Toxicity of Ashes Produced During the Combustion and Co-combustion of Coal and Meat and Bone Meal in a Fluidized-Bed Reactor

Rui Barbosa*[(1)], Nuno Lapa*, Helena Lopes, Ibrahim Gulyurtlu**, Benilde Mendes***

[*] UNL, FCT, UBiA

[**] INETI, DEECA

[(1)] UBiA-Faculdade de Ciências e Tecnologia-Universidade Nova de Lisboa, 2829-516 Caparica, Portugal, Tel.: 351-21-2948543, Fax: 351-21-2948543, E-mail: rfb@fct.unl.pt

Executive Summary

The replacement of fossil fuels by renewable fuels can contribute to improve the environmental performance of the power production and to move forward in the sustainability way. The experience has shown that the availability of alternative fuels can be an obstacle for its extensive use for energy production, since biomass is not always available. The use of non-hazardous wastes may be a good alternative to biomass, mainly if they are economically unattractive for recycling or if they present a high cost for land-filling. The co-firing of non-hazardous wastes with coal is, therefore, a subject of great interest for the sustainability of energy production and the reduction of the emissions of fossil greenhouse gases. The use of these wastes for energy is promising if they combine well with other fuels during the conversion process for energy production and have no negative effect on the combustion system, on the ash quality and on the gaseous emissions.

The DEECA/INETI team has performed three combustion assays in a pilot fluidized-bed combustor: a) combustion of coal; b) co-combustion of coal (85%) and meat and bone meal (MBM) (15%); c) combustion of MBM. Each combustion test has produced three types of ashes: 1) bottom ashes; 2) fly ashes collected in the first cyclone; 3) fly ashes collected in the second cyclone. The UBiA/FCT/UNL team has analyzed of the bulk content of the fuels and of the ashes. The quantification of the bulk content was performed according with the USEPA Method 3051A. The ashes were also characterized for chemical and ecotoxicological parameters. The ashes were submitted to the leaching test defined in the European Standard EN 12457-2. The eluates were also characterized for chemical and ecotoxicological characterization. The chemical characterization of the eluates included the following parameters: pH, dissolved organic carbon (DOC), cyanides, chlorides, sulfates, phenolic compounds, total dissolved solids (TDS), fluorides, As, Sb, Se, Hg Cd, Cr, Cr(VI), Cu, Ni, Pb, Mo, Zn, Ba, Ca, Na and K. The ecotoxicological characterization included the following parameters: i) luminescent

inhibition of *V. fischeri*, after 15 and 30 min exposure; ii) growth inhibition of *P. subcapitata*, after 72h exposure; iii) mobility inhibition of *D. magna*, after 48 h of exposure.

According to Council Decision 2003/33/EC, the fly ashes produced in the three combustion tests need stabilization prior to landfilling, except the first cyclone ash produced in the co-combustion test of coal and MBM. This fly ash could be landfilled in a hazardous waste landfill. Based on this same Council Decision, the bottom ashes produced during the combustion tests were classified as non-hazardous residues.

The ecotoxicological characterization of the eluates followed the principles defined in a modified version of the Criterion and Evaluation Methods for Waste Ecotoxicity CEMWE (1998). The chemical and ecotoxicological characterizations have the same significance. Therefore, if at least one chemical or ecotoxicological parameter has a value higher than the limit fixed on the French regulation, the residues shall be classified as ecotoxic. Otherwise, it must be concluded that there is no significant evidence to classify the residue as ecotoxic. The bottom ashes and the first cyclone ashes produced during the combustion of coal and during the co-combustion of coal and MBM have not shown evidences of ecotoxicity. All the ashes produced during the combustion of MBM are ecotoxic, due to the results of the chemical characterization.

