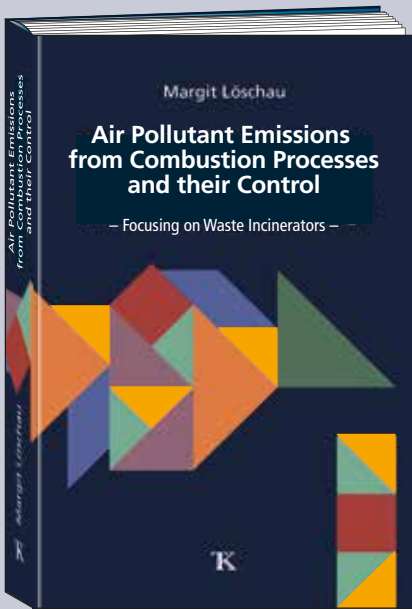


# Air Pollutant Emissions from Combustion Processes and their Control



## Air Pollutant Emissions from Combustion Processes and their Control

– Focusing on Waste Incinerators –

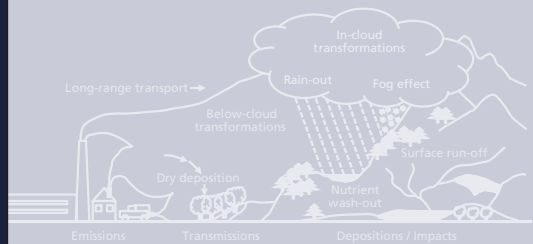
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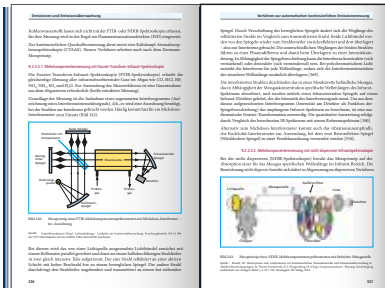
This comprehensive text and practical handbook thoroughly presents the control of air pollutant emissions from combustion processes focusing on waste incinerators. Special characteristics are emphasised and the differences to emission control from combustion processes with other fuels are explained.

The author illustrates the origin and effects of air pollutants from incineration processes, the mechanics of their appearance in the incineration process, primary and secondary measures for their reduction, processes of measuring the emissions as well as the methods of disposing the residues. In particular, the pros and cons of procedural steps and their appropriate combination under various conditions are emphasised.

Moreover, the book contains information and analyses of the emissions situation, the consumption of operating materials and of backlog quantities as well as of the cost structure of waste incinerators with regard to their applied control system. Furthermore, the author explicates the contemporary legal, scientific and technological developments and their influence on air pollutant emission control. An evaluation of the status quo of air pollutant control at waste incinerators in Germany, practical examples about possible combinations and typical performance data complete the content.

Accordingly, this book is a guideline for planing a reasonable overall concept of an air pollutant control that takes the location and the segregation tasks into consideration. This book is addressed to students, decision makers, planners and the operating practitioners if for example the construction of a new system or the implementation of improvement measures have to be conducted.

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# Overview of the Pyrolysis and Gasification Processes for Thermal Disposal of Waste

Jürgen Vehlow

1.	Background.....	431
2.	Brief technology description .....	432
3.	Pyrolysis .....	433
3.1.	Stand-alone processes.....	433
3.2.	Pyrolysis in combination with combustion/ash melting .....	434
3.3.	Pyrolysis upstream of a power plant.....	436
4.	Gasification .....	437
4.1.	Shaft furnace processes.....	437
4.2.	Fluidised bed gasification.....	438
4.3.	Gasification as upstream installation.....	441
5.	Conclusions and outlook .....	442
6.	References .....	443

## 1. Background

Thermal treatment of waste started in the 1870s in England with the first waste incineration plants and this technology was in short time adopted by many industrialised countries. Starting in the late 1970s waste incineration was blamed for emission of toxic compounds, in particular of dioxins, and public pressure initiated the decree of more and more stringent air emission standards in all countries which, again, induced significant improvement of the environmental performance of waste incineration.

Anticipated lower pollution was one reason to look for other thermal treatment technologies, another one was the option to convert waste into useful products other than energy. That is why pyrolysis and gasification came into consideration. Both processes avoid or at least minimise on the one hand the formation of dioxins and, on the other hand, end up with products which can be used as fuel in high efficient combustion processes or in the transportation sector. Another appealing option is their use as raw materials in chemical industry.

The application of both technologies has a long history, e.g. in charcoal pits or during and after world war II for the production of car fuel from wood. First serious attempts to use one or the other of these so-called alternative processes for treating municipal solid waste (MSW) started in the 1970s. The term alternative suggested that these processes would prove as superior in view of technical performance, environmental quality, and economy. In the mid-1990s some experts predicted their break through and expected 41 such plants in Europe with two thirds for MSW and the remainders for residues from recycling and for bio-wastes [22]. However, there were also sceptics who expected problems in view of complexity, energy efficiency, and costs and asked for careful monitoring of the further development and performance of these processes [14].

This report will concentrate on selected processes for treating MSW which found technical application and will discuss advantages and problems emerged after long-term operation. Air emission quality will not be referred to in detail since all technical plants have to comply with legislative standards of similar stringency all over the world. New two-stage combustion processes which are mainly for acceptance and/or tax reasons marketed under the label gasification, e.g. Energos [2] or Cleergas [4], will not be included. The final conclusions do not claim catholicity but reflect the personal view of the author.

