

# Aging behaviour of bottom ashes from RDF-power plants

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## Abstract

In Germany waste materials are not only burnt in conventional waste incineration plants, but have also been increasingly used energetically in RDF power stations for some years. The quality properties and the aging behaviour of bottom ash from municipal waste incineration were examined thoroughly in the past. Based on a changed fuel spectrum compared to conventional waste combustion, changes in their aging behaviour are not to be excluded. For this reason investigations concerning quality and aging behaviour of bottom ashes from RDF power plants were carried out. The results were compared with well-known mechanisms, observed in bottom ash from municipal waste combustion

*Keywords: ash, bottom ash, RDF, aging behaviour*

## 1. INTRODUCTION

Waste materials are not only burnt in conventional waste incineration plants, but have also been increasingly used in RDF power stations. In contrast to conventional waste combustion selected, mostly higher-calorific waste fractions are used in RDF power stations as a solid fuel which differs in its composition from the fuel mix of the waste combustion. While the interest in the thermal use of RDF in the past mostly focused on questions concerning processing, price, availability, corrosion and combustion behaviour, the composition of the combustion residues, e.g. the bottom ashes has been more or less neglected. The quality properties and the aging behaviour of bottom ashes from municipal waste incineration were examined thoroughly in the past, mainly in the 1990s. The examinations were mainly carried out with the aim of a complete utilisation of the bottom ashes. Many projects, which were funded by the industry or the government, were accomplished in the field of the aging behaviour and technical leaching, melting or sintering processes. In Germany e.g. publications of Lahl [7, 8], Vehlow [20], Pfrang-Stotz [15], Reimann [17], Zwahr [22], Gallenkemper [5], Sager, Heuss-Asspichler, Heindl [18] and Fraunhofer UMSICHT [9, 13, 10, 11, 14] are to be mentioned. In Europe e.g. the work of Comans and van der Sloot [2], Ecke [3], Polletini [16], Mostbauer and Lechner [12] and Astrup [1] has to be referred to.

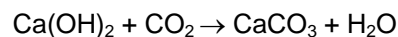
There is relatively little published information regarding bottom ashes from RDF power plants. Based on a changed fuel spectrum compared to conventional waste combustion, changes in the composition of bottom ashes and in their leaching and aging behaviour are not to be excluded. For this reason investigations were made by ThyssenKrupp Xerxon Energy and Fraunhofer UMSICHT concerning quality and aging behaviour of bottom ashes from RDF power stations. In this respect different RDF power stations were sampled to examine the influence of aging processes. The results were compared with well-known mechanisms, observed in bottom ashes from municipal waste incineration.

## 2. AGING OF BOTTOM ASHES FROM MUNICIPAL WASTE INCINERATION

Today, a mechanical treatment of bottom ashes is common practice for the recycling of ferrous and non-ferrous metals and the separation of different corn fractions. In Germany this mechanical treatment is combined with an aging period of three months, in which the bottom ash is stored in heaps.

The proposal of an aging period of bottom ashes was made by Kluge [6]. In 1979 he observed mechanical defects at a construction sector of a road, where bottom ashes from municipal waste incineration were used as a construction material. The defects had been caused by the oxidation of elementary aluminium and the formation of swell able aluminium hydroxide. So it seems to be advantageous to have a storage and aging period before the ashes are utilised. After this period the reactions, which damage the road are completed. Kluge also expected that aging processes have a positive effect on the leaching behaviour of the bottom ashes.

Early investigations by Lahl [7, 8] in the beginning of the 1990s show that natural aging processes lead to a decrease of the leachable content of heavy metals. Lahl stored the bottom ashes in a thin layer and observed a clear decrease of the pH-value in the leachate within a time period of 7-8 days. This effect is related to the reaction of calcium hydroxide from the ashes with carbon dioxide from the air to calcium carbonate.



The carbonatisation leads to pH-values of 10 in the German leaching test DEV S4. At this pH-value the solubility of amphoteric heavy metal compounds like lead and zinc is minimised. The pH-values of fresh ashes in the DEV S4 leaching test are found to be about 12.5. This corresponds reasonably well with the pH-value of a saturated aqueous solution of calcium hydroxide. The high pH-value in the leachate of fresh ashes leads to increasing solubility of the amphoteric heavy metal compounds.

Investigations [9] made in the end of the 1990s show that there are different aging mechanisms which take place in different layers of an ash heap (Figure 1).