INTRODUCTION

The best solution for power production has to be profitable, environmentally sound and sustainable. It is widely accepted that the use of fossil fuels is profitable, but it is not sustainable. The replacement of fossil fuels by renewable sources of energy can contribute to improve the environmental performance of the power production and to move forward in the sustainability way (Morais et al., 2008). The experience has shown that the availability of alternative fuels can be a serious obstacle for its extensive use for energy production. Biomass is not always available to be used as an alternative fuel in fairly small boilers or as a co-fuel in larger mostly coal-based units. The use of non-hazardous wastes may be a good alternative to biomass, mainly if they are economically unattractive for recycling or if they have a high cost for land-filling (Khan et al., 2009). The co-firing of non-hazardous wastes with coal is, therefore, a subject of great interest for the sustainability of energy production and the reduction of the emissions of fossil greenhouse gases (Arias et al., 2008). The use of these wastes for energy is promising if they combine well with other fuels during the conversion process for energy and have no negative effect on the combustion system, on the ash quality and on the gaseous emissions. It is therefore imperative that there will be a satisfactory synergy between coal and wastes and no negative impacts will come to the combustion equipment and to the environment (Lopes et al., 2003).

The utilization of MBM as animal feedstock was forbidden, by the European Union, since the 1st of July of 1994 (Directive 94/38/EC), since it was in the origin of the spreading of Bovine Spongiform Encephalopathy (BSE) which can promote the equivalent human disease (Creutzfeldt-Jakob disease). One possible way for the elimination of the prion that is responsible for this disease, is to submit the MBM to a thermal treatment. The incineration of MBM is efficient to inactivate the prion (Cummins, 2006; National Agricultural Biosecurity Center Consortium, 2004). Therefore, using this residue as a co-fuel can promote the elimination of the prion which is a very positive contribution to the protection of human health and environment.

A European project named COPOWER was developed to assess the possibility to replace coal by MBM. Three combustion tests were performed in the pilot scale combustor of the Departamento de Engenharia Energética e Controlo Ambiental (DEECA) of Instituto Nacional de Energia, Tecnologia e Inovação (INETI): 1) mono-combustion of coal; 2) co-combustion of coal and MBM (85% + 15%); and 3) mono-combustion of MBM. Each one of these combustion tests has promoted three types of ashes: 1) Bottom ashes; 2) Ashes collected in the first cyclone; 3) Ashes collected in the second cyclone.

MATERIALS AND METHODS

FBC, Fuels and Combustion Conditions

The combustion and co-combustion tests were performed in a bubbling fluidized-bed combustor (FBC) (Fig. 2) of INETI/DEECA. The combustor has an internal square section of 0.3 m x 0.3 m and 5 m height. The internal chamber is made of refractory steel (AISI 310). Further details are shown in Lapa et al. (2007). The bottom ashes are collected at the bottom of the combustor and the fly ashes are collected by two containers located at the bottom of each cyclone. The bed material used was cleaned river sand. The fossil fuel used was bituminous coal from the Colombian mine of El Cerrejón with a lower heat value (LHV) of about 24.8 MJ/kg. The MBM was produced in Germany and the LHV was of about 13.10 MJ/kg.

Bulk Characterization of Ashes

The quantification of the bulk content was performed according with the USEPA Method 3051A. This quantification was achieved through Atomic Absorption Spectrometry (AAS) after an acidic digestion with nitric acid (65% v/v) and hydrochloric acid (37% v/v) (1:3), in a Milestone microwave system (model Ethos) using closed vessels. The acidic eluates were filtered through fiber glass filters (Schleicher & Schuell) and the following chemical elements were analyzed: As (EN ISO 11969, 1996), Hg (ISO 5666/1, 1983), Cd, Cu, Ni, Pb and Zn (ISO 8288, 1996), Sb, Se, Mo and Ba (AAS flame quantification – APHA et al., 1996) and Cr (AAS flame quantification/Method A – ISO 9174, 1990). The moisture content of the ashes was determined according to ISO 11465, 1993.

Leaching Tests

All ashes were submitted to the leaching test described in the European leaching standard number EN 12457-2. This European standard describes a leaching test of 24 hours, with one extraction cycle which is performed at a L/S ratio of 10 l/kg. The standard applies to materials of a particle size <4 mm. The extraction cycles were performed under a controlled temperature of 20±1ºC.