## 2. Brief technology description

Pyrolysis and gasification are integral sub processes of the incineration of waste in grate systems which starts with drying, continues at higher temperature with pyrolysis and gasification and ends finally up with combustion of the waste. The technologies described here are also in combined processes strictly separated from each other.

Pyrolysis is a process for disintegration of organic substances at elevated temperatures under inert atmosphere. The technically preferred reactor is a heated rotary drum. The products are, depending on the operation temperature and the feed composition, gaseous organic compounds, oils, water, and coke rich ashes.

The term gasification denotes the partial combustion of an organic substance at high temperatures. The reactor is usually a fluidised bed or a shaft furnace. Primary products are hydrogen (H<sub>2</sub>), carbon monoxide (CO) and carbon dioxide (CO<sub>2</sub>), the solid residues are in almost all cases molten slags. Table 1 compiles the main reaction conditions and the products of pyrolysis and gasification.

Table 1: Reaction conditions and products of pyrolysis and gasification processes

	unit	pyrolysis	gasification
reactor		rotary drum	shaft furnace/fluidised bed
temperature	°C	250 to 700	800 to 1,600
pressure	mbar	≤ 1,000	1,000 to 45,000
reactant		none	O <sub>2</sub> , H <sub>2</sub> O, air
stoichiometry	–	0	< 1
main products		C <sub>n</sub> H <sub>m</sub> , H <sub>2</sub> , CO, pyrolysis oil, H <sub>2</sub> O	H <sub>2</sub> , CO, CH <sub>4</sub> , CO <sub>2</sub>
solid residues		ash, coke	ash/slag

The gaseous products of both processes can be used as feedstock in chemical industry e.g. for production of transportation fuels. These so-called stand-alone processes will not be discussed in this paper in case of gasification. In most technical implementations all products are, after some conditioning, either burnt in directly coupled combustion chambers or transferred into power plants respectively industrial furnaces.

### 3. Pyrolysis

#### 3.1. Stand-alone processes

The development of pyrolysis plants for MSW started in the 1970s in Germany and in the USA. First attempts in the USA failed for various reasons [2]. In the early 1970s Kiener built a rotary drum pyrolysis test facility in Germany. The waste was pyrolysed at 450 °C and the pyrolysis gas was fed into a gas engine for energy generation. This technology was later acquired by Siemens and was advanced to the Thermal Waste Recycling Process (Schwel-Brenn-Verfahren) [1].

At about the same time the Destrugas Process was developed to pyrolyse e.g. MSW, light industrial waste, and spent tyres in an externally heated shaft reactor at temperatures up to 1,050 °C [16]. The pyrolysis gas was partly used for heating the reactor, the solid residues were ought to be separated for metal and coke recovery. Two plants were built in Denmark and one test facility in Japan, but all were shut down and the process is not marketed any longer.

A much more successful story is that of the Burgau Pyrolysis Plant which was built in the small Bavarian town of Burgau. The main driver for the decision to go for pyrolysis was in addition to political preferences of alternative technologies the necessity to treat chromium sludge for what the reducing pyrolysis atmosphere is very much suited. The plant has been taken into operation in 1983 with an annual throughput of approximately 30,000 Mg [3].

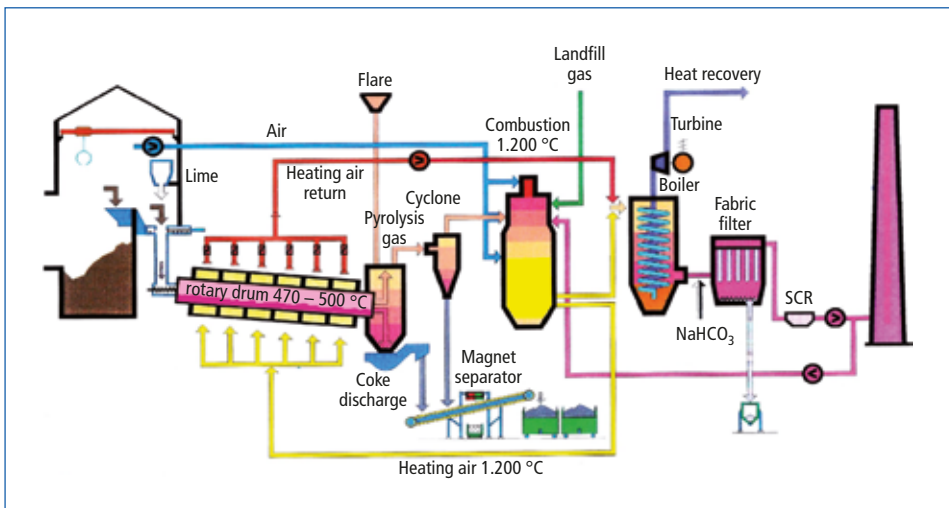


Figure 1: Scheme of the Burgau pyrolysis plant

Figure 1 shows the scheme of the plant. Shredded waste was fed by metering screw conveyors into two externally heated rotary drums with a throughput of 2 Mg/h each where it was pyrolysed at 470 to 500 °C. The combined pyrolysis gas passed a cyclone and was burnt at 1,200 °C in a combustion chamber which was followed by a boiler, a dry scrubbing system using NaHCO<sub>3</sub> as neutralising agent, and a SCR denitrification unit. The high carbonaceous pyrolysis residue was after ferrous scrap removal together with the cyclone ash deposited on a special landfill site.