- The aging process at the surface of a heap is determined by the process, which was described by Lahl [7, 8], a carbonatisation of calcium hydroxide with carbon dioxide from air or rain water. By the formation of hardly soluble calcium carbonate the pH-value and the concentration of calcium in the leachate decreases. Caused by the decreasing pH-value the concentration of dissolved compounds of zinc, lead and copper decreases as well.
- In deeper layers of a heap no carbonatisation was observed. This is due to a limited transport of carbon dioxide within the heap. Instead, processes take place which are similar to the formation of ettringite ( $3\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot 3\text{CaSO}_4\cdot 31\text{H}_2\text{O}$ ) in concrete and lead to an immobilisation of sulphate. A decrease of leachable lead and other metals could also be observed in deeper layers. But this reduction is not as effective as on the heap's surface. Ettringite was detected by Gallenkemper [5] in aged bottom ashes.

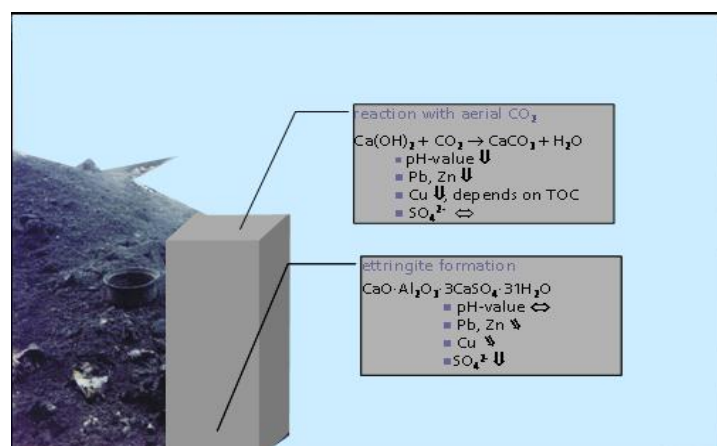


Figure 1: Aging of bottom ashes

Therefore, concepts for a rapid aging process, which accelerates the aging of the bottom ashes by contact with carbon dioxide containing gases, were developed. E.g. laboratory tests were carried out in the 1990s at Fraunhofer UMSICHT to investigate the technical feasibility of such a process [13]. In 2002 a large scale test was carried out, with 170 tonnes of bottom ash and a carbondioxide content of

8 vol-% in the gas [10, 11]. The reaction was carried out in a compost tunnel and the carbonatisation of the whole amount of bottom ash was completed within a period of a few days. The carbonatisation was coupled with a decrease of the pH-value and with a decrease of soluble lead, copper and molybdenum concentrations in German and Dutch leaching tests. Rapid aging makes sense, if there is a free of charge delivery of CO<sub>2</sub>, as in the utilisation of landfill gas and biogas.

### 3. SAMPLING AND PROCEDURE

The investigations were carried out with the bottom ashes of several RDF power plants with grate firing system and a medium flow combustion chamber. The sampling follows the VGB guideline M216H [21] and FDBR guideline [4]. Therefore eight samples between 30 and 100 kg were collected in intervals of one hour and sieved to corn fractions < 40 mm. Large conglomerates, metals and other impurities were separated. The samples of a whole day (8 h) were combined to a master sample of 800 to 1000 kg bottom ash. The sampling at each power plant lasted two days.

The master samples were stored in heaps, with a height of 1 meter. For testing the influence of different aging conditions some heaps were protected from rain with a tarpaulin. Other heaps had direct contact to rain. Initially, during the investigations there was an analytic characterisation of the fresh samples (sample „0“). The heaps were sampled after 7, 30 and 90 days of storing time at the heap surface and in a depth of 30 cm. Among other parameters the leaching test DIN 38414-S4 was carried out and the composition of the leachate was compared with parameters of the German landfill regulation.

### 4. RESULTS

Selected results of the investigations will be discussed in the following. Initially, the influence of different aging conditions on the pH-value and the concentrations of heavy metals in the leachate are explained. Subsequently, there will be a separate contemplation of the specific characteristics of the parameter sulphate. Focus will also be put on the comparison of the new results from RDF power plants to former results from municipal waste incineration. Therefore the physical and chemical character of the solubility behaviour of bottom ash is described by the help of experimental data and theoretical considerations.

