

ENVIRONMENTAL AND HEALTH ASPECTS OF ASHES PRODUCED AT CO-COMBUSTION OF BIOMASS

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ABSTRACT

An agreement was made between the Dutch government and the electricity producing companies for reducing the CO₂-emissions with 3 million ton in 2008. In practise this means that about 25% of the coal by dry mass will be replaced by secondary fuel. Since 1993 KEMA has researched the effect of direct co-combustion in more than fifty test series. In these test series secondary fuels were co-combusted in proportions up to 42% by dry mass. During a lot of these tests the relationship was established between fuel composition on the one hand and the composition of the ash, gypsum and flue gases on the other hand. It appears that the behaviour of (trace) elements during co-combustion is (almost) similar to the behaviour during full coal firing.

An assessment of technical, environmental and health aspects of the co-combustion ashes was made. The health and safety properties of coal fly ash from co-combustion are assessed accordingly to the KEMA-DAM[®] (Dust Assessment Methodology). The judging whether the ash is hazardous or not is done accordingly to the EWC by the KEMA TRACE MODEL[®].

It appeared that co-combustion coal ash, as produced in the Netherlands, is equivalent to coal ash from full coal-firing with respect to environmental, technical and occupational health properties. Research made clear that ashes generated by high percentages of co-combustion are able to meet the technical requirements of the European standard (EN 450) for utilisation as pozzolanic filler in concrete. Performance tests of concrete confirmed the suitability of these fly ashes. The applied research indicates that there is no reason to regard coal fly ash "dust" as harmful. No increased health risk will be caught under circumstances, which generally meet the requirements laid down for nuisance dust in the occupational environment. This means that the standards for nuisance dust can be applied. Accordingly to the EWC (European Waste Catalogue) the coal fly ash is categorised as non-hazardous.

INTRODUCTION

The Dutch government promotes co-combustion of biomass or other secondary fuels to reduce CO₂ emissions. An agreement has been made between the Dutch government and the electricity producing companies aimed at reducing CO₂ emissions by 3 million metric tons in 2008. In practice this means that about 13% of the coal in terms of energy or about 20% in terms of dry mass will be replaced by secondary fuel. Today co-combustion is daily practice in almost all coal-fired power stations.

The by-products that are produced during co-combustion of secondary fuels must comply with European and Dutch legislation. KEMA has examined co-combustion by-products since 1993. In this paper the results of research into the environmental and health properties of coal-fly ash, produced at co-combustion, are discussed. Besides some concluding remarks are made regarding the technical suitability of co-combustion ashes for utilisation as pozzolanic filler in concrete.

OVERVIEW OF THE TEST SERIES

Since 1993 KEMA has researched the effect of direct co-combustion in more than fifty test series. In the first three years seven tests were performed at the KEMA 1MW_{th} test boiler, followed in the next seven years by twenty-three tests on real scale at all seven Dutch coal-fired power stations. In these tests secondary fuels were co-combusted in proportions up to 10% by dry mass.

In 2001 test series were started in which secondary fuels were co-combusted in proportions more than 10% by dry mass. At first three test series in the KEMA 1MW_{th} test boiler, followed by five tests on real scale at three different power stations.

The ashes were used as a raw material to make concrete test cubes. These cubes were subjected to leaching tests and tested to determine the appropriate technical parameters^(3,7,8). Moreover, in twenty-five of these tests the relationship was established between fuel composition on the one hand and the composition of the ash, gypsum and flue gases on the other hand. Also leaching tests were performed on the ashes. The importance of the relationship is discussed in the next chapter.