In particular, during the first years of operation several modifications had to be made but some technology related weaknesses stayed during the entire lifetime of the plant. The sealing of the rotary drum required high maintenance efforts, the deposition of the pyrolysis coke needed special approval, and the operational costs were high, even if the special landfilling was not taken into consideration. These facts and the limited landfill capacity reasoned the decision to close the plant at the end of 2015. No other plant of that type has ever been built.

### 3.2. Pyrolysis in combination with combustion/ash melting

To solve the problem of treatment and disposal of the pyrolysis coke, Siemens KWU developed the Thermal Waste Recycling Process which was in principal an enhancement of the Kiener rotary drum pyrolysis by additional treatment of the pyrolysis residues [1]. A simplified scheme of the process is shown in Figure 2.

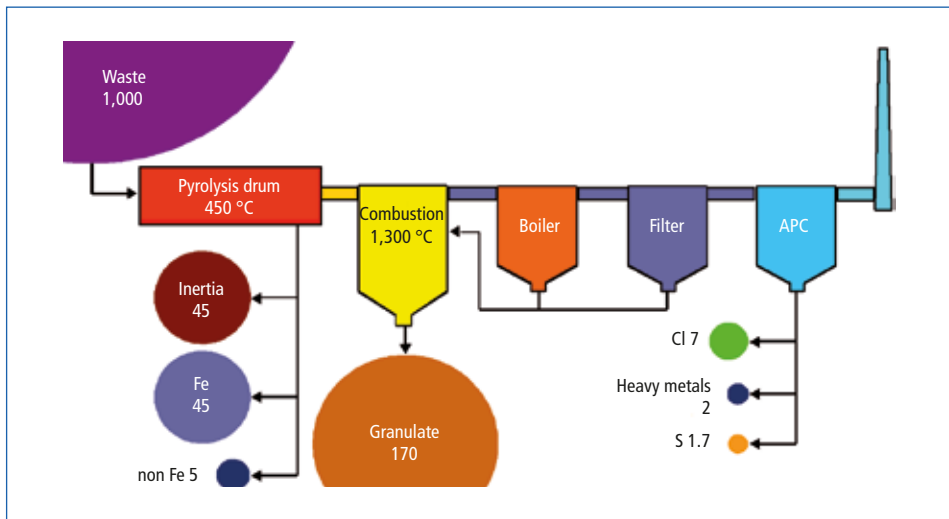


Figure 2: Simplified scheme and mass balance (in kg) of the thermal waste recycling process

The rotary drum accepted waste of a particle size < 150 mm, it contained internal tubes for heating the material to 450 °C. The pyrolysis lasted 1 to 2 h. The pyrolysis gas was directly transferred into the combustion chamber where it was burnt at 1,300 °C. The flue gas entered a boiler, followed by a filter and a conventional air pollution control (APC) system. The dry residue was sieved, the >5 mm fraction underwent a separation

of ferrous and non-ferrous metals, the remaining mineral fraction could be utilised in the building sector. The <5 mm fraction was ground to <1 mm and blown together with the boiler and filter ashes into the combustion chamber. The high combustion temperature cared for the total melting of all mineral matter, the liquid slag was quenched in a water tank, the resulting granulate could also be used in the building sector. The process scheme in Figure 2 contains a global mass balance based on typical German MSW composition.

The process was tested in a single-line 4 Mg/d test facility between 1988 and 1996. The feed comprised not only MSW but also other residues including auto shredder residue [15]. In 1994 a double line 100,000 Mg per year pilot plant was erected in Fürth, Bavaria, and taken into operation in 1997. The estimated operational costs were higher than those of grate incineration plants although the recovery of metals from MSW was much more efficient and the boiler efficiency was expected to reach 23 percent.

Various technical problems showed already up during commissioning, in particular plugging of waste in the drum, failures of the drum sealing, and clogging in the gas transfer tube. After months of improvement a sealing broke again during operation for reaching technical approval. Gas leaked, some people were hurt, and the plant was finally closed. Two other orders in Germany were never realised and Siemens stopped marketing the process.

However, the company had given licenses to two Japanese companies already in 1991. The Mitsui MES R21 process has an almost identical design as the Thermal Waste Recycling Process. Mitsui built 7 plants with 15 lines and a total capacity of 1,840 Mg/d between 1997 and 2009. The Toyohashi plant is with two lines and 200 Mg/d the biggest one. A three-dimensional scheme of the design of that plant is shown in Figure 3.

