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# IMPROVEMENT OF BOTTOM ASH QUALITY FROM INCINERATION OF MUNICIPAL SOLID WASTE (MSW) BY PROCESS INTEGRATED MEASURES AT GRATE SYSTEMS

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## ABSTRACT

The improvement of ash quality from incineration of municipal solid waste (MSW) is a very important R&D task for sustainable waste management. Reusable products would be: minerals as secondary raw material for building applications and heavy metals as ore concentrates. Such improvements will be accepted only if they can be realised with moderate additional costs. Grate systems offer the technical potential for the intended improvement. In the present work the approach used for improvement of bottom ash is the separation of the heavy metals from the mineral matrix by evaporation. Corrosion aspects must be considered separately.

Experiments have been carried out on a 0.5 MW pilot grate system to investigate the heavy metal evaporation process. Zinc as volatile and copper as a more or less non-volatile metal were investigated. The operation conditions of the incineration where changed between combustion and gasification operation.

Two independent methods were used to determine the metal partitioning and to demonstrate the improvement of bottom ash quality. One was based on a mass balance of the heavy metals, the other on a radio tracer method which was applied for the first time for monitoring heavy metal evaporation in a grate system. The radio tracer method offers additionally the opportunity to identify directly the evaporation process on the grate. Zinc and copper isotopes were injected as tracers at the beginning of the grate. Their gamma radiation was measured with detectors positioned alongside the grate and the flue gas system. With this installation the transport of the radio tracers through the incinerator could be monitored, especially it allows to localise the place where the evaporation occurs.

The results show the expected trends. Copper is negligibly evaporated whereas the radio tracer zinc is evaporated during a very short period at high temperatures and reducing conditions. By changing the operation conditions from combustion to gasification the amount of evaporated zinc can be increased i.e. by increasing the homogeneous zone with reducing conditions on the grate. The operation conditions have an effect on the evaporation place of zinc which is postpone under gasification conditions.

#### KEYWORDS

MSW, bottom ash, incineration, heavy metals, evaporation, metal partitioning, tracer

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#### INTRODUCTION

Grate systems have been used for decades for the thermal treatment of municipal solid waste (MSW) and similar wastes from industry. With regard to the reduction of pollutant concentrations the emphasis lay on the development of an efficient purification of flue gas and the post-treatment of residuals (i.e. secondary measures). Plants with the state-of-the-art pollution control equipment generally comply with the emission limits for air, water and soil.

The developments of the past few years in the area of the conventional processing with grate technology indicate a considerable potential for optimisation by so-called primary measures with regard to minimisation of pollutants, improvement of efficiency etc.. The optimisation of the combustion chamber design, the flue gas recirculation and mixing of flue gas, oxygen enrichment of the primary air, water-cooled grate elements and further development of control systems (e.g. IR-camera) can be mentioned in this context.

The improvement of the ash quality is a very important research and development task for sustainable waste management. Per tonne of MSW 250 kg of mineral residues are left. Depending on the operating conditions, heavy metals (HM), chlorides (Cl) or sulphates (SO<sub>4</sub>) are released into the flue gas (gaseous, aerosols) or concentrated in the incineration residues (bottom ash). Operating conditions influencing the ash quality with respect to HM, Cl, SO<sub>4</sub>, etc. have to be considered in conjunction with corrosion problems.

To meet the goals of sustainable resource management a significant improvement of the quality of residues is required. Reusable products would be: minerals as secondary raw material for building applications and heavy metals as ore concentrates. It is clear that such improvements will be accepted only if they can be realised with moderate additional costs.

The experience of the last 10 years has shown that for the improvement of bottom ash a thermal process step is essential. This step can be combined with additional process steps such as mechanical processes or washing processes. For the improvement different approaches are practised today: separation of the HM from the mineral matrix by evaporation, vitrification of HM in the mineral matrix, and leaching or mechanical separation of the remaining HM after the incineration process.

Grate systems offer the technical potential for the intended improvement. It has been shown previously that an improvement of bottom ash quality with process integrated measures is possible. This would also have a positive effect on the costs. On the other hand there is still a lack of physico-chemical information about the evaporation/immobilisation process which occurs on the grate.

In the presented work the approach used for improvement of bottom ash is the separation of the HM from the mineral matrix by evaporation. There exist hypotheses, under which conditions HM such as Zinc, Lead, Cadmium etc. can significantly by evaporated. The factors, which influence the heavy metal evaporation are temperature, partial pressure of oxygen and the chlorine in the gas, e.g. [1-8]. Macro- and microscopic mixing and the mean residence time can have an influence on the heavy metal evaporation.

