

QUALITY IMPROVEMENT OF MSWI BOTTOM ASH BY ENHANCED AGING, WASHING AND COMBINATION PROCESSES

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Abstract

Natural aging of MSWI bottom ash is known to have the effect that the leaching of most constituents decreases, especially with respect to heavy metals. Tauw Milieu has developed a process to speed up the aging process by controlling storage conditions and leading a CO₂ containing gas mixture through the material. Aging process conditions were optimized and the process was then tested on bottom ash samples from Dutch incinerators with a volume of up to 500 kg. Results showed that within several weeks a quality improvement is achieved that corresponds to one or more years of natural aging. Leaching of heavy metals, DOC and most macro-elements decreased by max. 90%. Leaching of salts did not decrease. This means that generally speaking, the requirements of the Dutch Building Materials Decree are met for the material after this way of processing, although this may not always be true for bromide.

Washing processes are especially suitable for the removal of soluble salts like chloride- and bromide salts. The removal of sulphate is more difficult owing to its poor solubility. Research into washing processes has focused on minimization of the amount of water and optimization of conditions such as pH, percolation rate etc. Although a quality improvement was achieved, relatively strongly leaching bottom ash did not meet the requirements of the Building Materials Decree.

With a combination of washing and aging, the leaching of salts as well as heavy metals can be reduced to low levels.

Based on a preliminary design of an installation for the treatment of 100,000 tonnes of bottom ash per year, the exploitation costs of aging, washing and combination processes were calculated.

1 INTRODUCTION

In the Netherlands, the environmental quality of building materials to be applied on or in soil is mainly assessed on the basis of its leachability. The limit values for the immission of inorganic contaminants into the soil are listed in the Building Materials Decree [1], in mg/m² for a period of 100 years. These limit values are based on the principle of 'marginal burdening of the soil': the quality of the soil or groundwater below the used material may only be affected marginally by the material's leaching. This has been translated into a contaminant concentration increase of 1% of the target value over a period of 100 years, in a 1-metre-thick soil layer directly below the used material.

This leaching standard results in a classification of materials into two categories:

- 1) materials that hardly leach can be freely used; no special demands are set on their application other than that it must be possible to remove the materials, and that therefore they should not be mixed with the soil (category 1).
- 2) materials that leach more than that may be applied on the condition that a watertight top lining be applied (infiltration \leq 6 mm/year) and that the material be used only at a height of \geq 0.5 m above the mean highest groundwater level (category 2).

Materials that do not meet the immission limit values at an infiltration rate of ≤ 6 mm/year are not allowed to be applied. There are a few exceptions to this. One of these is MSWI bottom ash, for which however extra stringent demands are imposed on the top lining of the construction.

The immission requirements laid down in the Building Materials Decree can be translated into emission values as measured in column tests, with the help of a calculation model. Table 1 presents the emission requirements for some components.

Table 1 Maximal leaching of building materials, as measured by means of a column test, at an application height of 0.7 m. Emissions in mg/kg d.w. at a cumulative Liquid/Solid value of 10.

Parameter	Category 1	Category 2
Antimony*	0.045	0.43
Arsenic	0.88	7.0
Cadmium	0.032	0.066
Chromium	1.3	12
Copper*	0.72	3.5
Lead	1.9	8.7
Molybdenum*	0.28	0.91
Nickel	1.1	3.7
Zinc	3.8	14
Bromide*	2.9	4.1
Chloride	600	8,800
Sulphate	1,140	22,000

* potentially critical parameters for MSWI bottom ash

Although the application of MSWI bottom ash can continue for the time being (against higher costs, as stricter demands are set towards the top sealing), the waste processing branch is striving for a quality improvement of up to category 2 level. In the long run, category 1 should be attainable.

The quality improvement of MSWI bottom ash can be attained by input measures (such as the MSWI refusal to accept certain waste materials for processing), by process measures and by an end-of-pipe treatment of the bottom ash. The article at hand concentrates on quality improvements achieved by the after-treatment of slags, a process in which the leaching behaviour is of major importance.

In order to improve its leaching behaviour, MSWI bottom ash can be subjected to several processes, that amount to either the fixation or the removal of contaminants. Contaminants can be fixed by e.g. a melting process, adding agents, or aging the material. Tauw Milieu has developed an enhanced aging process, in which the leachate quality rapidly improves within a short period of time [2]. This method is particularly successful when applied to heavy metals. On the other hand, the quality of the material can be improved by washing processes; these are particularly effective for easily soluble salts.

