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ScienceDirect
Journal of Hazardous Materials 153 (2008) 544–550
Journal of Hazardous Materials
www.elsevier.com/locate/jhazmat

Implications of moisture content determination in the environmental characterisation of FGD gypsum for its disposal in landfills

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Received 4 July 2007; received in revised form 28 August 2007; accepted 29 August 2007
Available online 1 September 2007

Abstract

The leachable contents of elements of environmental concern considered in the Council Decision 2003/33/EC on waste disposal were determined in flue gas desulphurisation (FGD) gypsum. To this end, leaching tests were performed following the standard EN-12457-4 which specifies the determination of the dry mass of the material at 105 °C and the use of a liquid to solid (L/S) ratio of 10 l kg^{-1} dry matter. Additionally, leaching tests were also carried out taking into account the dry mass of the material at 60 °C and using different L/S ratios (2, 5, 8, 10, 15 and 20 l kg^{-1} dry matter). It was found that the dry mass determination at 105 °C turns out to be inappropriate for FGD gypsum since at this temperature gypsum transforms into bassanite, and so, in addition to moisture content, crystalline water is removed. As a consequence the moisture content is overvalued (about 16%), what makes consider a lower L/S ratio than that specified by the standard EN-12457-4. As a result the leachable contents in FGD gypsum are, in general, overestimated, what could lead to more strict environmental requirements for FGD gypsum when considering its disposal in landfills, specially concerning those elements (e.g., F) risking the characterisation of FGD gypsum as a waste acceptable at landfills for non-hazardous wastes.

Keywords: FGD gypsum; Landfilling; EN-12457-4; Leaching; Moisture content

1. Introduction

During the last decades most coal-combustion power plants in order to comply with the legislation regarding the SO_2 emissions to the atmosphere [1–3] have been equipped with flue gas desulphurisation (FGD) installations. Although several FGD processes are available to this end, the wet limestone FGD process is that most widely applied, taking up to about 80% of the market. In this process SO_2 is removed from the flue gas by absorption into limestone slurry, then oxidised to produce sulphate, extracted from the absorber as gypsum slurry, and finally dewatered, producing the so-called FGD gypsum. The process taking place can be summarised in the following reaction:

$$CaCO_3(s) + SO_2(g) + (1/2)O_2(g) + 2H_2O(l) \rightarrow CaSO_4{\cdot}2H_2O(s) + CO_2(g) \quad (1)$$

It is important to note that the limestone slurry used in the desulphurisation process acts as a scavenging system also for volatile elements other than S, such as F, Cl, As, Se and Hg [4–9], present as well in the flue gas emissions, or condensed on the particulate matter (PM) escaping the electrostatic precipitators (ESPs), likewise for other trace elements of environmental concern present in this PM (Cd, Cr, Cu, Ni, Pb, Zn, Ba, Sb and Mo).

FGD gypsum produced from the indicated process can find application in the wallboard manufacture or be used in the cement industry. Nevertheless, it is also employed in a great proportion as a landfill material in mine reclamation or sent to landfills for its disposal. In such cases the leaching of elements of environmental concern must be controlled according to the Council Decision 2003/33/EC [10], presently in force. The Council Decision establishes the criteria and procedures for the acceptance of waste at landfills, their application being compulsory (by 16 July 2005) by European Union (EU) Member States. The procedure to determine the acceptability of waste at landfills consists of the basic characterisation, com-

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doi:10.1016/j.jhazmat.2007.08.088

pliance testing and on-site verification. Basic characterisation constitutes a full characterisation of the waste (type and origin, composition, consistency, leachability and, where necessary and available, other characteristic properties). A waste is deemed acceptable for a landfill class on the basis of the basic characterisation. Subsequently, the compliance testing and the on-site verification shall be performed with the aim of verifying the classification derived from the basic characterisation. In accordance with the Council Decision 2003/33/EC [10], and as it is specified in the section 3 of its annex, listing the methods to be used for the sampling and testing of waste, the EN standards must be used for the determination of the basic waste properties. Thus, the standards EN-12457/1–4 [11–14] must be employed as leaching tests for granular waste materials and sludges. These standards differ in the liquid to solid (L/S) ratio to employ, the particle size of the waste and the number of stages to perform in the batch leaching test. Among them, the standard EN-12457-4 [14] is generally that followed due to its simplicity (the corresponding leaching test is performed in only one stage), and because it requires lesser material size reduction. Physical modification of wastes should be avoided since as a result their properties could be altered, including their leaching behaviour.