Material flux analysis is a method for determining, describing and analysing the metabolism of e.g. furnaces. It analyses the flow of different materials through a defined space, within a certain time. This method is explained for example in [6]. By definition no conclusion is possible, within the balance area, about the location of processes such as heavy metal evaporation. In practice, the entire furnace must be balanced, although only a certain area

within the furnace is of interest. The allocation of evaporation phenomena to one specific area can therefore be difficult or impossible. The incineration process on the grate is complex, as different interacting reactions take place in parallel and in series. Therefore detailed information from inside the process is needed. For this, the material flux analysis has to be complemented by other methods.

With the proposed new radio tracer method, detailed information about evaporation and immobilising phenomena should be detectable. During stationary operation of the incinerator, activated heavy metals are injected as tracers to the solid waste by pulses. The chemical properties of these tracers are identical to the inactive heavy metals. The mass flux of the tracers is negligible in comparison to the flow of inactive heavy metals. The gamma radiation of the tracers can be measured with detectors positioned alongside the grate and the flue gas system. With this installation the transport of the radio tracers through the incinerator could be monitored, especially it allows to localise the place where the evaporation occurs.

To reach the ambitious aim, three partners with complementing competence worked together. *Paul Scherrer Institut* (PSI) was responsible for the evaluation of promising operation conditions by laboratory tests and the sampling of radio tracers in the flue gas. *Clausthaler Umwelttechnik-Institut* (CUTEC) was responsible for the pilot plant operation and *Fraunhofer Institut für zerstörungsfreie Prüfverfahren* (IzfP) for the radio tracer measurements.

#### **OBJECTIVES**

The objectives of the experiments are the following:

- Verification of hypotheses for an adapted incineration operation to improve bottom ash quality
- Improvement of a physico-chemical database for the heavy metal evaporation (zinc, copper)
- Verification of the applicability of a new radio tracer method

Zinc and copper are today two of the important heavy metals which cause the environmental problems in the boutom ash. Both have quite different physico-chemical properties. Under the prevailing conditions zinc is a volatile and copper is a more or less non-volatile heavy metal.

For the incineration a synthetic model waste was produced using similarity theory. The main components were urban waste wood, plastics and lava as mineral component. With this approach the results of the pilot plant should be comparable with the one form municipal solid waste (MSW) incineration. As the composition was held constant during the whole measurement campaign the expected effect must be due to changes in operation and not due to changes in the input waste stream.

## METHODOLOGY

#### Description of the Pilot Plant

The pilot plant with a thermal power of 0,5 MW consists of the main components (figure 1)

forward-acting grate system,

- · post combustion chamber system and
- · flue gas purification.



Fig. 1. Pilot plant and detector positions

A feeding ram discontinuously ensures the charging of the first grate stage with waste material from the storage hopper. Then the motion of the grate elements provides the transport down the grate. The reaction gas (usually air) enters the chamber beneath the grate and is independently adjustable in the three grate zones with regard to mass flow and oxygen concentration. After the last grate stage the bottom ash is falling into a box and discontinuously discharged by a screw conveyor.

When operating the grate process understoichiometrically, a combustible gas is generated on the grate stage, which is directed to a combustion chamber system for the self-sustained, independent, multi-staged incineration. In the case of an overstoichiometrical operation of the grate, the flue gas merely passes through the hot combustion chamber without undergoing post combustion.

The flue gas purification unit starts with cooling down the hot flue gas by controlled input of quench air and a successive heat exchanger. The separation of the flue dust is achieved by means of a cloth filter and the adsorption of remaining gaseous trace pollutants (e.g. heavy metals, HCl, SO<sub>2</sub>) takes place in an activated coal filter which is installed before the flue gas reaches the chimney.

The process flow chart of figure 1 roughly conveys the quantities measured during operation (e.g. gas concentrations, temperatures and flow rates). Those measuring quantities are significant to the plant's operation and balancing. They are recorded and stored for further processing.

To carry out the radio tracer measurements, the pilot plant was extended by a movable tracer tube for injecting tracer materials on the combustible bed. Alongside the grate 11 detectors

are positioned to measure the gamma radiation (figure 1). These detectors are located immediately after the injection point of the tracers, at the middle and the end of each grate zone, approximate 100 mm above the grate. Further detectors are placed alongside the flue gas system: at the outlet of the post combustion chamber, at the cloth filter and at the activated coal filter. To determine the amount of evaporated radio tracer a portion of flue gas was suck off the post combustion chamber and washed in an absorber.