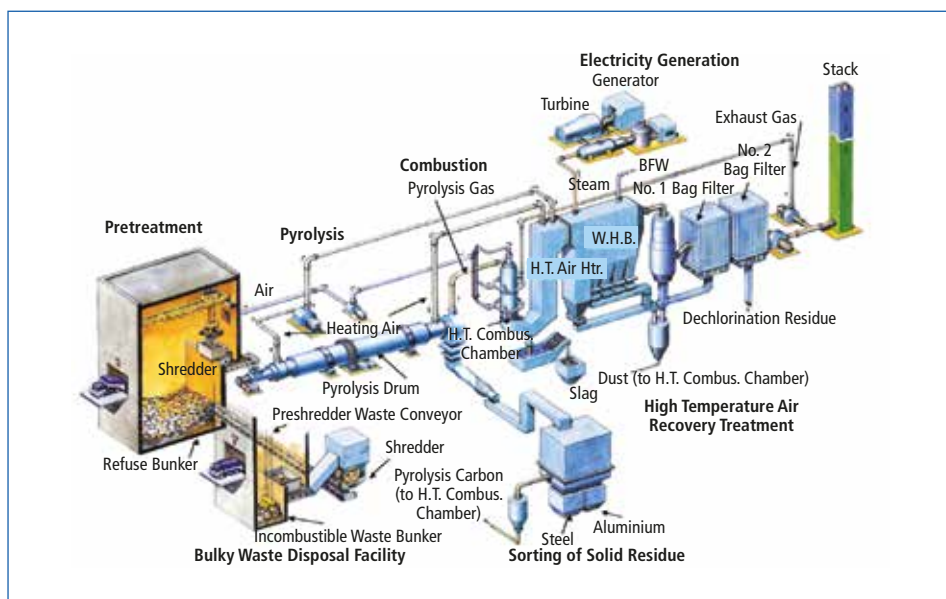


Figure 3: Three-dimensional scheme of the Toyohashi MES R21 plant

Several visits in two of such plants left the impression of more or less permanently regular operation. One reason could be the low ash content of Japanese MSW which is typically 10 to 12 wt.-% only.

A second license of the process was given to Takuma in 1995. The company built 5 plants until 2012 with 10 lines and a total capacity of 785 Mg/d which are still running without major problems.

Although it is difficult to get reliable cost data for Japanese plants it can be expected that the investment and operational cost are higher than those of other technologies since both companies stopped marketing the process meanwhile. A further reason might be the low energy efficiency of the process. Data obtained from two MES R21 plants showed 15 percent of power efficiency without indicating whether this was the gross or the exported power.

### 3.3. Pyrolysis upstream of a power plant

Between 2000 and 2001 a rotary drum pyrolysis, ConTherm, was built in Hamm, Westphalia, to pyrolyse high-calorific refuse derived fuel (RDF), mainly plastics, and burn the pyrolysis gas and the coke in a power plant [18]. A simplified scheme of the plant is shown in Figure 4.

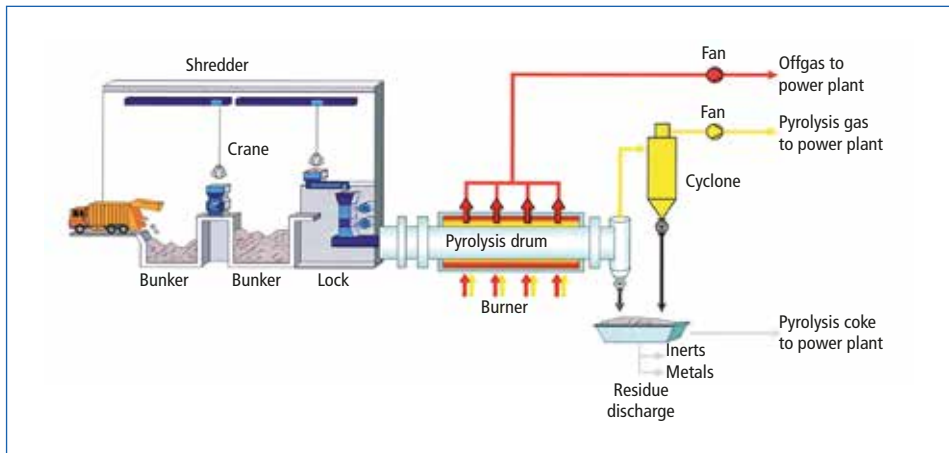


Figure 4: Scheme of the ConTherm plant

The RDF was pyrolysed in an externally heated rotary drum at 500 °C. The extracted pyrolysis gas passed a cyclone and was blown by a fan into the power plant. The solid residue was separated into an inert fraction which was landfilled, a metal fraction which was sold, and the coke fraction which was also blown into the power plant. The power plant was equipped with an air pollution control system.

The name plate capacity of the two parallel lines plant was 120,000 Mg per year but had to be reduced already during the service trials to 70,000 Mg per year since the rotary drums were underdesigned: the waste in the drum centre did not reach the scheduled

temperature. The sealings of the drum caused always problems with pyrolysis gas leakage or air penetration and the control of the underpressure in the pyrolysis part pointed out to be difficult.

The gas transfer line tended to clogging by dust and tar and had to be shortened and externally heated. The emergency stack became too hot and lost stability. It finally bent over in late 2010 which was reason to shut the facility down. An additional reason might have been rising fuel prices.

Most of the problems had been solved with time and the design deficits could be avoided, but the technology is complex and requires a lot of maintenance. Nevertheless, respective market conditions might make the process attractive again.