According to the specifications of the standard EN-12457-4, the leaching of waste materials must be performed using: (a) representative samples containing 0.090 ± 0.005 kg of dry mass with a grain size of at least 95% (mass) less than 10 mm, (b) samples without being subjected to drying processes, unless their moisture content does not allow their crushing to reach the required sample grain size (in such cases drying temperature must not exceed 40 °C), (c) deionised water or water of equivalent purity ($5 < pH < 7.5$, conductivity $<0.5\,mS\,m^{-1}$) as leachant, employing a L/S ratio of $10\,l\,kg^{-1}$ dry matter ±2% and (d) an agitation period of 24 ± 0.5 h, using a rotary system at about 5–10 rpm. The standard EN-12457-4 also establishes that, even though the leaching test must be performed using samples without being dried, the leachable contents in the waste material must be calculated on its dry mass, specifying the determination of the dry mass of the material at 105 ± 5 °C according to ISO 11465.

The determination of moisture content at the temperature specified by the standard EN-12457-4 could turn out to be inappropriate for certain materials, namely those risking to lose crystalline water at temperatures about or below 105 °C, as could be the case of FGD gypsum. It has been reported that gypsum transforms into bassanite ($CaSO_4{\cdot}0.5H_2O$) by losing 1.5 molecules of crystalline water per molecule of gypsum when heating at moderate temperature; although there is some controversy, the onset of the dehydration temperature of gypsum to bassanite is generally accepted to be about 100 °C [15].

The main objective of the present work is to assess the implications of moisture content determination as specified by the standard EN-12457-4 in the establishment of FGD gypsum leachable contents, and so in the environmental characterisation of this by-product for its disposal in landfills.

2. Materials and methods

2.1. FGD gypsum

Samples of FGD gypsum were collected from a Spanish coal-combustion power plant equipped with a wet limestone FGD installation. Sample collection was carried out just after the dewatering process of FGD gypsum slurry performed at the power plant before sending it for disposal. Sampling was performed in three consecutive days, obtaining three different samples that were mixed and homogenized to give a single sample.

2.2. Mineralogical characterisation

The mineralogical composition of FGD gypsum samples dried at both 60 °C and 105 °C was determined by X-ray diffraction (XRD) using the reference intensity method (RIM) described by Chung [16,17]. XRD analysis was performed on a Siemens D 501 diffractometer using Cu Kα radiation. Solids were scanned as unoriented powder samples from 4° to 60° 2θ with a 0.04° 2θ step interval and a 1 s per step counting time.

2.3. Moisture content analysis

Moisture content of FGD gypsum samples was determined both at 105 °C (according to ISO 11465, as the standard EN-12457-4 establishes) and at 60 °C. With this aim FGD gypsum samples were dried in an oven at 105 ± 5 °C and at 60 ± 5 °C to constant weight. Moisture content determinations in this way were carried out by triplicate and only mean values are presented.

2.4. Leaching tests

2.4.1. Environmental characterisation of FGD gypsum

Leaching tests were performed following the standard EN-12457-4 as the Council Decision 2003/33/EC establishes. Thus, FGD gypsum samples were subjected to an agitation period of 24 h with deionised water on a vertical rotary shaker (10 rpm), using a L/S ratio of $10\,l\,kg^{-1}$ dry matter. FGD gypsum samples underwent this process without being dried and without being crushed since their particle aggregate size was already below 10 mm, as the mentioned standard requires. Such leaching tests were carried out by triplicate. Leachates were analysed for all those elements considered in the Council Decision 2003/33/EC [10], thus As, Ba, Cd, Cr, Cu, Mo, Ni, Pb, Sb, Se and Zn were analysed by inductively coupled plasma-mass spectrometry (ICP-MS) using a Thermo Electron Corporation ICP-MS X Series II apparatus, Cl and sulphate by ion chromatography (IC) using a KONTRON-WATERS unit, Hg using a mercury analyser (model AMA-254) equipped for thermal decomposition, amalgam formation and atomic absorption measurement, and F by fluoride selective electrode using a Thermo Orion ISE-meter (model 710). The leachable content of constituents of environmental concern in FGD gypsum was calculated on the dry mass of the material dried at 105 °C as specified by the standard EN-

12457-4. Also leaching tests were performed taking into account the dry mass of the material dried at 60 °C, following, except for this aspect, the procedure and the analytical techniques mentioned before. Leaching tests in this way were also performed by triplicate.

2.4.2. Effect of L/S ratio in the leaching behaviour of FGD gypsum

The actual L/S ratio employed when leaching test are carried out considering the dry mass of FGD gypsum dried at 105 °C is 8 l kg^{-1} dry matter instead of 10 l kg^{-1} dry matter. Therefore, in order to assess the influence of the L/S ratio in the leaching behaviour of FGD gypsum, leaching tests following the procedure described before were performed at different L/S ratios, namely 2, 5, 8, 10, 15 and 20 l kg^{-1} dry matter. The dry mass of the material derived at 60 °C was that employed for the accomplishment of these leaching tests (also by triplicate). Leachates were analysed for all those elements considered in the Council Decision 2003/33/EC using the analytical techniques aforementioned.