BEHAVIOUR OF ELEMENTS DURING COMBUSTION

In the tests series were the elemental composition of the fuel and the outgoing streams were established, parameters such as the relative enrichment factors of the elements in the ashes and the removal efficiencies of the gaseous elements in the ESP and FGD were determined. These relative parameters are independent of the particular fuel and can be compared to those, which are obtained by 100% coal-firing. It appears that these parameters obtained for co-combustion are about equal to 100% coal firing. This is clear established for co-combustion in proportion up to 10% by dry mass. For co-combustion of more than 10% the first results indicate that this is also true for most elements. Only the so called "class-three" elements, which are the elements that leave

the power station entirely or partly in the gaseous phase via the flue gases, are in some cases captured in the ash to a larger part. It concerns sulphur, mercury, chlorine and fluorine. Particularly calcium present in the secondary fuel is an important parameter with respect to the capture of these elements.

If the behaviour of the elements hardly changes, it can be assumed that the speciation will not change either. This hypothesis is confirmed by thermodynamical calculations for almost all-relevant elements and by direct measurements of the speciation of phosphorus and chromium. If the behaviour hardly changes, it is justified to apply the models developed for 100% coal firing to simulate co-combustion.

KEMA has developed a model that calculates the composition of the ash and the emission into the air as a function of fuel and plant data. The model is called the KEMA TRACE MODEL[®]. Not only the total elemental composition of the ash is calculated, but also the composition of the inhalable fraction of the ash, the order of magnitude of the leached elements and the radioactivity. The composition of the inhalable fraction of the ash is needed for the judgement of the health properties of the ash. This will be discussed in the next chapter.

JUDGEMENT OF THE HEALTH PROPERTIES OF COAL FLY ASH

In contradiction to coal, for coal fly ash there is no exposure limit for occupational health. If a certain substance has to be judged on its toxic properties, various toxicological tests have to be performed. A great deal of research has been conducted into the health implications of working with coal fly ash produced at 100% coal firing. Data from cell test systems and animal experiments indicates that normal levels of exposure (i.e. exposure to levels below the limit for nuisance inhalable substances) are not likely to have any significant health implications^(6,2). The results of epidemiological research confirm this conclusion⁽¹⁾. The applied research indicates that there is no reason to regard coal fly ash "dust" as harmful. No increased health risk will be caught under circumstances, which generally meet the requirements laid down for nuisance dust in the occupational environment. This means that the standards for nuisance dust can be applied^(4,5). In the Netherlands the standards for nuisance dust are 5 mg.m⁻³ for the respirable fraction and 10 mg.m⁻³ for the inhalable fraction.

If toxicological test have to be performed on the ash originate from combustion of every other type of coal as that was investigated or every case of co-combustion, this will be time consuming and expensive. Therefore KEMA has developed a methodology in order to judge the ash. The KEMA Dust Assessment Methodology (KEMA-DAM[®]) is a methodology to detect, through simple judgement, whether in case of exposure to suspended matter the TLVs and/or health limit values of certain substances in the dust can be exceeded. The KEMA-DAM[®] can detect whether the dust in question is regarded as a nuisance dust. Nuisance dust is such that it does not have any specific toxic characteristics. The method has been concisely summarized in table 1.

TLV stands for Treshold Limited Value; air limit values in the case of occupational exposure are meant here. The air limit value of a substance is the maximally accepted concentration of a gas, vapour, fume or of a dust-formed agent in the air at one's workplace. When determining the air limit value the principle that, even upon repeated exposure and even during long working periods, this concentration should, in general and, obviously, keeping the level of knowledge on this in mind, not harm the employees' and their offspring's health, is used as often as possible. Air limit values do not protect against exposure via other routes (skin, ingestion).

1. Acceptance maximum allowable exposure = $10 \text{ mg}\cdot\text{m}^{-3}$
2. Choice of elements to be considered
3. Determination concentrations of elements
4. Calculation concentrations of elements in the inhalable fraction
5. Determination of element's speciation
6. Determination of the choice of TLVs and conversion into elements
7. Calculation exposure per individual element
8. Calculation quotient exposure and TLV
9. Choice components with similar toxic action on the same organ system
10. Summation of results of step 8 on the basis of step 9 (addition rule)
11. Determination of which components are carcinogenic
12. Determination of concentration of elements from 11 in the total amount of dust
13. Testing result #8, criterion is < 0.5 (statistic inaccuracies)
14. Testing result #10, criterion is < 1 (addition rule)
15. Testing result #12, criterion is $0.1\% \Rightarrow$ determination carcinogenity
16. Determination whether the substances in question can be considered as a nuisance dust