## **Radio Tracer Measurements**

The application of short-lived radioactive substances is well-known from nuclear medicine, e.g. from function test of the thyroid gland. In technology such radio tracers are often used for measuring the residence time distribution (RTD) in continuously operating processes, optimisation of mixing processes, identifying leaks, blockages and other malfunctions. Radio tracer methods have been applied for example to determine the mixing quality of flue gases in a post combustion chamber [9] or the transport behaviour of waste material in a rotary kiln [10]. The application of radio tracers for the localisation and quantification of heavy metal evaporation is new and has the following advantages:

- · continuous measurement without taking samples, even through non-transparent walls,
- · observation of one and the same sample during the whole process.

The tracer must fulfil following requirements:

- a heavy metal isotope with the same chemical properties as the inactive heavy metal
- · an emitter of gamma rays
- short radioactive half-life (some hours until a few days).

The isotopes <sup>64</sup>Cu and <sup>69m</sup>Zn meet these requirements. The isotope <sup>64</sup>Cu has a half life of 12.7 hours and emits gamma rays with an energy of 511 keV with a probability of 37 % due to positron annihilation. The isotope <sup>69m</sup>Zn has a half life of 13.8 hours and emits gamma rays with an energy of 439 keV. Both isotopes are possible to generate in a research reactor by neutron activation. To generate <sup>64</sup>Cu a pure sample of copper metal is necessary, to generate <sup>69m</sup>Zn a small quantity of highly enriched <sup>68</sup>Zn is required.

These isotopes were applied to determine the heavy metal evaporation. For each experimental run one irradiated ampoule with a sample of <sup>64</sup>Cu or <sup>69m</sup>Zn, respectively, was used. The ampoule was cracked, its content was mixed with 20 grams of waste material and pressed into a pellet. This single pellet was injected at the beginning of the grate through the tracer tube. This causes a so called DIRAC-pulse.

By means of 1"-NaI(Tl)-scintillation-detectors, shielded and collimated by special blocks made from tungsten alloy, it was possible to localise the labelled metal in the incinerator. The detectors were positioned at selected opposite points alongside the grate of the pilot incinerator. By using the same detectors but labelling the input mass flow with non volatile  $^{113m}In(OH)_3$  or  $^{113m}In_2O_3$  it was also possible to determine the solid residence time distributions in the whole incinerator as well as in several parts of it.

For the determination of the quantities of copper or zinc in the flue gas an absorber filled with diluted nitric acid was used. A partial flow of the flue gas was continuously sucked off the combustion chamber and passed through this absorber. An unshielded 1,5"-NaI(Tl)-scintillation-detector positioned in the centre of the absorber recorded the absorbed quantity of radioactive metal as a function of time. At the end of each experimental run the dissolved

content of an irradiated ampoule containing the small quantity of the corresponding metal was given directly into the absorber for calibration.

To determine the location of heavy metal evaporation on the grate two independent measurement are needed. One is the radio tracer measurements with zinc and copper from where we get the heavy metal evaporation in function of time. The other one is the radio tracer measurements with In which determines the waste material flow on the grate.

#### RESULTS AND DISCUSSION

#### Residence time distribution (RTD) of waste material on the grate

The RTD information of waste material on the grate is a prerequisite to be able to calculate from the time-dependent evaporation signal a space-dependent signal. Due to specification of the detectors only one tracer at the time can be applied. Therefore the RTD of waste material was measured separately. Nine radio tracer measurements were performed with different radio tracers (indium, zinc and copper). Alongside the grate the isotope radiation was measured at six different detector positions (see also figure 1).

Waste mass flow and grate velocity of the forward-acting grate were held constant for the whole measurement campaign. In figure 2 an example of measured residence time distribution with Indium is shown. The shape of the transients recorded by the individual detectors is predominantly due to an insufficient shielding of the detectors and not due to material transportation. This effect is apparent for the measurement at the beginning of the grate (position 0.0 meter). After injecting the tracer as DIRAC pulse a continuous signal decrease is measured during ten minutes. Each detector measures therefore isotopes within the zone of 0.3 meters up- and downstream of the detector position. The measured deformation of the detector signal at position 0.3 meter is due to an unexpected release of indium into the flue gas during a very short period of three minutes.



Fig. 2. Residence time distribution (RTD) measurement of waste material on the grate

The residence time is defined as the time at which each detector measures maximum impulses. As expected a linear correlation between detector position and residence time was determined. The RTD was detectable for all nine measurements independent of tracer

material and tracer concentration. In the example given in figure 2 the measured residence time for the whole grate is 45 minutes.

With the existing raw data a calculation of the axial mixing or respectively axial dispersion is difficult. Fortunately different indications point out that axial mixing is negligible. Therefore the system is comparable to a plug flow stream. Indications are the constant variance of the signal distribution alongside the grate, symmetric signals for each detector and residence times which correspond to the time with signal maximum.