3. Results and discussion

3.1. Mineralogical characterisation

Fig. 1 shows the XRD diagrams of FGD gypsum samples dried at 60 °C and at 105 °C. The FGD gypsum sample dried at 60 °C presents gypsum as main mineral phase with some impurities of calcite (<5%) and montmorillonite (traces). The occurrence of calcite is related to its incomplete sulphation during the desulphurisation process according to the reaction indicated in Eq. (1), and that of montmorillonite to its occurrence in trace amounts in the limestone used as scavenging material in the desulphurisation process described before. The XRD pattern of the FGD gypsum sample dried at 105 °C only displays the diffraction peaks corresponding to bassanite, and those of the calcite impurity (<5%). No diffraction peaks related to gypsum are observed, proving a complete phase transformation of gypsum to bassanite, what implies the loss of 1.5 molecules of crystalline water per molecule of gypsum. This transition involves a structural change; it has been established that the gypsum–bassanite transition is associated with a rearrangement of sulphate ions in the crystal structure because of the fact that they are strongly coupled with the water molecules [15]. Thus, the crystalline structure of gypsum is monoclinic-prismatic, while that of bassanite is orthorhombic. Therefore, when FGD gypsum is dried at 105 °C, in addition to moisture content removal, gypsum crystalline water is partially lost, being completely transformed into a new mineral phase. The mass loss corresponding to crystalline water removal theoretically accounts for 15.7% of the initial gypsum mass, this mass loss cannot be in any case attributed to moisture content as the standard EN-12457-4 regarding this aspect does.

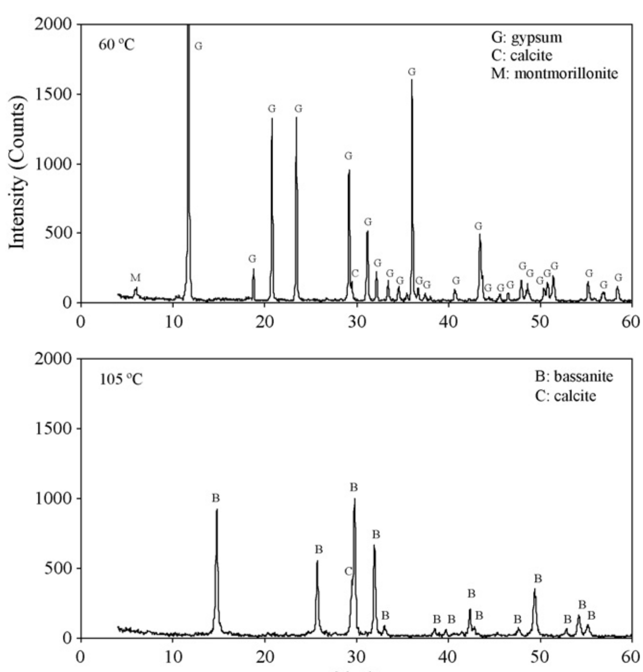

Fig. 1. XRD patterns of FGD gypsum dried at 60 °C and at 105 °C.

3.2. Moisture content analysis

The moisture content ratio (MC) (%) of FGD gypsum samples dried at 60 °C and at 105 °C are 13.8 and 29.7%, respectively, and the dry matter content ratio (DR) (%) of FGD gypsum samples dried at 60 °C and at 105 °C are 86.2 and 70.3%, respectively. The MC was calculated as follows:

$$\mathrm{MC}\,(\%) = \frac{100 \times (M_\mathrm{W} - M_\mathrm{D})}{M_\mathrm{W}} \quad (2)$$

where M_W is the mass of undried test portion and M_D is the mass of the dried (either at 60 °C or at 105 °C) test portion.

The DR was calculated as follows:

$$\mathrm{DR}\,(\%) = \frac{100 \times M_\mathrm{D}}{M_\mathrm{W}} \quad (3)$$

These results show that the determined MC and DR values are different depending on the drying temperature at which FGD gypsum samples were subjected. Much greater MC and, therefore, much lower DR values were derived when FGD gypsum samples were dried at 105 °C. The difference between the MC and DR values obtained at both temperatures are found to be 15.9%, which is in correspondence with the crystalline water loss produced when FGD gypsum samples were dried at 105 °C. As was mentioned before, the crystalline water loss corresponding to the transformation of gypsum into bassanite theoretically accounts for 15.7% of the initial gypsum mass. Hence, all the mass loss produced when FGD gypsum is dried from 60 °C to 105 °C is attributable to the removal of the crystalline water necessary for gypsum to be transformed into bassanite, since no significant differences were found between both values. Therefore, it could be concluded that, in order to properly determine the MC and DR of FGD gypsum, the drying process should be performed at temperatures below which the transition gypsum–bassanite starts; a temperature of 60 °C seems suitable to totally remove moisture content from FGD gypsum while keeping crystalline water.