Chemical characterization of the eluates

The eluates produced through the European Standard EN 12457-2 were submitted to the following chemical characterization: As (EN ISO 11969, 1996), Hg (ISO 5666/1, 1983), Cd, Cu, Ni, Pb and Zn (ISO 8288, 1996), Sb, Se, Mo, Ca, Na, K and Ba (AAS flame quantification – APHA et al., 1996), Cr (AAS flame quantification/Method A – ISO 9174, 1990), pH, DOC, CN^-, SO_4^{2-}, F^-, TDS (APHA/AWWA/WPCF, 1996), Cl^- (ISO 9297, 1989), Cr (ISO 9174, 1990), Cr(VI) (NF T90-043, 1988), phenol compounds (ISO 6439, 1990).

Ecotoxicological characterization of the eluates

The eluates were also characterized for the following ecotoxic parameters: a) Luminescence inhibition of the bacteria *Vibrio fischeri* (Microtox® system, Azur Environmental) (ISO 11348-3, 2003); b) Mobility inhibition of the crustacean *Daphnia magna* (Daphtoxkit F magnaTM of Microbiotests Company, ISO 6341); and c) growing inhibition of the algae *Pseudokirchenriella subcapitata* (ISO 8692).

RESULTS AND DISCUSSION

Bulk characterization of fuels and ashes

Table 1 shows the moisture content of the ashes produced in the combustion tests.

Table 1 – Moisture content of the bottom, 1st cyclone and 2nd cyclone ashes produced in the combustion tests

Origin of the ash	Combustion test	Moisture Content (wb%)
Bed ashes	MC Coal	0.04
	CC Coal + MBM	0.08
	MC MBM	0.09
1st cyclone ashes	MC Coal	3.3
	CC Coal + MBM	2.6
	MC MBM	0.60
2nd cyclone ashes	MC Coal	2.7
	CC Coal + MBM	3.3
	MC MBM	0.56

MC Coal: Mono-combustion of coal; CC Coal+MBM: Co-combustion of coal and MBM; MC MBM: Mono-combustion of MBM; wb: wet basis

The bed ashes produced in all the combustion tests and the cyclone ashes produced in the mono-combustion of MBM have shown low moisture content (less than 0.1 wb%). The moisture content of the cyclones ashes produced in the mono-combustion of coal and the co-combustion assay was between 2.6 and 3.3 (wb%). The ashes retained in the first and second cyclone produced in the combustion of MBM was of about 0.6 wb%.

Table 2 shows the bulk composition of the fuels for a set of metals.

Table 2 – Bulk composition of the fuels used in the combustion tests

Fuel	Parameter											
	Ba	Sb	Mo	Se	As	Hg	Cd	Pb	Cu	Ni	Zn	Cr
	mg/kg											
Coal	<3.7	<0.07	<22.4	<0.2	<0.7	<0.3	<7.3	<22.8	<9.4	<14.4	36.8	33.5
MBM	452	0.1	117	0.3	<0.7	<0.2	<6.5	<20.4	9.9	<14.0	94.3	<10.2

MBM has shown the highest concentrations of the set of heavy metals analyzed. The major differences in the concentration were observed for the parameters Ba, Mo and Zn. The high concentration of these elements, in MBM, can be explained by the fact that, when consumed by the cattle, they are rapidly transported in blood plasma, mainly to bone (World Health Organization, 2004; World Health Organization, 2003a World Health Organization, 2003b). The concentration of Cr was higher in coal than in MBM. The concentration of As, Hg, Cd, Pb and Ni were below the detection limit in both fuels.

Table 3 shows the bulk composition of the bottom, first cyclone and second cyclone ashes produced in the combustion tests, for a set of metals. According with the data shown in Table 3, the substitution of coal by MBM has promoted a higher concentration of metals in the ashes produced in the co-combustion test, specially, in the mono-combustion test of MBM.