Table 1. Concise description of KEMA-DAM[®]

The KEMA-DAM[®] has been checked up on coal fly ash, for which real toxicity tests were performed. The KEMA-DAM[®] was primarily set up to deal with inorganic substances. The TLVs sometimes refer to elements, but mostly to compounds. In the latter case the chemical form or speciation for the element in question has to be known. In order to get more insight into the chemical form of the elements in coal fly ash, a research program was started. Firstly a literature search was performed. Secondly, thermodynamical calculations by FACT (Facility for the Analysis of Chemical Thermodynamics) were done and thirdly measurements were performed.

Based on measurements the arsenic concentrations were established on calcium arsenate ($\text{Ca}_3(\text{AsO}_4)_2$), the phosphor concentrations on calcium phosphate ($\text{Ca}_3(\text{PO}_4)_2$) and chromium(VI) is present between 4 and 9% of the total chromium. The speciation measurements apply to 100% coal firing and co-combustion in proportions up to 10% by dry mass.

The TLVs refer to the inhalable fraction of the substances. The concentrations in the inhalable fraction of the ash are calculated by the KEMA TRACE MODEL[®]. The KEMA-DAM[®] procedure is applied to the calculated coal fly ash composition at co-combustion in proportions of 10%, 30% and 50% by dry mass of five selected secondary fuels. It

appears that up to 50% co-combustion of paper sludge, sewage sludge, residual wood, chicken manure and RDF of average composition does not lead to individual occupational exposure limits being exceeded at an inhalable coal fly ash dust exposure of 10 mg.m^{-3} . The sum of the average concentrations of the potential carcinogenic trace elements As, Be, Cd, Co, Cr(VI) and Ni in total coal fly ash amounts in all cases to less than 40% of the limit value for carcinogenic components and mixtures of 1000 mg.kg^{-1} . For the cases studied, it is concluded the co-combustion ashes could be assigned as "nuisance dust".

α -QUARTZ

Quartz is of interest because it can have an adverse effect on human health, if inhaled, such as "black lung" or more precisely pneumoconiosis or silicosis. Especially the malignant condition progressive massive fibrosis (PMF) is serious. In relation to the development of silicoses the following limiting conditions have to be fulfilled: The particulate material containing the quartz must be respirable (i.e. sufficiently fine that it is able to penetrate deep into the lungs). The surface of the material is also very important, since it is believed that surface radicals act as the trigger. Surface radicals are found mainly on freshly created surfaces and their formation can be inhibited by weathering/ageing and by the presence of other substances, such as aluminium and some forms of iron. As well as causing silicosis, it is recently known that quartz is a human carcinogen at concentrations above a certain threshold.

Since quartz is found in coal and pulverised fuel ash, it is important to know the concentrations in which it is present and whether its presence can cause PMF. Therefore extensive measurements of α -quartz in ashes of 100% coal firing and of co-combustion were performed with Scanning Electron Microscopy (SEM). From the SEM-results the share of respirable α -quartz in coal ashes was determined with a special developed computer program, called KEMPHASE. With this technique an automatic distinction is made between several size fractions and between free α -quartz particles and α -quartz that is enclosed in fly ash particles.

In Dutch power stations, when coal is fired, approximately 50 per cent of the quartz is vitrified. This vitreous material is one of the main components of pulverised fuel ash. The remainder of the quartz finds its way into the pulverised fuel ash in non-vitreous form. Most of this quartz is found in the non-respirable fraction of the ash; the respirable fraction contains only about 1 per cent of the quartz. In absolute terms, quartz accounted for roughly 0.1 per cent of the respirable fraction of the pulverised fuel ash samples tested. Between 60 and 86 per cent of this quartz was embedded in the ash particles and therefore not available at the surface. Thus, only a very small amount of the quartz is biologically available.