#### 4.1 Aging conditions

In figure 2-5 examples are given for an aging process in open heaps with a direct contact to rain water. Figure 2 shows the dependence of the pH-value of the storage time at the surface of a heap. It becomes clear that the carbonatisation of the heap surface is completed no later than after 30 days. After 7 days the leachate shows pH-values of 11. This decrease of the pH-value is low enough to assure concentrations of lead in the leachate which reach the target values stipulated by the German landfill guideline and regulations for the utilisation of bottom ashes. Figure 3 shows the leachates lead concentration in dependence of the aging time. It could be shown that there is no notable lead concentration after a storage time of 7 days. Similar effects can be observed for the concentration of copper and zinc compounds. It is a fact that there is a correlation between the TOC and the copper concentration in the leachate of some bottom ashes from municipal waste incineration [10]. Such a correlation could not be observed in the leachates of the investigated RDF bottom ashes.

Similar to bottom ashes from municipal waste incineration, the processes in the inner layers are different from the processes of the heap's surface. In the inner layers with a depth of 30 cm, a decrease of the pH-value was observed, too, but the pH reaches only values between 11.5-12.0 after a storage time of 90 days (figure 4). So the pH-values in inner layers are still more alkaline than the pH-values at the heap's surface. But the observed pH-value in a depth of 30 cm is low enough to assure low concentrations of lead as is pointed out in figure 5.

Figure 6 and 7 show some examples for aging processes in heaps, which were protected against a direct contact to rain water with a tarpaulin. The carbonatisation at the surface is completed after a storage time of 7-30 days (figure 6). In contrast to this, no decrease in the pH-value could be observed in inner layers (figure 7). Leachates from the samples of inner layers also show higher concentrations in the species of copper, lead and zinc than samples from the heaps surface.

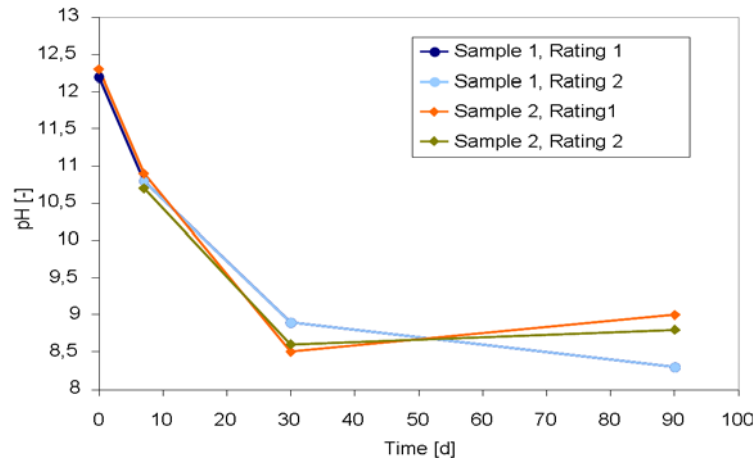


Figure 2: Aging in an open heap with direct contact to rainwater - pH-value versus aging time at the heap's surface

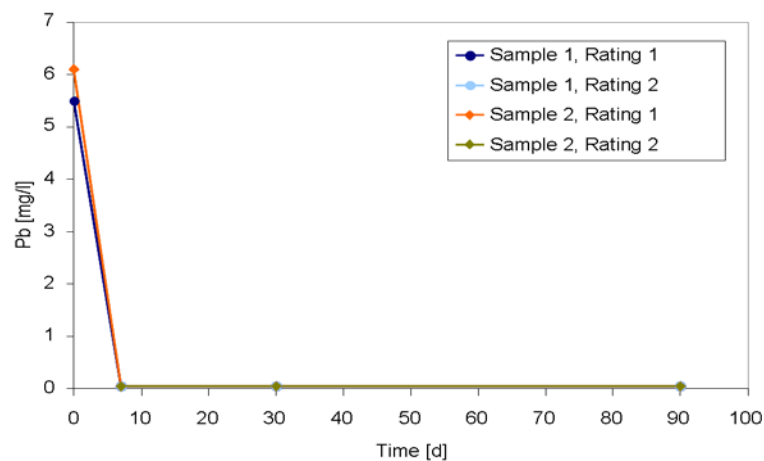


Figure 3: Aging in a open heap with direct contact to rainwater - lead in the leachate versus aging time at the heap's surface