Table 3 – Bulk composition, to a set of metals, of the bottom, 1st cyclone and 2nd cyclone ashes produced in the combustion tests

Parameter	Bed ashes			1st cyclone ashes			2nd cyclone ashes		
	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM
	mg/kg								
Cr	172	162	133	313	308	572	59	292	4,800
Zn	18.3	28.6	128	148	178	233	167	234	1,495
Ni	69.6	30.3	43.5	298	173	202	156	158	3,828
Cu	<8.4	<10.4	<9.3	47.8	49.9	81.1	68.7	73.4	470
Pb	<17.4	<18.9	<17.6	<26.2	<22.5	81.1	44.7	35.6	470
Cd	19.5	22.5	<0.70	<9.1	<18.6	177	19.8	19.7	5,7
Hg	<0.42	<0.45	<0.42	<0.45	<0.61	<0.44	<0.44	<0.75	0.9
As	1.4	0.89	<0.70	3.5	3.4	<0.73	6	6.2	4.8
Ba	<10.4	133	3,110	1,238	1,608	485	1,086	1,428	1,782
Mo	<34.8	<37.7	<35.2	<37.8	73.3	140	90.3	102	508
Sb	<0.10	<0.11	<0.11	<0.11	<0.11	<0.11	<0.11	<0.11	<0.11
Se	<0.70	<0.75	<0.70	32	1.9	<0.73	9.7	12.9	<0.73
Ca	48,056	18,078	129,617	15,880	51,336	238,378	9,185	16,463	210,427
K	4,016	8,070	5,705	14,082	14,442	9,583	14,735	17,890	27,016
Na	3,129	7,731	8,121	6,778	8,585	15,544	6,733	9,300	23,236

MC Coal: Mono-combustion of coal; CC Coal+MBM: Co-combustion of coal and MBM; MC MBM: Mono-combustion of MBM

Concerning the bottom ashes, there were no significant differences between the three bottom ashes analyzed. The exception was for Ba and Zn, since they were retained in higher concentrations in the bottom ashes of the combustion of MBM. This fact is probably associated with the higher concentration of these elements in MBM. The concentrations of Cr, Ni and As were very similar in the bottom ashes. The concentration of Cd was considerable higher in the bottom ashes produced in the combustion of coal and co-combustion test of coal and MBM. This may signify that the concentration of Cd was higher in the coal, although the concentrations of this element in both fuels are below the detection limit. The concentration of the remaining heavy metals was below the detection limit. The introduction of MBM as a co-fuel has increased the levels of Ca, K and Na in the ashes.

The fly ashes retained in the second cyclone, specially those produced in the combustion of MBM, have presented the highest concentration of Cr, Zn, Ni, Cu, Pb and Cd, which can be attributed to the lower particle size of the ashes that usually present enrichment in heavy metals due to volatilization/condensation phenomena, especially in presence of high levels of Cl (Miller et al., 2002; Van de Velden et al., 2008; Barbosa et al., 2009).

The first and second cyclone ashes, produced in the combustion of coal and co-combustion of coal and MBM, have retained As and Se in higher levels than those observed in the ashes of the first and second cyclone ashes produced during the combustion of MBM.

Leaching behavior of ashes

Chemical characterization of the eluates

Table 4 shows the release of chemical species from the ashes under the leaching test conditions. The eluates were produced according to EN standard EN 12457-2. The pH values of the eluates produced by the bottom ashes were of about 11. The eluates produced by first cyclone ashes were slightly less alkaline, around 10, and the pH of the eluates produced by the second cyclone were of about 8. The alkalinity of the bottom ashes was probably associated with the high concentration of oxides in this material. The lower alkalinity of the eluates produced by the cyclone ashes are, probably, associated with the acidic condensates in this material.

The elements Cr(VI), Zn, Ni, Cu, Pb, Cd, Ba and Sb have presented concentrations below the detection limit, except the second cyclone fly ashes produced during the combustion tests of MBM and coal. Cr was detected in the eluates of the ashes retained in the second cyclone of all combustion tests. The concentration of chlorides, fluorides and sulfates was high in the eluates produced by ashes in the co-combustion test and in the combustion of MBM. This fact was probably due to the high concentration of these elements in MBM (Gulyurtlu et al., 2007). It was also observed an higher accumulation of these species in the ashes retained in the second cyclone. This fact may be associated with the accumulation of particles, in this cyclone, with high content of these species and more soluble forms (Barbosa et al., 2009).