At the request of the Waste Processing Association, Tauw Milieu has optimized the aging process and scaled it up to 500 kg. At the same time, research was carried out into optimizing the washing process, and into combined washing and enhanced aging processes.

The research objectives were the process-technical upgrading of the above processes and finding the most cost-efficient way of making MSWI bottom ash meet the category 2 demands of the Building Materials Decree.

2 THEORETICAL BACKGROUND

It is a generally known fact that when MSWI bottom ash is stored outdoors, the quality of its leachate improves considerably after some months to some years [2-4]. This improvement is the result of several different reactions, namely hydration, carbonation, the (microbiological) oxidation of organic substances, the oxidation of metals, various precipitation reactions (including the formation of proto-clays) and the weathering of glass phases. Most of these reactions help to improve the leaching characteristics. Through carbonation, a pH of about 8.5 is obtained, at which the solubility of many metals in the form of cations (copper, lead, zinc, etc.) is minimal. The oxidation of organic substances results in decreasing emissions of COD and metals that are complexed by organic substances. The oxidation of metallic iron and aluminium, various precipitation reactions and the formation of clay minerals improve the MSWI sorption behaviour. In addition, metals can be incorporated within the newly-formed matrices. One unfavourable reaction during aging is the mobilization of sulphate, which is believed to be the result of the slow hydration of anhydrite [5].

The aging process can be enhanced by bringing the material into contact with air containing a raised CO_2 concentration (enhancement of carbonation), raising the temperature, optimizing the moisture content, and cocultivation with microorganisms (which play an important role in the degradation of organic substances).

In the washing processes, readily and poorly soluble components can be distinguished. Readily soluble components, e.g. chloride and bromide, can be easily removed with little water. Poorly soluble components, e.g. heavy metals, can only be partially removed by washing. Even if only the leachable fractions of these elements are to be removed, more washing water will be needed than for such a substance as chloride. As these elements are usually subsequently still supplied from the matrix, a greater quantity of contaminants should be removed in the washing process than one would expect based on the leaching test.

As the treatment of the washing water is one of the major cost items, the volumes of washing water should be kept to a minimum; in other words, the solubility of the contaminants should be maximal. In order to obtain maximal solubility there are various treatment methods, e.g. pH correction, the addition of complexers, or raising the temperature. Earlier research [6] has shown that the use of such complexers as EDTA is less effective than pH correction. One of the reasons may be that complexers partly adsorb to the bottom ash and are released again during the next leaching test. However, pH correction has the drawback that salts remain behind in the material (chloride, if hydrochloric acid is used for pH correction). These remainders can be removed by again washing with water.

3 SETUP OF THE INVESTIGATION

As two of the most critical elements are copper and molybdenum, the tests were predominantly carried out on the bottom ash of two Dutch MSWI plants with the highest copper and molybdenum leaching concentrations. At a later stage, a third MSWI, the bottom ash of which showed more average leaching results, was involved in the investigation. In total, batches from 12 different production periods were investigated.

The aging process was optimized by varying the process parameters at a small scale (1 - 2 kg). The range of the investigated conditions is presented in Table 2. In total, 26 experiments were carried out. Next, the kinetics of the process were investigated at a larger scale (50 - 100 kg) by regularly taking samples during the treatment. These tests were carried out at optimal conditions with only the CO_2

concentrations being varied (between 4 and 40%). Finally, the established optimal process conditions were verified at a scale of 500 kg, paying attention to such technical aspects as pressure drop and possible quality differences as a function of the layer thickness.

Table 2 Ranges of investigated parameters for optimization of enhanced aging

Parameter	range
Temperature (°C)	25 - 80
Moisture content (% of d.w.)	15 - 30
Gas flow rate (dm ³ /kg, h)	0.5 - 5
Microbiology	inoculation, addition of nutrients
CO ₂ -content (vol.percent.)	4 - 40
O ₂ -content (vol.percent.)	9 - 20
Residence time (days)	3 - 56

Two kinds of washing processes can be distinguished: processes intended to remove salts (metals are fixated through aging, therefore it is in fact a combined process) and processes aimed at removing both salts and heavy metals. For both process types, the process conditions were optimized by varying the different parameters at a scale of 10 kg. Table 3 presents the parameter variation ranges.