## 4. Gasification

### 4.1. Shaft furnace processes

Gasification has a long tradition in Japan. Nippon Steel started the development of a gasification process for MSW based on its experiences with the shaft furnace technology. A first two lines fixed bed gasification plant with a capacity of 50 Mg/d each line was taken into operation in 1979. This was the prototype of the nowadays widely used Direct Melting System (DMS) [17]. A scheme of a shaft furnace with its reaction zones and of the entire gasification plant in the Munakata Eco Park, Kiushu, is shown in Figure 5 [8].

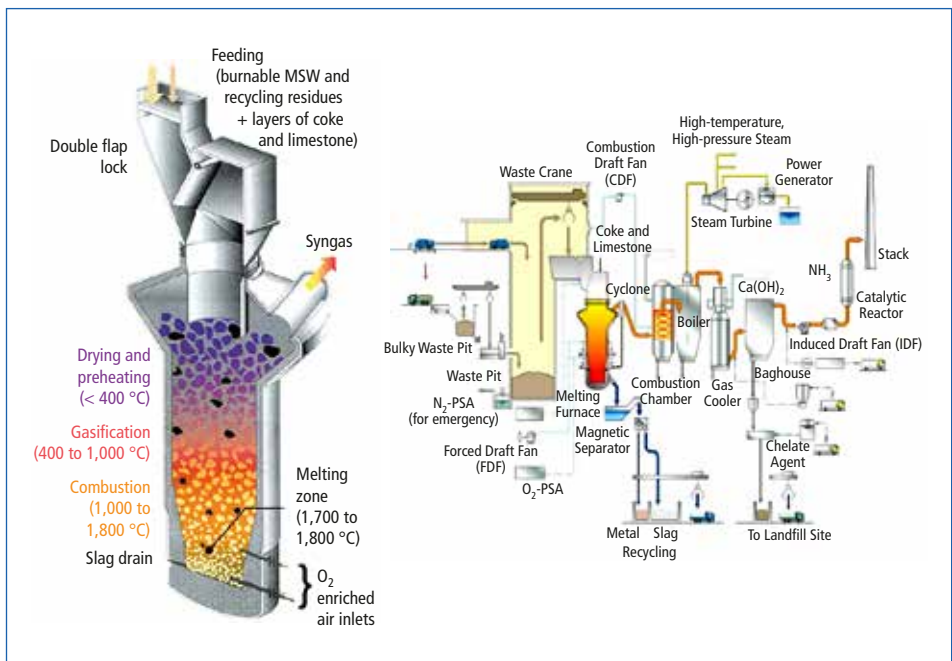


Figure 5: Reaction zones in a shaft furnace (left) and scheme of the Munakata Eco Park DMS plant (right)



The waste enters the furnace via a double flap lock which cares for tight separation of the furnace atmosphere from the environment. About 5 to 10 wt.-% of coke is added to the MSW, sometime also limestone is added to bind sulphur. Oxygen enriched air (36 vol.-%) is blown into the furnace through 4 nozzles in the lower part; through another 6 nozzles higher up air is injected. The energy for the gasification process is delivered by the combustion of residual carbon at 1,000 to 1,800 °C in the lower part of the furnace. This high temperature causes melting of the slag which is drained through an outlet at the bottom of the furnace.

The gasification takes place in a temperature range between 400 and 1,000 °C above the combustion zone. The residence time in the furnace is 4 h. The hot syngas leaves the furnace after heating the mix of waste, coke, and lime stone. Its main components are CO with 15 to 20 vol.-% and H<sub>2</sub> with 2 to 5 vol.-%. The calorific value of the gas is in the order of 6.7 MJ/m<sup>3</sup>. The extracted syngas passes a cyclone and is burnt at approximately 950 °C in a combustion chamber which is followed by a boiler and a conventional APC system.

For the Munakata plant the design throughput per line is 80 Mg/d of waste and the design operation time is 240 d/a per line. This operation time is typical for such type of plants in Japan and indicates significant efforts in terms of maintenance.

The DMS process is the mostly applied gasification technology in Japan with already 28 plants with 57 lines and a total capacity of 6,200 Mg/d.