Based on these indication the time-dependent evaporation signal measured in the flue gas can therefore easily be transformed into a space-dependent signal. Thus the place of zinc and copper evaporation can be localised clearly.

#### **Copper evaporation**

Copper was chosen as a representative for a more or less non-volatile heavy metal. Concentration in MSW ranges up to 2000 mg/kg. Typically, 90 to 95% of the input copper is concentrated in the bottom ash [6]. Aspects of toxicology and preservation of resources motivates a copper reduction in the bottom ash. About 80% of copper is metallic and the particle size distribution ranges from 0.1 millimetres up to several centimetres [11]. The remaining 20% is chemically bound copper. For the experiments metallic copper was used with particle sizes of 0.2 to 0.6 millimetres.

Two experimental measurement with copper tracer are shown in figure 3. Both measurements were executed under identical operation conditions (high temperature, oxidising conditions) and show a good reproducibility. The evaporation occurs alongside the whole grate and can not be allocated to a specific place. The determined 3 to 5 % of copper evaporated confirm the hypotheses that even under chemically favourable conditions no substantial evaporation of metallic copper occurs. Unfortunately it is not distinguishable what mechanism is responsible



Fig. 3. Normalised Cu amount detected in flue gas scrubber after the pulsed addition of Cu tracer at position 0.0 m. Accumulation is monitored while tracer is transported along the grate (0.0 to 2.0 m). oxi.: oxidising conditions; w: water content of waste; evaporation: % of tracer transferred to flue gas.

for the small copper evaporation. It could either be due to fine copper particles which were entrained with the flue gas, or reacted metallic copper which is evaporated as e.g. CuCl. As up to 95% remain in the bottom ash, it is not so important to know the mechanism for the determined release. For a significant reduction of copper in the bottom ash other separation technologies such as mechanical processes are needed, as they are presented, e.g. in [12].

Dispersed chemically bound copper species are not separable with mechanical separation technologies. For a estimation of evaporation potential of these species further investigations are needed.

#### Zinc evaporation

Zinc was chosen as a representative for a volatile heavy metal. The concentration in MSW ranges up to 2500 mg/kg. Typically, 50% of the input zinc is concentrated in the bottom ash [6]. For the experiments, metallic zinc granulates were used (2 millimetre in diameter).

For three different operation conditions the measured zinc evaporations are shown in figure 4. The two parameters which were varied on the grate were the oxygen partial pressure and temperature. The redox conditions of the incineration was changed from combustion to gasification operation by decreasing the flow rate of primary air to one third. But even in the so called combustion operation mode reducing conditions dominate in the solid bed in the first primary air zone. The variation of temperatures was achieved by adjusting the water content in the waste material. Grate temperature was increased approximately by 100 °C.

Figure 4 reveals that the trend of zinc evaporation is independent of the operating conditions and the amount of evaporated zinc. The evaporation takes place very fast i.e. in a narrow area on the grate. However the place of the evaporation depends on the operating conditions. The



Fig. 4. Normalised Zen amount detected in flue gas scrubber after the pulsed addition of Zen tracer at position 0. Accumulation is monitored while tracer is transported along the grate (0.0 to 2.0 m). oxi.: oxidising conditions; red.: reducing conditions (gasification); w: water content of waste; evaporation: % of tracer transferred to flue gas.

evaporation occurs always at location with high temperatures and with reducing conditions and can reach up to 100% evaporation. For the run yielding only 50% evaporation of the radio tracer (wet, oxidising) we expect the following: First, the location of evaporation is shifted to places further down the grate, due to delayed reaching of necessary temperature. Second, the shift in the temperature profile brings the evaporation zone for zinc into the region where oxidising conditions prevail (air zone 2). More and more zinc oxide is produced which is not volatile and therefore a further zinc evaporation is stopped.

## CONCLUSION AND OUTLOOK

The measurements on the pilot plant confirm the initially mentioned hypotheses that reducing conditions in connection with high temperatures improve the evaporation of zinc. Copper as non-volatile heavy metals is not evaporated significantly form the furnace bed as expected. A detoxification and reduction of the copper in the bottom ash must be obtained by other measures such as mechanical separation technologies. Corrosion aspects must be considered separately.

The new radio tracer method was successfully applied. Different improvements in the method are planed in the future, such as an improved shielding of the grate detectors, an increase in the number of tracer particles per experimental run, absorber configuration etc. For an application of the presented radio tracer method on an industrial incinerator no major modifications of the method are expected.

The gained set of data is useful for the validation of the physico-chemical modelling of zinc and copper evaporation. Future investigations with other metals, species of metals and operation conditions are foreseen.

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