Table 3 Ranges of investigated conditions for optimization of washing processes

Parameter	range
Washing processes for salts	
L/S-value (l/kg)	0.5 - 5
Percolation-rate (l/kg,day)	0.2 - 2
Intensity of water contact	percolation - shaking
Temperature (°C)	20 - 60
Washing processes for metals and salts	
pH	3 - 4
L/S-value (l/kg)	1 - 20

Finally, combined processes were tested, in which the sequential order of the processes, the L/S-ratios and the scale sizes were varied. At a small scale (1-2 kg) and L/S values of 1 - 5, three process sequences were tested: washing-aging, aging-washing, and aging-washing-aging. At a scale of 100 kg and L/S value of 1, two process sequences were tested, namely washing-aging and aging-washing. Based on the test results, global designs were drawn up for practice installations for some of the tested process configurations, and cost estimates were made of both the investment and exploitation costs.

4 WORKING ROUTINE

The bottom ash was sampled by employees of the concerned MSWIs, in conformity with the common quality assurance procedures. The samples were mixed samples taken over a 2-week production period.

All aging tests were performed in the form of columns through which a gas mixture was blown upwards. The gas mixture was composed of air, CO₂ and N₂, at the desired ratios. The mixture was furthermore saturated with water vapour. The temperature in the columns was created by supplying heat from an external source.

Samples were taken during (intermediate samples) or at the end of the tests by emptying the columns and composing mixed samples. The large-scale test material was also sampled per layer of ca. 0.5 m; the total bed height was 1.5m. For the small-scale tests (up to 2 kg), crushed material (<4 mm) was used; the other tests were performed with material of the usual size. Any released condensate or drainage water was recirculated. The leached concentrations were determined at the beginning and at the end of the experiments, by means of shaking tests (L/S 20) and column tests (L/S 10), in accordance with NEN 7343 and NEN 7349, respectively. Intermediated leaching tests only involved shaking tests. The leachate was analyzed for As, Cd, Cu, Cr, Mo, Ni, Pb, Sb, Zn, Ca, Cl, SO₄, COD, pH and E.C. The treatment performance was expressed in terms of the decrease (in %) of the leached quantities in relation to the initial values.

The washing tests were performed by leading tap water upwards through the columns filled with bottom ash. The columns were flushed until the desired L/S ratio was obtained. Afterwards, leakage water was added to the washing water. pH-control experiments were conducted as stirring tests after the addition of concentrated hydrochloric acid. In combination tests, columns were both vented and flushed in certain sequential orders. The analysis package was the same as that for the aging tests.

As the composition of the material remained more or less the same, it was extensively analyzed before the test, but after the test only dry weight and organic substance were determined. Furthermore, civil-technical parameters were determined before and after the large-scale tests, namely grain size distribution, crushing factor, and iron- and unburned material content.

5 RESULTS

5.1 Enhanced aging

The optimal process conditions for enhanced aging are: a temperature of 60°C, a moisture content of 15-20%, gas composition of 4-8% CO₂ and 18-20% O₂, a gas flow rate of 0.5 m³/tonne/hour, and a maximum residence time of 4 weeks. The required residence time depends on the initial quality of the bottom ash. Where category 2 values are only slightly exceeded (copper up to 200%, molybdenum up to 50%), a residence time of a week will suffice. Stimulation of the microflora is not required.

Table 4 Leached emissions (in mg/kg d.w.) for MSWI 1 before and after enhanced aging (four weeks), and performance (% decrease leaching). Shaking test L/S 20, averages of 4 tests at a scale of 100 - 500 kg.

	fresh material		aged material		performance
	emission	class	emission	class	(%)
pH	10.7	-	8.1	-	-
E.C. (1)	913	-	905	-	-
Cl	1430	Cat. 2	1430	Cat. 2	0
SO ₄	3170	Cat. 2	6790	Cat. 2	-114
Sb (2)	0.23	Cat. 2	0.24	Cat. 2	-4
Cu	6.1	>Cat. 2	0.63	Cat. 1	90
Zn	0.82	Cat. 1	<0.13	Cat. 1	>84
Pb	0.78	Cat. 1	<0.06	Cat. 1	>92
Mo	2.00	>Cat. 2	0.72	Cat. 2	64

(1) electrical conductivity, in $\mu\text{S}/\text{cm}$

(2) Sb is column test emission (L/S 10)

Table 4 summarizes the results of the large-scale tests with material from MSWI 1. These show that good results (80-90% decrease in leaching) can be obtained for cationic metals and COD. After treatment, the material meets the category 1 requirements for these parameters. The performance with regard to molybdenum is less effective (over 60%), whilst for antimony and chloride little or no improvement is measured, and sulphate leaching increases, as expected. In spite of this, the material as a whole meets the requirements for classification as a category 2 material.