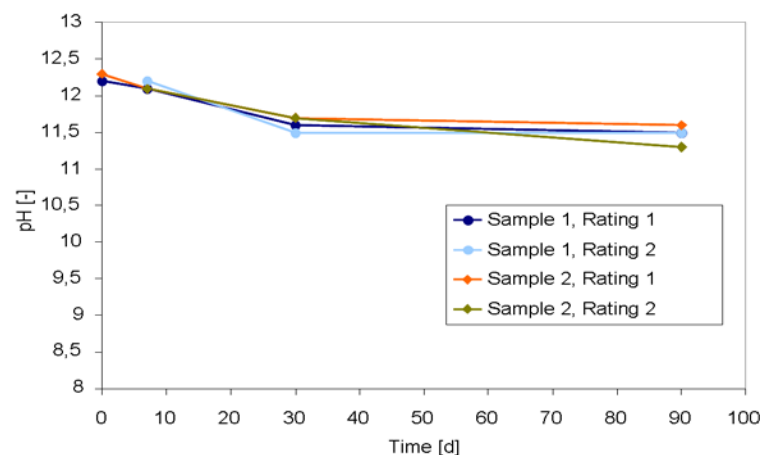


Figure 4: Aging in a open heap with direct contact to rainwater - pH-value versus aging time in inner layers (depth 30 cm)

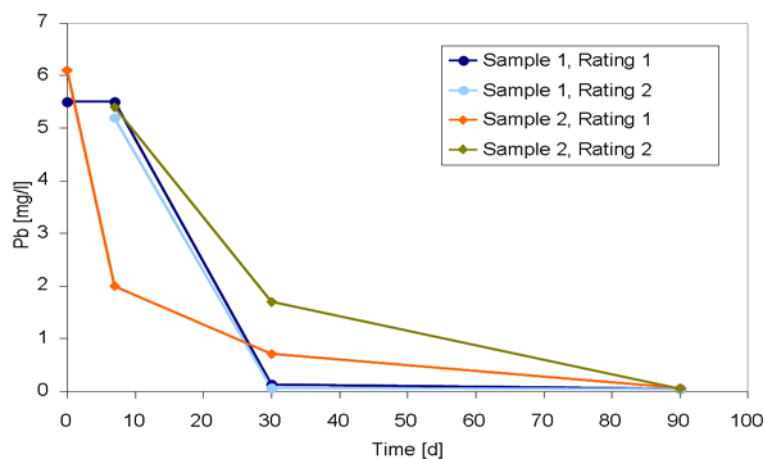


Figure 5: Aging in a open heap with direct contact to rainwater - lead in the leachate versus aging time in inner layers (depth 30 cm)

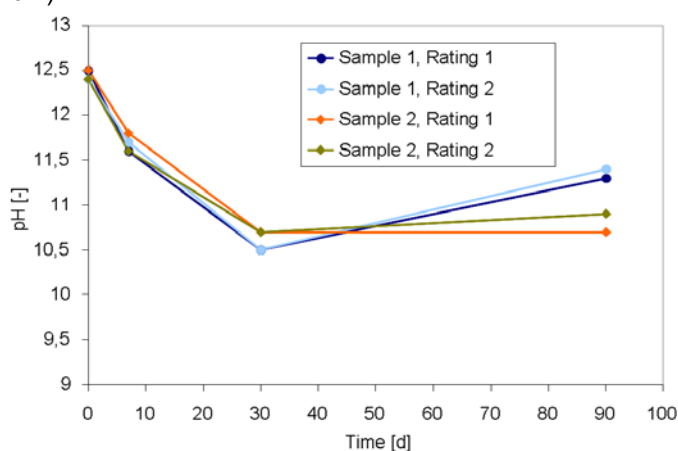


Figure 6: Aging in a heap without direct contact to rainwater (protection with a tarpaulin) - pH-value versus aging time at the heap's surface

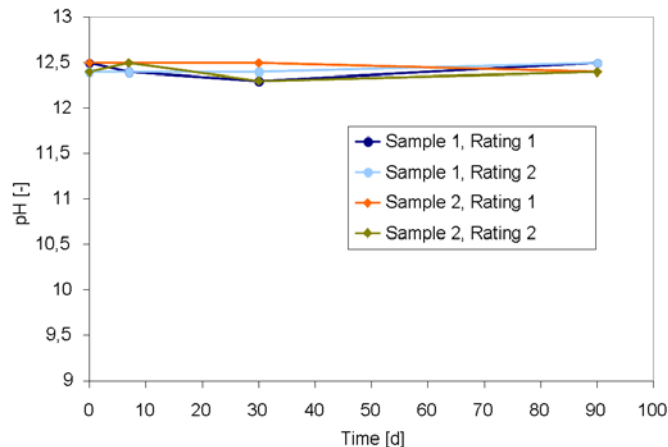
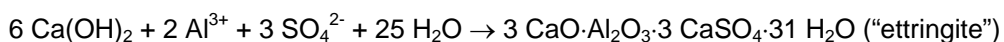


Figure 7: Aging in a heap without direct contact to rainwater (protection with a tarpaulin) - pH-value versus aging time in inner layers (depth 30 cm)

#### 4.2 Solubility of Sulphates

The concentration of sulphate in a leachate is influenced by different drivers which are dedicated by the aging conditions and the power plants fuel composition. In chapter 2 it is described that the mineral "ettringite", a hardly soluble sulphate compound, is formed in the inner layers of heaps from municipal waste incineration bottom ashes.