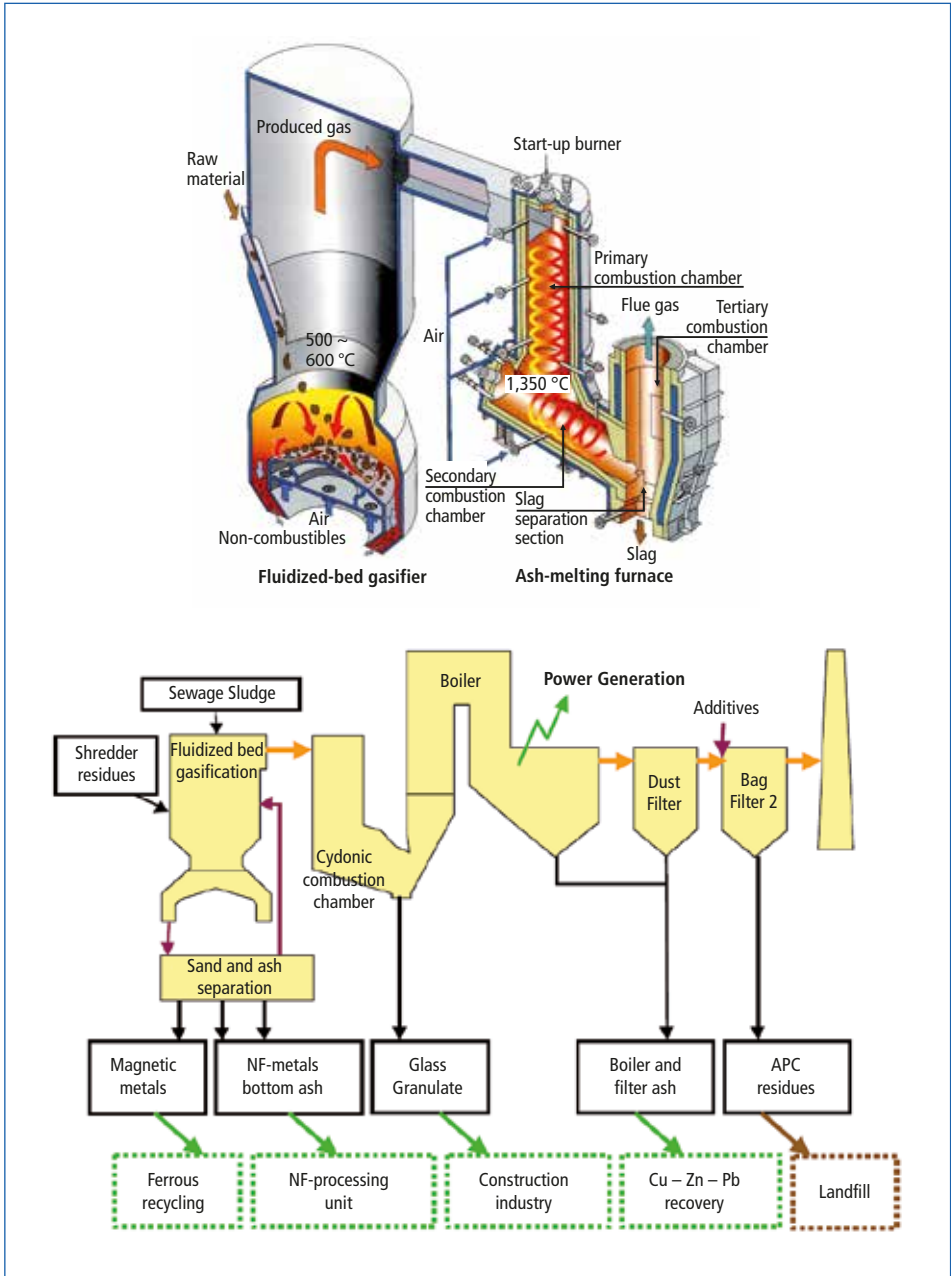
A similar process is the JFE High-Temperature Gasifying and Direct Melting Furnace System with 10 plants erected and operated since 2003 [12]. The design of the shaft furnace differs from that of the DMS one since it is a combination of a fixed bed and a fluidised bed furnace. It is fueled from above with a mix of RDF, coke, and limestone. Oxygen enriched air (35 vol.-%) is injected at the bottom together with a certain amount of water to enhance the water gas shift reaction and to modify the gas composition. Additional air supply higher up causes fluidisation of the material in the upper part of the furnace. The dust laden syngas is directly transferred into a combustion chamber followed by a boiler and an APC system.

## 4.2. Fluidised bed gasification

The Japanese company Ebara had a long tradition in building fluidised bed incinerators based on the German Hölter rotating bed design. In the 1990s they used this principle to develop a gasifier and took a first 7 Mg/d test facility into operation in 1995. The first full- scale two lines plant of 225 Mg/d each line started operation in 2000 [21]. The core of this TwinRec Process (TIFG) called process is a combination of a rotating fluidised bed gasifier, two cyclonic, and one conventional combustion chambers as can be seen in the left graph in Figure 6.

The process accepts shredded MSW, but also other residue streams such as light industrial waste, auto shredder residues, or plastics. The gasifying temperature is 500 to 600 °C, the combustion temperature reaches 1,350 to 1,450 °C with the result that all

mineral matter is molten. The gasifier-combustor part is followed by a boiler and an APC system. A simplified flow diagram of the process is shown in the lower graph in Figure 6 which also indicates the management of the solid residues.



Pyrolysis/Gasification

Figure 6: Gasifying and combustion part and simplified flow sheet of the TwinRec process

Like the shaft furnace gasification plants, this one, too, has a limited operation time of 240 to 280 d/a and indicates similar maintenance efforts. Until 2009 the company erected 12 plants with 21 lines and a total capacity of 3,100 Mg/d. The operation of the plants is said to cause no major problems.

Fluidised bed gasification plants have also been developed and built by the Japanese companies Kobelco [9] and Hitachi Zosen [6]. Both companies use bubbling fluidised beds for gasification, followed by two combustion chambers, boiler and APC system. Kobelco built until 2013 14 plants in Japan and 2 in Korea with a total capacity of 2,600 Mg/d. Hitachi Zosen had 9 plants in operation in Japan and one in construction in Korea.

A different type of gasification process, developed by Thermostelect, started its test phase in a small facility in Fondotoce, Italy, in 1992. The basic idea was to realise a process which converts MSW into marketable products with almost no residues which would require final disposal [19]. A simplified scheme of the process with a mass balance is shown in Figure 7.