Table 4 – Chemical characterization of the eluates produced by the ashes

Parameter	Bed ashes			1st cyclone ashes			2nd cyclone ashes		
	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM
	Sorensen scale								
pH	11.51	9.69	8.00	10.50	9.61	7.44	11.27	10.81	7.34
	mg/kg								
Cr	<0.20	<0.20	1.8	<0.21	<0.21	14.3	<0.27	<0.21	3.3
CrVI	<1.1	<1.1	1.6	<1.1	<1.1	<1.1	<1.1	<1.1	1.7
Zn	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Ni	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	<0.20	17.2
Cu	<0.11	<0.10	<0.19	<0.17	<0.10	<0.11	<0.10	<0.10	<0.10
Pb	<0.49	<0.49	<0.49	<0.51	<0.51	<0.50	<0.50	<0.52	<0.50
Cd	<0.02	<0.02	0.23	<0.02	<0.02	0.03	<0.02	<0.02	<0.02
Hg	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.05	<0.01	<0.01
As	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	0.12	<0.02	0.17
Ba	<0.30	6.0	<0.30	4.5	<0.30	6.5	<0.30	2.7	4.1
Mo	6.9	6.1	<0.99	33.3	18.2	46.1	71.3	79.7	35.5
Sb	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003
Se	0.19	<0.02	0.46	29.7	0.10	0.09	0.82	9.6	0.28
Ca	757	799	113	1,939	288	1,610	2,953	1,234	6,621
K	52	153	2986	650	610	3,852	341	1,033	243
Na	127	244	2,310	781	958	3,782	658	2,302	22,739
Chloride	98.5	<25.0	993	179	206	1,559	103	156	302
Fluoride	95.7	1.5	79	135	108	52.3	110	95.4	641
Sulfate	1,580	2,897	1,863	18,925	18,734	1,786	13,531	10,320	1,338
Phenolic compounds	<0.49	<0.50	<0.50	<0.51	<0.51	<0.50	<0.51	<0.52	<0.50
DOC	54.2	77.4	<0.99	4.2	129	12.8	<1.0	98.9	72.3
TDS	4,652	4,775	11,685	26,401	31,519	23,056	23,955	35,098	120,056
CN^-	<0.13	<0.13	<0.13	0.30	0.47	0.21	<0.13	<0.13	0.25

The combustion tests in which MBM was used as co-fuel and fuel, have shown higher concentration of TDS. The fly ashes retained in the second cyclone have promoted an high release of TDS for the leaching agent. This fact may be associated with higher contents of soluble species in these particles (Barbosa et al., 2009).

Table 5 shows the ecotoxicological classification of ashes, based on the chemical parameters defined in CEMWE.

Table 5 – Ecotoxicological classification, based on the chemical characterization of the eluates, according with the CEMWE criterion

Parameter	Bed ashes			1st cyclone ashes			2nd cyclone ashes		
	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM
Classification	Non-ecotoxic	Non-ecotoxic	Ecotoxic	Non-ecotoxic	Non-ecotoxic	Ecotoxic	Non-ecotoxic	Non-ecotoxic	Ecotoxic
Due tothe concentration of…	n.a.	n.a.	CrVI	n.a.	n.a.	Cr	n.a.	n.a.	Ni and CrVI

n.a.: not applicable

According to CEMWE, the chemical characterization of the eluates has led to the following classification of the ashes: 1) the bottom ashes and the first cyclone ashes produced in the combustion of coal and co-combustion of coal and MBM were classified as non-ecotoxic; 2) the ashes produced in the combustion of MBM were classified as ecotoxic due to Cr(VI) (bed ashes), Cr (first cyclone ashes) and Ni and Cr(VI) (second cyclone ashes).

Ecotoxicological characterization of the eluates

Table 6 shows the ecotoxicological levels of the eluates of ashes produced in the three combustion assays. It is also shown the Toxicity Units (TU) obtained for each eluate (TU=100/ECx, where EC is effective concentration that promotes an effect to fifty or twenty percent of the population exposed to the eluate, depending on the organism used).