In bottom ashes from RDF power plants the formation of ettringite takes place, too. Figure 8 shows ettringite formation in form of the resulting decrease of the sulphate concentration. Ettringite is only stable in an alkaline milieu, and its degradation starts at pH-values < 12. The degradation of ettringite is illustrated in figure 9. Firstly, there is a decrease of the sulphate concentration caused by the formation of ettringite, but after 30 days an increase to higher sulphate concentrations could be examined, again. This comes from CO<sub>2</sub>-saturated rain water, which reaches the deeper layers of the heap and leads to a carbonatisation of the inner layers, a decrease of the pH-value and a degradation of formed phases of ettringite.

The composition of the RDF is important as well. As has been pointed out in the upper reaction equation, aluminium cations are necessary for the ettringite formation. Normally they are formed in the pore water by the reaction of elementary aluminium<sup>1</sup> with hydroxide ions. So, no ettringite formation can be observed if there is only a low amount of elementary aluminium in the bottom ashes or accordingly the RDF.

Leachates from the heap surface often show an increase of the sulphate concentration (figure 10) versus aging time. This effect is caused by the specific properties of an aqueous system of calcium hydroxide and calcium sulphate which will be described in the next chapter.

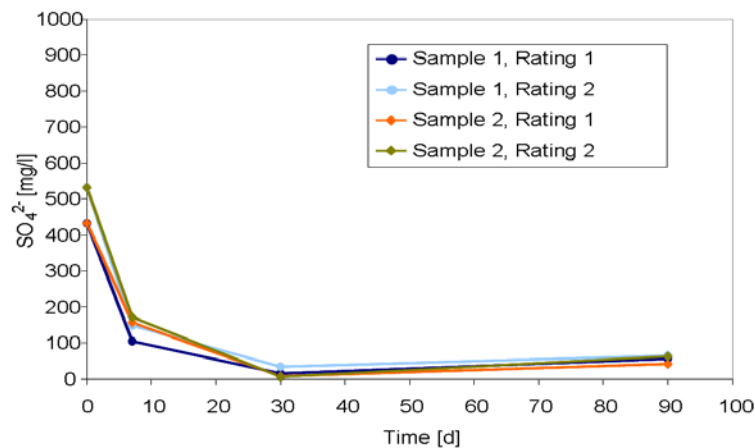


Figure 8: Sulphate concentration in deeper layers (depth 30 cm, open heap with direct contact to rainwater), Case (a) - Formation of ettringite

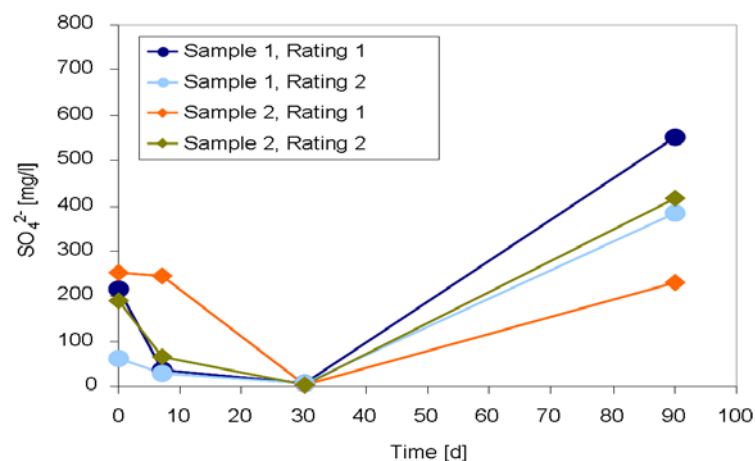


Figure 9: Sulphate concentration in deeper layers (depth 30 cm, open heap with direct contact to rainwater), Case (b) - Formation and degradation of ettringite