The results of tests on bottom ash of other investigated MSWIs are largely the same, with higher performances for antimony, (58% for MSWI 2, and 42% for MSWI 3).

A general conclusion is that although metals and metalloids can be fixed to a sufficient degree, enhanced aging is not effective for salts. If bromide is critical, or the category 1 level should be obtained, aging will have to be combined with a washing process.

5.2 Washing

The tests showed that chloride can be removed up to category 1 level, using little washing water (L/S \approx 0.5) at short retention times (0.5 day, shorter retention times were not tried). As bromide has similar chemical properties, these conditions probably also apply to this element.

Sulphate is much harder to remove. The category 1 value was not even obtained after washing at L/S 5 and a residence time of 5 days. The removal performance improves only slightly (> 10%) when raising the temperature from 20 °C to 60 °C. Lowering the percolation rate with a factor 10 is more effective, with 28% more sulphate being removed (refer to Table 5). However, also at these conditions - that are financially not very attractive - the category 1 value is not attained.

**Table 5 Effect of percolation rates during washing of MSWI bottom ash.
Washing up to L/S = 5, T = 20 °C.**

Parameter	percolation rate 0.2 l/kg,d ($\tau = 25$ d)		percolation rate 2 l/kg,d ($\tau = 2.5$ d)	
	Load (mg/kg dm)	Performance (%) (1)	Load (mg/kg dm)	Performance (%) (1)
Removal				
Chloride	1390	102	1000	74
Sulphate	2390	68	1235	35
Leaching				
Chloride	100 (2)	93	260 (2)	81
Sulphate	1020 (2)	71	1660 (2)	53

(1) removal (in per cent) with the washing water, or decrease (in per cent) in leachate of washed material in relation to the initial leaching values, as measured during shake test L/S 20.

(2) leaching of washed material, measured during shake test L/S 20.

Table 5 shows that kinetic aspects play a role in the washing process. This may both be related to transport (diffusion from the pores of larger ash particles) and to mineral conversions. It is remarkable that even for the readily soluble component chloride, percolation rate/residence time differences have an effect. This effect is however significantly more noticeable for sulphate: at low percolation rates the removal performance is 30% higher. The performance on the basis of leaching is only 18% higher; in absolute quantities, an additional 1,115 mg of sulphate has been removed, but the leaching has decreased by 640 mg/kg dw. This illustrates that the relation between removal and residual leaching is not linear - 'new' sulphate becomes available through subsequent supply. Tests during which the material was shaken while being washed did not result in higher removal efficiencies in comparison to the percolation washing method.

Conditions at which metals always meet the category 2 standards were not found. Lowering the pH to values of < 4 , or raising the L/S ratio to 20 had an unsatisfactory and sometimes even negative influence on the residual leached quantities. At L/S 1 and pH 4 the best results are obtained. It is however possible that in the case of less strongly leaching bottom ash, the category 2 level can be attained by washing.

In summary, it can be concluded that chloride and bromide can easily be removed (little washing water, short residence time), but that there are no easy methods to remove sulphate to category 1 level. The bottleneck probably is the mineral form in which the sulphate is present.

As regards metals, the category 2 level could not be reached in any of the tested samples by means of washing at acid pH and variable L/S. Washing may hold good perspectives for bottom ash in which metals only slightly exceed the category 2 limits.

5.3 Combinations of washing and aging

The sequential order of washing and ageing can be important for several reasons:

- 1) there may exist positive or negative interactions between both processes;
- 2) washing after aging will result in less contaminated washing water;
- 3) after aging, sulphate is more mobile, and so less washing water might be required in order to attain the category 1 value.

Three variants were investigated in small-scale tests, namely washing (L/S 5) followed by aging (6 weeks); 3 weeks aging followed by washing (L/S 5) followed by 3 weeks aging; and aging (6 weeks) followed by washing (L/S 1). The investigation showed that the end quality of all samples was almost the same, always well under category 2 limits. For many components, category 1 values were attained, but not for antimony. Sulphate only approached this value after washing up to L/S 5.