In the workplace underneath the E-filter, the measured stationary respirable atmospheric quartz concentrations under normal stationary conditions average

0.0005 milligrams per cubic metre. That is less than 1 per cent of the TLV for quartz, which is 0,075 milligrams per cubic metre.

All the research undertaken, including epidemiological, *in vivo* and *in vitro* studies, indicates that quartz in pulverised fuel ash does not have the same effect on humans or animals as pure quartz or some quartz containing substances and does not constitute a fibrogenic risk. However, exposure to respirable pulverised fuel ash in concentrations of more than 5 milligrams per cubic metre can result in functional impairment of the lungs and respiratory complaints. At even higher concentrations, there is a risk of chronic bronchitis. However, these effects are what one would expect from any particulate material (nuisance dust); they are not specific to pulverised fuel ash and are certainly not attributable directly to the presence of quartz in the ash.

The absence of the effects normally associated with quartz is attributable to the fact that the quartz in pulverised fuel ash is mainly enclosed within vitreous material. This has been established by electron microscopy of roughly eleven thousand cross-cutted pulverised fuel ash particles. Moreover, it appears that quartz loses its fibrogenic properties when heated to temperatures of more than 1200 °C. All pulverised fuel ash particles undergo heating in excess of this level.

The Dutch Health Council's Expert Committee on Occupational Standards assumes that inhaled quartz particles can cause lung fibrosis, leading to the development of tumours, as a result of prolonged irritation of the lung tissue. Since pulverised fuel ash does not have any of the effects normally associated with quartz, it is highly unlikely that the quartz in pulverised fuel ash is carcinogenic.

From the results of the study it is concluded that pulverised fuel ash of the kind produced at power stations in the Netherlands does not have any of the effects (e.g. silicosis) normally associated with quartz. Hence, the TLVs for quartz are not appropriate for the total-quartz found in pulverised fuel ash.

EUROPEAN WASTE CATALOGUE

In January 2001 the European Commission has adopted a decision in order to come to a harmonised list of hazardous and non hazardous waste, the European Waste Catalogue (EWC). The EWC includes an annex with a list of about 800 wastes. The different types of waste in the list are fully defined by the six-digit code for the waste and the respective two-digit and four-digit chapter headings. Any waste marked with an asterisk (*) is considered as a hazardous waste.

Ashes produced at 100% coal-firing are defined as non-hazardous waste in the EWC, with EWC-codes 10 01 15 for bottom ash and 10 01 17 for coal fly ash. For co-combustion ashes things are not as clear. The classification has to be established based on the chemical composition of the ashes and limit values for several classes of hazard properties. If the ashes contain hazardous components above one of the limit values, the EWC-codes are 10 01 14* and 10 01 16* for bottom ash and fly ash respectively. In that case the ashes are considered as "hazardous waste". If the concentra-

tions are below the limit values, the ashes get the same EWC-code as ashes from 100% coal-firing and are considered as "non-hazardous waste".

An overview of the Risk-statements (R), hazard properties and maximum weight percentages is included in table 2.

The hazard properties can be divided into:

- physical-chemical properties (H1 - H3)
- health aspects (H4 - H11)
- environmental properties (H12 - H14)

For the categories H1, H2, H9, H12, H13 and H14 there are as yet no limit values available, so that at this time these categories cannot be included in the evaluation. More detailed content for these categories is being investigated within the EU.