<sup>1</sup> Aluminium of tin foils

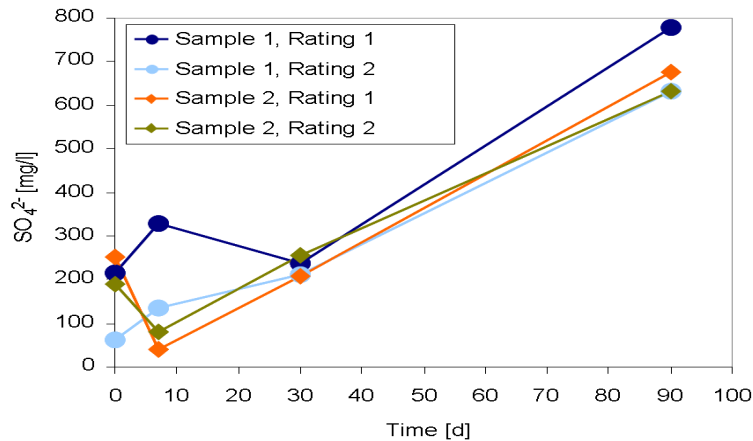


Figure 10: Sulphate concentration at the surface (open heap with direct contact to rainwater), Case (c) - Increasing sulphate concentrations

#### 4.3 Bottom Ashes from Municipal Waste Incineration and RDF Power Plants: Comparison of the Physical and Chemical Behaviour

The aging processes and the composition of leachates from bottom ashes are dominated by the species calcium sulphate dehydrate ( $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$ ) and calcium hydroxide ( $\text{Ca}(\text{OH})_2$ ). It is possible to describe the physical and chemical characteristics of a leachate of fresh bottom ashes by an aqueous system with a solid phase of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} - \text{Ca}(\text{OH})_2$ . Figure 11 describes such a system. The diagrammed dependence of the calcium and sulphate concentration of the pH-value was calculated by the solubility products of calcium sulphate dehydrate and calcium hydroxide and the electro neutrality condition. In the leachates of fresh bottom ashes calcium hydroxide is the dominating species. The calcium concentration is determined by the solubility product of  $\text{Ca}(\text{OH})_2$ , which leads to a pH-value of 12.5-12.6 in aqueous systems. The solved sulphate concentration, which comes from the solid phase  $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$  is determined by the solubility product of  $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$ . At high pH-values an extensive amount of the solved calcium ions are coming from the species  $\text{Ca}(\text{OH})_2$ . This amount of calcium reduces the soluble sulphate concentration, because sulphate is correlated with calcium by the solubility product of  $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$ .

$$K_L (\text{Ca}(\text{OH})_2) = [\text{Ca}^{2+}] \cdot [\text{OH}^-]^2 \cdot f_2 \cdot f_{12} \cdot a_w^2$$

$$K_L (\text{CaSO}_4 \cdot 2\text{H}_2\text{O}) = [\text{Ca}^{2+}] \cdot [\text{SO}_4^{2-}] \cdot f_{22} \cdot a_w^2 \text{ or}$$

$$[\text{SO}_4^{2-}] = K_L (\text{CaSO}_4 \cdot 2\text{H}_2\text{O}) / [\text{Ca}^{2+}] \cdot f_{22} \cdot a_w^2$$

- Solubility constants at room temperature [19]
- $K_L (\text{Ca}(\text{OH})_2) = 2,5 \cdot 10^{-5} \text{ mol}^3/\text{l}^3$  and  
 $K_L (\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}) = 6,31 \cdot 10^{-6} \text{ mol}^2/\text{l}^2$ ,
- [ ] = ion concentration in mol/l,
- $f_1$  and  $f_2$  = activity coefficients of mono and divalent ions (by neglect ion of individual ion properties,
- $a_w$  = water activity (diluted dilutions:  $0,995 < a_w < 1$ )

The calculations of figure 11 are based to the theory of an ideal behaviour with an activity coefficient of 1. In reality there are deviations from an ideal behaviour because further soluble components like chloride influence the ionic strength, the activity coefficients and the solubility behaviour.

At the beginning of the aging process, calcium from the species  $\text{Ca}(\text{OH})_2$  dominates the leachate, so that the sulphate concentration is relatively low. During the aging process  $\text{Ca}(\text{OH})_2$  reacts with carbon dioxide. Hydroxide ions are neutralised and calcium ions are fixed in a solid phase of calcium hydroxide. The pH-value decreases and the system in figure 11 moves to the left side. At pH-values < 11,  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  is the dominating calcium species in the leachate and more sulphate ions become

soluble. Therefore the sulphate concentration in figure 10 increases versus time.