Table 6 – Ecotoxicological classification, based on the ecotoxicological characterization of the eluates, according with the CEMWE criterion

Parameter	CEMWE limit	Toxicity units								
		Bed ashes			1st cyclone ashes			2nd cyclone ashes		
		MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM
D. magna	10	1.95	<1.05	<1.05	<1.05	<1.05	1.57	<1.05	<1.05	3.39
V. fischeri	10	4.59	<1.01	2.58	<1.01	<1.01	2.35	<1.01	<1.01	<1.01
P. subcapitata	1000	4.63	1.91	<1.05	<1.05	<1.05	<1.05	28.57	NC	2.53

NC: not characterized due to unsufficient volume sample

The eluates produced by the ashes have presented low ecotoxicological levels and below the CEMWE limit values. According with to the CEMWE, the ecotoxicological characterization has led to the classification of all ashes as non-ecotoxic.

The bed ashes produced by combustion of coal have promoted higher ecotoxicity levels probably due to the high pH levels (Barbosa et al., 2009; Barbosa, 2005; Lopes et al. 1999; Seco et al., 2003). The second cyclone ashes have produced eluates with the highest ecotoxicological levels. The second cyclone ashes produced in the combustion of coal have shown an ecotoxicological level higher than those observed in the remaining eluates.

Overall ecotoxicological classification of the ashes according to CEMWE

The bottom ashes and the first cyclone ashes produced during the combustion of coal and during the co-combustion of coal and MBM have not shown evidences of ecotoxicity. All the ashes produced during the combustion of MBM are ecotoxic, due to the chemical composition.

Classification of ashes according with the Council Decision 2003/33/EC

Table 7 shows the classification of the ashes according the Council Decision 2003/33/EC

Table 7 – Classification of the ashes according with the Council Decision 2003/33/EC

Parameter	Bed ashes			1st cyclone ashes			2nd cyclone ashes		
	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM	MC Coal	CC Coal + MBM	MC MBM
Classification	N-H	N-H	N-H	DnA	H	DnA	DnA	DnA	DnA
Due to the concentration of …	Hg, Mo, Se, F^-, sulfate and TDS	Hg, Mo, sulfate and TDS	Cd, Cr, Hg, Mo, Se, Cl^-, F^-, Sulfate and TDS	Mo and Se	Mo	Mo	Mo	Mo and Se	Mo, F^- and TDS

N-H: Non Hazardous; H: Hazardous; DnA: Deposition not Allowed

According to the Council Decision 2003/33/EC, all the fly ashes require stabilization prior to landfilling, except the first cyclone ash produced in the co-combustion test. This fly ash can be landfilled in a hazardous waste landfill. According to this Council Decision, the bottom ashes produced during the combustion tests were classified as non-hazardous residues.

CONCLUSIONS

The substitution of coal by MBM has led to the production of ashes with higher content of heavy metals. However, the high content of heavy metals in the ashes has not promoted higher leaching rates of these chemical species. It was observed, nevertheless, higher leaching rates of Na, Cl, F and TDS. According to CEMWE and considering both chemical and ecotoxicological characterization, the bottom ashes and the first cyclone ashes produced during the three combustion tests have not presented evidences of ecotoxicity. All the ashes produced during the combustion of MBM were ecotoxic, due to the chemical composition of the eluates. According to the Council Decision 2003/33/EC, all the fly ashes need stabilization prior to landfilling, except the first cyclone ash produced in the co-combustion test that may be lanffilled in a hazardous waste landfill. According to this Council Decision, the bottom ashes produced during the combustion tests were classified as non-hazardous residues.

Despite of the chemical characterization has not conducted, according to the Council Decision 2003/33/EC, to different classifications of the ashes retained in the same place of the combustor, it is advisable to monitor the leaching behavior of the ashes produced during the co-combustion trials were MBM is used as fuel. The results also show that finer particles are more ecotoxic, hence a special attention should be given to the finer particles emitted.

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