Category	Limit value
H1 explosive	No limit value as yet
H2 oxidising	No limit value as yet
H3 flammable	55°C
H4 irritant	R36: 20%; R37: 20%; R38: 20%
H5 harmful	25%
H6 toxic	≥ 3%
H6 very toxic	0.1%
H7 carcinogenic category 1 or 2	0.1%
H7 carcinogenic category 3	1%
H8 corrosive	R34: 5%; R35: 1%
H9 infectious	No limit value as yet
H10 toxic for reproduction category 1 or 2	R60 ≥0,5%; R61 ≥0,5%
H10 toxic for reproduction category 3	R62: 5%; R63: 5%
H11 mutagenic category 1 or 2	R46: 0,1%
H11 mutagenic category 3	R40:1%
H12 release of toxic gases	No limit value as yet
H13 release of toxic eluates	No limit value as yet
H14 ecotoxic	No limit value as yet

Table 2. Overview of hazard properties with their limit values in the 14 different categories mentioned in the EWC

By order of the Dutch power stations, KEMA has determined whether the by-products of co-combustion earn the qualification "hazardous" or "non-hazardous" waste. In practise this means that the composition of the ash has to be known in detail: speciation and concentration. The composition of the elements is measured by extensive analyses or calculated with the aid of the KEMA TRACE MODEL®. The speciation of the elements is established in the same way as discussed above. The concentration of each com-

pound as present in the ash was compared to the appropriate limit as mentioned in the EWC and given above.

As an example, table 3 shows the results for bottom ash and pulverised fuel ash (PFA) originating from co-combustion of 20% (mass base dry) of wood in a Dutch coal-fired plant. For all hazard categories, the weight percentages of dangerous components in the evaluated co-combustion ashes fall (far) below the limit values listed in the EWC. This means that the co-combustion ashes evaluated are qualified as non-hazardous. Using a sensitivity analysis, the influence of a great many parameters was checked. It seems that especially the concentrations of Ni, As and Pb in the evaluated by-products have an influence on the results of the EWC assessment.

Co-combustion		20% (m/m) wood		13% (m/m) of biomass pellets and MBM	
hazard category	limit value (%)	bottom ash (%)	PFA (%)	bottom ash (%)	PFA (%)
H4	10	0.008	0.01	0.003	0.01
H4	20	0.06	0.12	0.07	0.12
H5	25	0.35	0.37	0.16	0.21
H6	0.1	0.01	0.01	0.003	0.01
H6	3	0.05	0.07	0.04	0.05
H7	0.1	0.03	0.04	0.01	0.02
H7	1	0.02	0.07	0.02	0.02
H8	1	0.002	0.003	0.002	0.002
H8	5	< 0.001	0.001	< 0.001	0.001
H10	0.5	0.001	0.01	0.001	0.01
H10	5	0.04	0.06	0.03	0.04
H11	1	0.04	0.06	0.03	0.04
H14	no limit	0.37	0.43	0.19	0.28

Table 3. Results of EWC test for co-combustion ashes

LEACHING AND ECO-TOXICITY

For application of by-products as building material in the Netherlands the demands of the Building Materials Decree (BMD) apply. This decree gives limit values for the composition and the leaching behaviour of the by-product. Demands are set for unbound as well as bound application of the by-product. KEMA has performed many leaching test on the ashes produced at coal-firing and co-combustion. From the results it is shown that almost all the bottom ashes produced in the Netherlands fulfil the category-I demands of the Dutch BMD. This means that unbound application of the bottom ashes is allowed without any further precautions. The leaching of the PFA is higher and as a result most of the PFA produced is categorized as a category-II building material. This means that unbound application of PFA as a building material is allowed, but that precautions have to be made to prevent contact of the ashes with ground- and rainwater.

At the moment the Dutch BMD is being revisited, which may result in adapted leaching limit values.

The bio-availability of compounds present in solid waste is dominated by the water-soluble part. Water is the prevailing mechanism for transport in the environment and intake in the organisms. The bio-availability of a material depends therefore on its leaching behaviour rather than on its chemical composition. So a leaching test is the appropriate first step for a bioassay. The choice for the leaching test is very important, e.g. the pH is a main factor. KEMA has chosen for the Dutch availability test (NEN7341), because this test is a batch leaching test at two constant pHs (4 and 7) and simulates the worst case leaching potential of the compound.