The calculated correlation between the calcium concentration and the pH-value from figure 11 is compared with experimental data in figure 12 and 13. The experimental data were taken from the current investigations with bottom ashes from RDF combustion and from former investigations [9] with bottom ashes from municipal waste incineration. It becomes clear that the behaviour of bottom ashes from RDF or municipal waste incineration is comparable to a large extent (figure 12). Samples from two power plants are not completely comparable (figure 13) and show partly higher calcium concentrations than calculated. This is partly caused by the higher ionic strength in the leachates of these samples. A calculated curve which describes the correlation between calcium concentration and pH-value for an ionic strength of 0,6 mol/l is illustrated in figure 13. The correlation with the higher calcium concentrations is satisfying. An increase of the ionic strength, which is caused by relatively high amounts of chloride, also leads to an increase of the soluble sulphate concentration.

The dependence of the calcium concentration on the ionic strength is illustrated in figure 14. It becomes clear that there is a linear correlation between the calcium concentration and the ionic strength. Furthermore it is possible to define two different areas in the diagram. The first one is the area which includes values of solved calcium  $\leq 400$ -500 mg/l and where the calcium concentration is influenced by the ionic strength. The second area includes values of solved calcium  $\geq 400$ -500 mg/l and belongs to samples which have a relatively high amount of calcium chloride. On the one hand calcium chloride is slightly soluble and increases the soluble calcium fraction, on the other hand more soluted calcium ions leads to an increase of the leachate's ionic strength, too. So there is an iterative back coupling effect which leads to increasing calcium concentrations.

It is known from former investigations [9] with bottom ashes from municipal waste incineration that the lead concentration in a leachate is determined by the pH-value. Figure 15 shows lead concentrations versus pH-value for the RDF bottom ashes investigated. These data are compared with the older data of bottom ashes from municipal waste incineration. It becomes clear that the results are comparable and that the solubility characteristics are determined by the same processes.

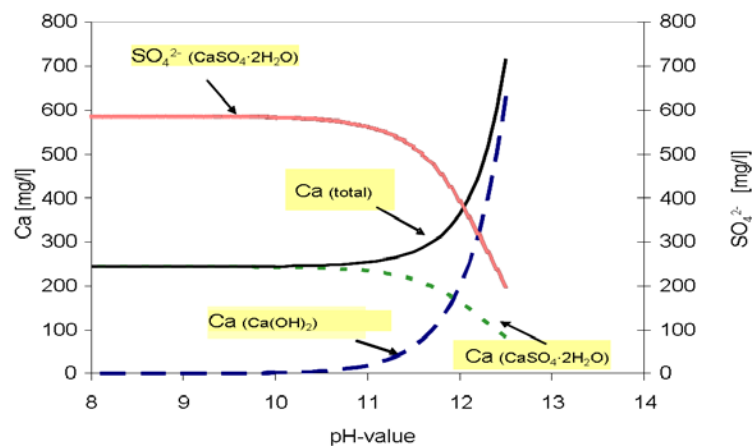


Figure 11: Solubility of sulphate and calcium ions in the system  $\text{Ca}(\text{OH})_2 - \text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ , ideal behaviour (ionic strength = 0, activity coefficient = 1)

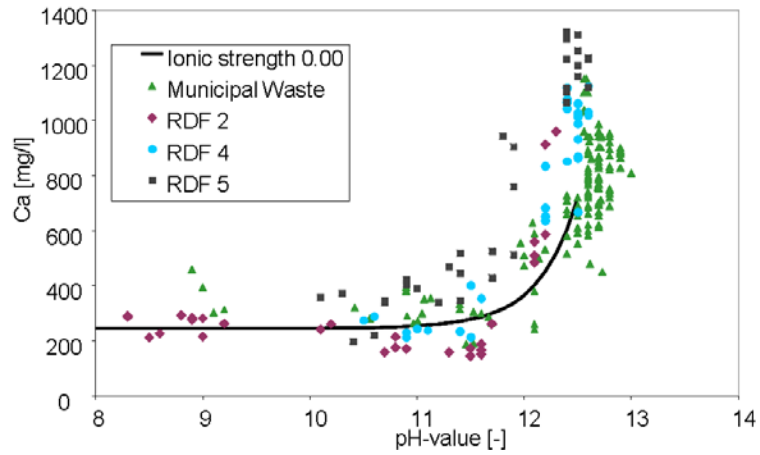


Figure 12: Calcium concentration in the leachate versus pH-value, comparison between bottom ashes from RDF and municipal waste incineration, additionally, the calculated theoretical behaviour for the ideal system  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} - \text{Ca}(\text{OH})_2$  is pointed out