Table 6 presents the contaminant loads that were removed during the washing process. Metal and metalloid loads in the washing water prove to decrease if aging of the material precedes the washing process. This is not so for salts - although sulphate loads are lower at the aging/washing sequence, the leaching results show that washing at L/S 1 is not sufficient for obtaining the desired quality improvement. Interpolation of the measuring data shows that for washing after aging, 20 - 25% less washing water is needed in order to attain category 1 sulphate values.

It is however obvious that this process order does not result in the desired low washing water volume of about 1 m³/tonne (L/S 1). The solubility of sulphate remains a limiting factor, also after aging has taken place, so that relatively much washing water (L/S 3 - 7, depending on the sulphate content in the bottom ash) will still be required.

Table 6 Loads in washing water in mg/kg ds in relation to process sequences of washing and ageing

MSWI	MSWI 1			MSWI 2		
	W(L/S 5)/ A(6 w.)	A(3 w.)/W(L/S 5)/A(3 w.)	A(6 w.)/ W(L/S 1)	W(L/S 5)/ A(6 w.)	A(3 w.)/W(L/S 5)/A(3 w.)	A (6w.)/ W(L/S 1)
Salts						
Chloride	1000	1175	940	1325	1175	1450
Sulphate	4500	4700	1950	6250	6250	2350
Metals						
Antimony	0.28	0.07	0.05	1.30	0.11	0.11
Copper	1.65	0.65	0.19	5.50	2.15	1.30
Molybdenum	1.35	0.11	0.09	0.85	0.11	0.14
Zinc	0.25	1.6	0.85	0.75	1.20	0.83

(1) W = Washing at stated L/S value; A = aging at stated residence time

Tests with bottom ash from three MSWIs, at a scale of 100 kg, during which washing at L/S 1 was applied before or after aging, proved that:

- 1) when the fresh material does not leach too much (values of about 50% above category 2), washing may suffice in order to reach category 2 levels;
- 2) washing in combination with aging is useful when chloride or bromide must be removed, or the molybdenum removal efficiency increased. The Mo-performance of the combination process (70 - 95%) lies on average about 20% above the performance of just aging. Therefore, washing is a good option for increasing the Mo-performance;
- 3) washing could furthermore be used to reduce the required aging residence time. Tests however show that for strongly leaching bottom ash, a far-reaching reduction of the residence time to 1 week is not feasible. Bringing down the residence time from 4 to 3 weeks however might be possible. Economic considerations also play a role in this.

6 COSTS OF QUALITY IMPROVEMENTS

The costs of MSWI bottom ash treatment were estimated on the basis of a conceptual design of an installation with a capacity of 100,000 tonnes per year. The installation comprises a number of roofed, concrete basins. The process evolves in batches, the same basins can be used for aging and washing by leading either water or gas through the ash. On the basis of MSWI bottom ash quality differences, four different treatment options were worked out:

- 1) enhanced aging during 1 week;
- 2) enhanced aging during 1 week, followed by washing up to L/S 1;
- 3) enhanced aging during 4 weeks;
- 4) enhanced aging during 4 weeks, followed by washing up to L/S 1.

Longer residence times of course lead to larger installations, whereas for the washing water an extra buffer tank is needed in order not to overload the water treatment installation.

The estimation of the investment costs includes the costs of construction of the concrete units, including roofing, foundation, costs of the land, ventilators, gas mixture feed and outlet pipes, transport belts for the supply of the material, and a switch box.

In first instance it was assumed that purified flue gas was to be used for carbonation, and as an alternative, the purchase of CO₂ was included. A schematic drawing of the installation is presented in Figure 1.