The solution (leachate) can then be used for an eco toxicity test. Duck weed (*Lemna minor*) was used first. The grow factor of the plants was measured for a blanc with a solution of a growth medium, and with addition of 25%, 40%, 55% and 70% of the leachates. As a reference under water soils were used of polluted rivers and clean brooks. However, the method is useful, but labour intensive. Therefore a straightforward option was taken by applying a standard bioassay: Microtox[®]. The Microtox[®] gives a relative figure, viz. the Toxicity Index (TI). This TI gives an indication of the acute toxicity. Microtox[®] is presently in wide use at KEMA and other test laboratories around the world for testing of various industrial water effluents. This testing is done in order to determine whether they comply with environmental protection regulations and water quality guidelines. The TI index is measured after during the time. A distinction can be made between the toxicity caused by heavy metals and by organic substances by measuring the TI after two periods.

Microtox[®] was applied to leachates of bottom ash, coal fly ash and soils. The coal fly ash was obtained during the firing of different coals and during co-combustion of secondary fuels or biomass. The Toxicity Index for leachates of coal fly ash was compared to the Toxicity Index for natural and contaminated soil. Soil is designated as contaminated according to the limits in the Dutch Guideline for Soil Protection. The limits in the guideline refer to 'target values' (for clean soil) and 'intervention values' (for seriously contaminated soil) of various elements and substances.

The KEMA study showed the following results. The Toxicity Index for coal fly ash is generally low and comparable to natural soil containing elemental concentrations between 'target value' and 'intervention value' of the Dutch Guideline for Soil Protection. Coal fly ash exposed in open-air landfill for six years showed a remarkable reduction in Toxicity Index. Co-combustion in proportions up to 10% secondary fuel by dry mass did not influence the Toxicity Index in the case of paper sludge and sewage sludge. It appears that the method is useful and not very labour intensive. The same leaching test could also be used for H13 (leaching properties). This can save money and time. The method is considered as a quick screening test. Depending on the results of the test further investigation may be necessary. Expensive and elaborative tests are avoided in most cases by this graduated approach.

TECHNICAL ASPECTS

Results of research made clear that fly ashes generated by high percentages of co-combustion are able to meet the technical requirements of the European standard (EN 450) for utilization as pozzolanic filler in concrete ⁽¹⁰⁾. Performance tests of concrete confirmed the suitability of these fly ashes. The critical factor is the nature of the co-fired fuel and especially on its inorganic matter and ash content. The performance of the fly ash could well be related to its mineralogical and chemical composition. The new ETA requirements (European Technical Agreement) are adapted to ensure a mineralogical and chemical composition that can provide good performance in concrete.

Interesting are the cases in which fuels containing phosphorus and calcium are co-fired. The glass-phase is only slightly enriched in Ca and P. There is no great interaction between the glass-forming matter in the coal and the solids in the secondary fuel. Otherwise there should be more influence of Ca and P on the bulk glass composition. The same is true for paper sludge as in this case there are two glass phases, namely from mineral matter in coal and mineral matter in paper sludge. This implies that there are two ash formation systems, with exchange between them only occurring to a small extent due to collision and in large part via volatile components in the fuels.

CONCLUSIONS

An extensive study conducted by KEMA on by-products of coal-fired plants shows that co-combustion ashes as produced in the Netherlands are considered "non-dangerous substances", according to the EWC (European Waste Catalogue). By showing that a type of waste belongs in the category "non-hazardous", it is possible to achieve considerable savings on the disposal costs.

The applied research indicates that there is no reason to regard co-combustion ash "dust" as harmful. No increased health risk will be caught under circumstances, which generally meet the requirements laid down for nuisance dust in the occupational environment. This means that the standards for nuisance dust can be applied. This at least holds for co-combustion percentages up to 10% and also for the so far tested scenarios at higher co-combustion percentages. Further studies at higher co-combustion percentages are foreseen.

Results of research made clear that fly ashes generated by high percentages of co-combustion are able to meet the technical requirements of the European standard (EN 450) for utilization as pozzolanic filler in concrete. Performance tests of concrete confirmed the suitability of these fly ashes.

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