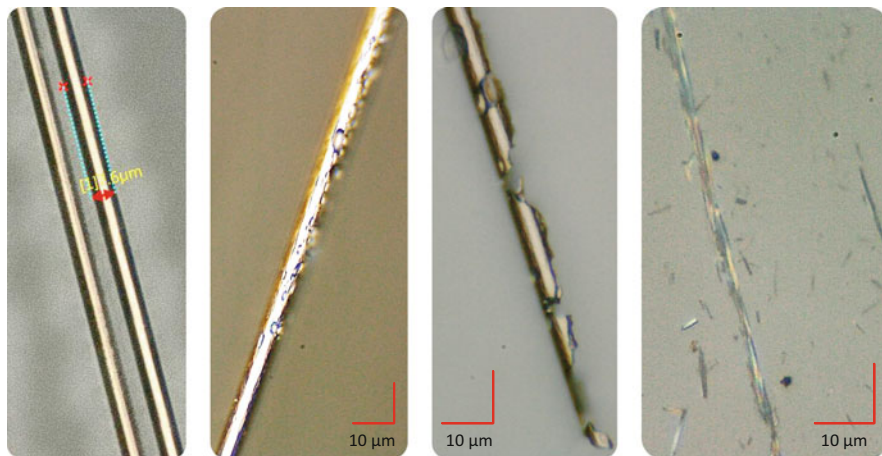


Fig. 19 Optical microscope pictures of carbon fibers, thermally treated at different temperatures and residence times (additionally to the time for heating up the sample) under ambient air atmosphere. From *left to right*: 700°C, 10 min – 800°C, 10 min – 900°C, 10 min – 900°C, ca. 40 min

Modern products and pre-concentrates from waste treatment facilities are often composed of different substances, components, and materials, e.g., WEEE, fiber-reinforced plastics, shredder fractions, etc., which cannot be separated by mechanical methods only. On the other hand, the incineration of the combustible part of the materials may damage some of the valuable components (e.g., carbon fibers and metals) in the composite materials. For these fractions, a pyrolytic treatment at relatively low temperatures and in the absence of oxygen may be the right processing step. Volatile plastic components can be easily removed, and the valuables in the materials are not negatively affected and can be recovered with high quality.

It is especially emphasized that the recovery of fuels or high-grade chemicals from MSW by thermochemical processes is not a reasonable pathway for the treatment of these fractions. Despite great efforts in the past, no economically feasible process for this purpose could show its practicability on an industrial scale and in a longtime operation.

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