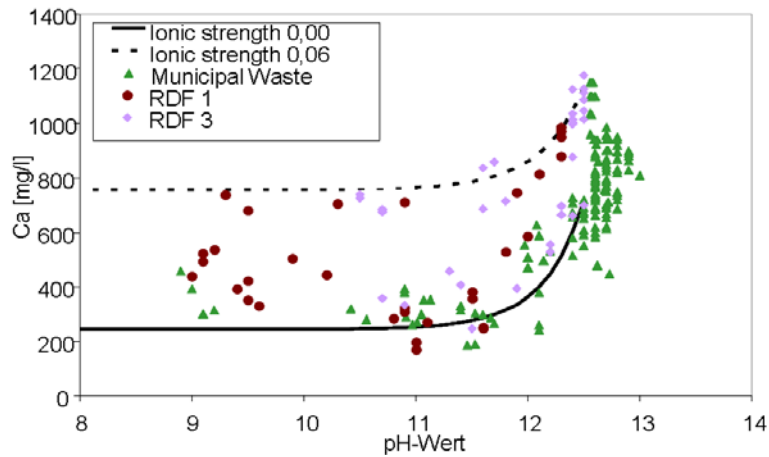


Figure 13: Calcium concentration in the leachate versus pH-value, comparison between bottom ashes of RDF and municipal waste incineration, additionally, the calculated theoretical behaviour for the system  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} - \text{Ca}(\text{OH})_2$  is pointed out for ideal conditions and an ionic strength of 0,6 and an activity coefficient of 0,3

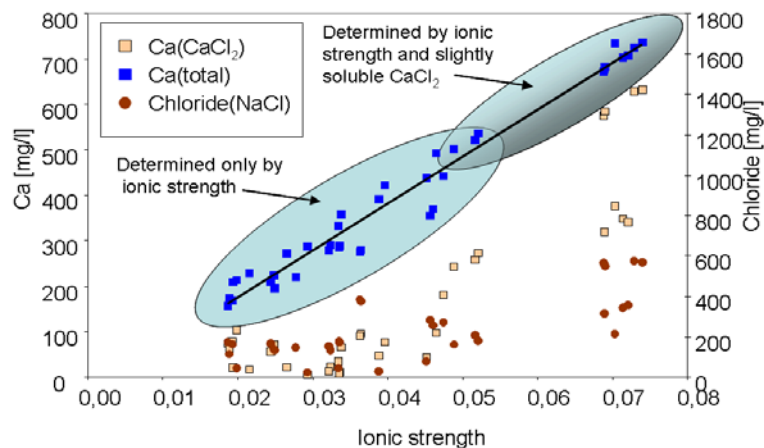


Figure 14: Correlation between the calcium concentration in the leachate and the the ionic strength (experimental data)

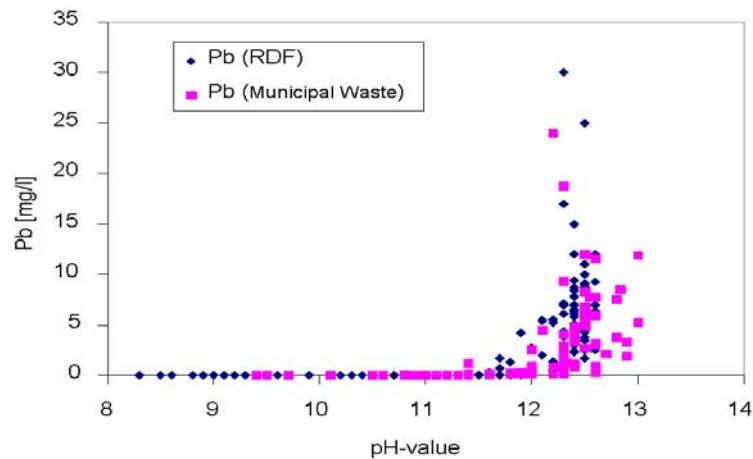


Figure 15: Lead concentration in the leachate versus pH-value, comparison between bottom ashes from RDF and municipal waste incineration

## 5. CONCLUSION

The aim of the project was the investigation of the aging behaviour of bottom ashes from RDF power stations. Therefore bottom ashes of different RDF power stations were sampled and stored in heaps under different aging conditions. The results were compared with data taken from former investigations with bottom ashes of municipal waste incineration. One of the results is the necessity to ensure, that rain water is able to reach the deeper layers of a heap. Finally, it can be pointed out that there was no difference in the aging behaviour of bottom ashes from the investigated RDF power plants and bottom ashes of municipal waste incineration. But the database of bottom ashes from RDF power plants is still very small, so that the results have to be verified with more samples.

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