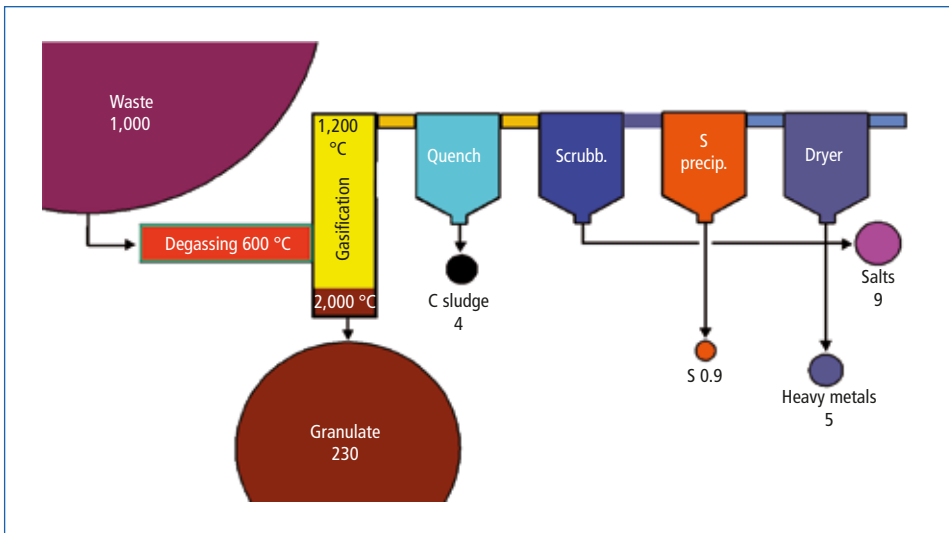


Figure 7: Simplified scheme of the Thermostelect process with estimated mass balance

A Thermostelect plant comprises a long channel with a standard load of 10 Mg/h where the waste is compacted and heated up to 600 °C. The partly degassed material is continuously pushed into a vertical reaction chamber where it is gasified by controlled injection of oxygen. The adjusting temperatures reaches from 2,000 °C at the bottom where a molten bath of minerals and metals is formed to 1,200 °C at the top. The syngas enters a quench where it is shock-cooled to 70 °C. This process causes the formation of high amounts of carbonaceous sludge the control of which is one of the critical issues of the process [5]. The further syngas cleaning is complex with recovery of sulphur, metal compounds, and salts since the original concept did not consider combustion but utilisation of the gas, e.g. in fuel cells.

The first full-scale plant with 3 lines and a total capacity of 225,000 Mg per year was built in Karlsruhe and started trial operation in 1999. Here the cleaned syngas is directly used for power generation in a combustion chamber. From the early beginning problems emerged which required some serious design changes: the heating of the degassing channel was eliminated, a combustion chamber with complete flue gas cleaning was built instead of the original emergency flare, the plant had problems to reach the name plate throughput.

In 2002 the German authorities approved the safety equipment of the plant and commercial operation started. However, the plant did never reach its planned operation mode and owner shut it down in 2004 already, claiming unbearable losses. The failure of this prototype plant stopped all other activities in Europe.

Kawasaki Steel, today JFE, got a license of the process for Japan and built since 1999 seven plants. The first one started operation in Chiba City, north of Tokyo, on the premises of a steel mill in 1999 and exported its syngas together with the blast furnace gas of the steel mill to a gas fired power plant via a huge gas pipe line. According to the operator this plant run without the problems seen in Karlsruhe. One reason might be the low ash content of Japanese MSW of 10 to 12 wt.-% only, since the operator disclosed that he could not reach full throughput during tests with auto shredder residue which had an ash content of almost 20 wt.-%.

### 4.3. Gasification as upstream installation

In 1998 a 60 MW circulating fluidised bed (CFB) gasifier, Kymijärvi I, was built in Lahti, Finland, which injected syngas derived from SRF and forest residues without cleaning into a coal fired power plant. This plant was successfully operated for a couple of years but had to be shut down due to new emission regulations [13]. A new two lines CFB gasification plant (2 x 80 MW), Kymijärvi II, was built instead which also uses SRF as fuel [7]. The gasification temperature is 850 to 900 °C, the syngas is first cooled down to 400 °C to condense corrosive metal chlorides which are precipitated in a battery of 6 series connected fabric filters. The gas is burnt in a gas fired power plant which is equipped with an APC system. A scheme of the plant is shown in Figure 8.

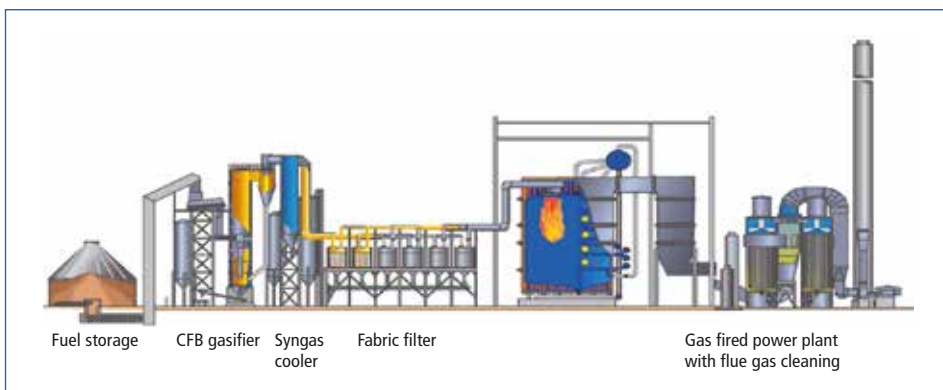


Figure 8: Simplified scheme of the Kymijärvi II plant

Test trials of the plant started in 2012. After obviously a number of modifications during the first years and 25,000 h of operation the plant has achieved its set targets in 2016 [10]. The plant supplies the base load of the Lahti district heating grid and has a gross power efficiency > 30 percent. Whether this design has a chance on the European market will greatly depend on its long-term performance and the costs. Reliable data on the latter ones are not yet available.