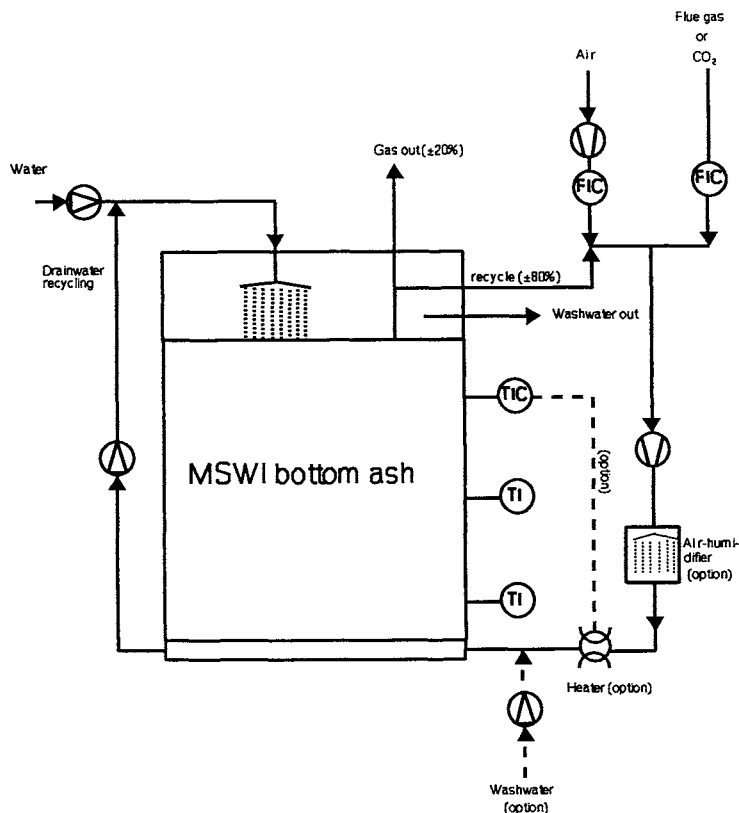


Figure 1 Schematic representation of the installation for enhanced aging and washing of MSWI bottom ash

For an assessment of the exploitation costs, the following items were taken into account:

- * interest and repayment (annuity depreciation, instalment of 10 - 20 years, 8% interest);
- * maintenance: 2% of construction and 6% of mechanical-electrical investments per year;
- * personnel: 0.5 - 0.8 man year for operation;
- * use of shovel (and personnel) for emptying the installation;
- * electricity, insurances;
- * costs of water treatment: all in costs of NLG 3 - 6 per m³ (provided that it is allowed to discharge such salts as chloride and sulphate);
- * costs of purchase of CO₂: NLG 0.25 per kg.

The results of the calculations are summarized in Table 7. The inaccuracy of these estimations is +/- 40%. Variations in depreciation periods and higher costs for CO₂ purchase are included in this margin.

Table 7 shows that enhanced aging is the cheapest way of improving the quality of MSWI bottom ash. If bromide and chloride are critical components, and washing is therefore required, applying only the washing process is an alternative provided that the leachate does not contain too high concentrations of e.g. copper. If strongly leaching bottom ash is the basic material, a combination of washing and aging will be the most attractive option from an economic viewpoint. As water treatment costs are the predominant cost factor in the washing process, higher washing water volumes will sooner lead to higher costs than longer residence times do.

Table 7 Costs of MSWI bottom ash quality improvement, in NLG per ton

Process	Costs in NLG per ton bottom ash
1 week aging	5.=
washing up to L/S 1	7.= - 10.50
1 week aging + washing up to L/S 1	9.50 - 12.50
4 weeks aging	10.50
4 weeks aging + washing up to L/S 1	14.50 - 17.50

7 DISCUSSION AND CONCLUSIONS

Enhanced aging is an effective technique for reducing the leaching of metals and organic substances within a short period of time (1 - 4 weeks). A reduction can be obtained of 80 - 90%, and for molybdenum of over 60%. Aging does not result in reduced leaching of salts - in order to achieve this the aging process should be combined with washing. Chloride and most probably also bromide salts can easily be removed by washing at an L/S value of 0.5. In order to remove sulphate, considerably higher L/S values are required, namely 3 - 7.

From the viewpoint of the desired final quality and costs, the following (combinations of) processes are the most attractive:

Objective	Bottom ash quality (1)	process choice
Category 2	metals critical, salts not salts critical, metals moderately raised salts critical, metals strongly raised	aging washing aging + washing
Category 1	difference not of importance	aging + washing

(1) quality in relation to category 2 limit values

The costs of aging are between NLG 5 - 10. = per ton bottom ash (depending on the required residence time), of washing NLG 7 - 11. =, and of combination processes NLG 10 - 18. =. In order to reach the category 1 level, costs of about NLG 25. = per ton should be taken into account.

At the moment, upscaling and optimization of the process is being investigated on pilot scale (50 ton). During this investigation, both aging and combinations of aging and washing are examined.

8 ACKNOWLEDGEMENT

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