## 5. Conclusions and outlook

Regarding the above described pyrolysis and gasification processes in view of their applicability for treating MSW it has to be stated, that all of them were not successful in Europe. The two large scale processes Thermal Waste Recycling and Thermoselect failed partly because of technical problems, partly because of costs. Upstream processes may have a chance on future markets depending on their costs and their energy efficiency. Typically, such processes treat SRF the production of which, however, is almost never taken into account in published costs and energy balances.

Considering other types of residues, such as packaging waste, auto shredder residues, or waste from electrical and electronic equipment, these processes can have advantages compared to grate fired waste incineration plants. They can more easily treat high calorific fuels and the recovery of metals can be more efficient, especially in pyrolysis processes. Hence waste type dedicated pyrolysis or gasification plants may emerge in future on the European market, in particular if they find direct application of their products.

One question remains: why were these processes successful in Japan? The share of these technologies in terms of total capacity was 10 percent in 2012 [11]. Some justifications are closely related and caused by national strategies. In Japan MSW has to be disposed of in the near neighbourhood of its generation. This results in a number of small facilities and the specific investment of plants reduces the cost difference between grate systems and gasification or pyrolysis plants. Especially the gasifiers have typically small capacities.

Another driver is the Japanese preference for 'novel' technologies the market launch of which is typically supported by the authorities. The Japanese waste management system is mainly financed by the public budget which includes the investment for new facilities as well as the major part of the operation costs. Especially the investment costs are commonly higher than in Europe although reliable data are hard to find.

Until recently the energy efficiency of the waste treatment plants was widely disregarded. One reason was the common vitrification of the solid residues from waste incineration, not only to reduce their volume but also to destroy dioxins in accordance with the total output limit of 5 ng(I-TE)/Mg of waste. Currently a change is seen and large energy efficient grate incinerators are built in big cities again.

Whereas during the early 2000s mainly gasification or pyrolysis plants were erected in Japan, the share of grate incinerators is increasing since 2008 again as can be seen in

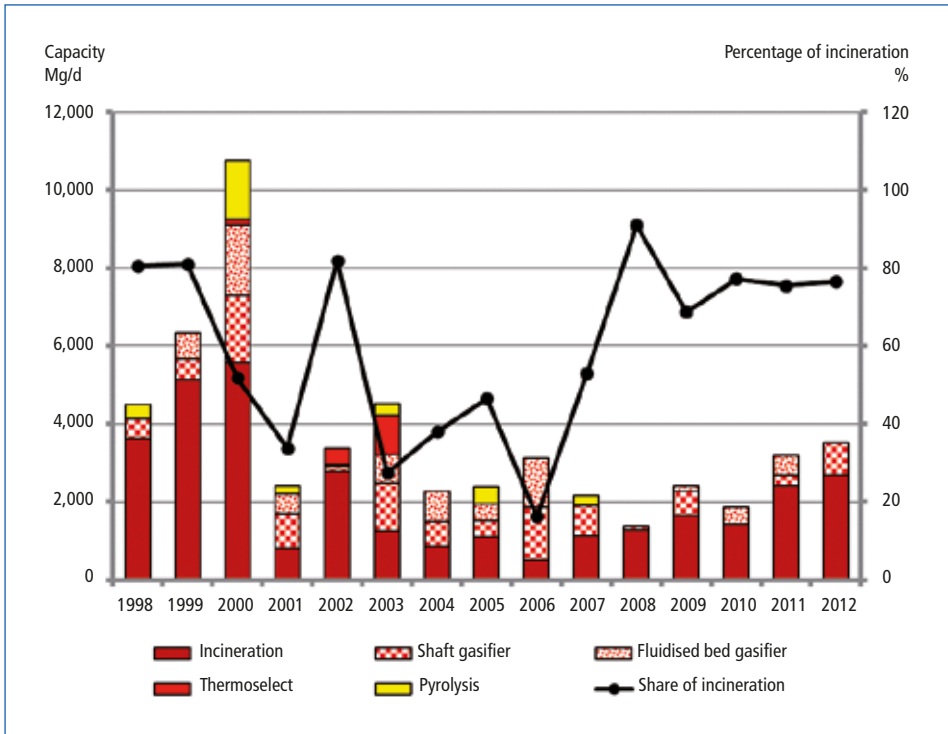


Figure 9: Total capacity and share of annual commissioning of thermal waste treatment plants in Japan

Figure 9 which shows the annual startup of thermal waste treatment plants in Japan between 1998 and 2012 [23]. The Japanese government expects a share of 10 to 20 percent in novel technologies for new plants [11] and that means that Japan will most likely be the main market for these technologies in future.

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