3.3. Leaching tests

3.3.1. Environmental characterisation of FGD gypsum

Table 1 shows the leachable contents in FGD gypsum for all the elements of environmental concern considered in the Council Decision 2003/33/EC derived from both the leaching tests performed taking into account the dry mass of the material dried at 105 °C, as the standard EN-12457-4 establishes, and the dry mass of the material dried at 60 °C (the reported contents represent mean values). All the leachable contents determined following strictly the standard EN-12457-4 remained below the required limit values for wastes to be accepted at landfills for non-hazardous wastes (also indicated in Table 1), therefore, regarding the leachable content of inorganic constituents, FGD gypsum could be disposed of in such landfills, following the additional stated dispositions (e.g., non-hazardous gypsum-based materials should be disposed of only in landfills for non-hazardous wastes in cells where no biodegradable waste is accepted). Anyway, the leachable contents of some of them, namely fluoride and sulphate, were very close to the mentioned limit values, what could risk the classification of this waste as acceptable at landfills for non-hazardous wastes. Measures to control their leaching from FGD gypsum when disposed of should be undertaken, especially in the case of fluoride, given that the Council Decision 2003/33/EC allows the use of the TDS (total dissolved solids) value (60,000 mg kg^{-1}) alternatively to those for sulphate and chloride, what could allow higher sulphate leaching concentrations. The minimisation of fluoride leaching when FGD gypsum is disposed of has already been recently subject of study [18], pointing out the concern brought about by this new environmental issue.

Nevertheless, when the leachable contents in FGD gypsum were derived considering the dry mass of the material dried at 60 °C such contents were found to be generally lower. This finding becomes particularly relevant for those elements risking the characterisation of FGD gypsum as acceptable at landfills for non-hazardous wastes when their leachable content is derived considering the dry mass of the material dried at 105 °C, as the standard EN-12457-4 establishes. In this aspect, and as is shown in the present study, fluoride is an element of interest. In addition, other toxic elements should be considered, especially selenium. This element has been reported to be removed in a high proportion from flue gas in the absorber by the formation of FGD gypsum [19,20], and its leachable content could be considered to be in the intermediate-upper range when comparing it with the limit value allowed to characterise a waste as acceptable at landfills for non-hazardous wastes.

Several aspects must be born in mind to explain the difference between the leachable contents in FGD gypsum derived at both temperatures (60 °C and 105 °C), all of them consequence of the wrong MC and DR determination at 105 °C. Firstly the L/S ratio employed when leaching tests are carried out considering the dry mass of the material dried at 105 °C is not 10 l kg^{-1} dry matter, but actually 8 l kg^{-1} dry matter, as calculated from the following equation:

$$L = [\left(\frac{L}{S}\right) M_D - (M_W - M_D)] \tag{4}$$

where the actual M_D, that derived at 60 °C, was considered; this aspect could influence the leaching behaviour of FGD gypsum as will be shown in the latter section. Secondly, when calculating the leachable contents of constituents in FGD gypsum from their concentrations in the leachates, a factor of 10 l kg^{-1} is being considered when it is actually 8 l kg^{-1}.

3.3.2. Effect of L/S ratio in the leaching behaviour of FGD gypsum

Fig. 2 shows the concentrations (μg l^{-1} or mg l^{-1}) of constituents of environmental concern present in the leachates arising from the leaching processes of FGD gypsum performed at different L/S ratios taking into account the actual MC and DR

Table 1
Leachable contents in FGD gypsum at different drying temperatures (105 °C and 60 °C) and limit values for wastes to be accepted at landfills for non-hazardous wastes

Constituent	FGD gypsum (105 °C) (mg leachable kg^{-1} dry matter)	FGD gypsum (60 °C) (mg leachable kg^{-1} dry matter)	Limit value (mg leachable kg^{-1} dry matter)
As	0.060	0.055	2
Ba	0.272	0.262	100
Cd	<0.001	<0.001	1
Cr	<0.001	<0.001	10
Cu	0.015	0.015	50
Hg	<0.001	<0.001	0.2
Mo	0.042	0.035	10
Ni	0.078	0.073	10
Pb	<0.001	<0.001	10
Sb	<0.001	<0.001	0.7
Se	0.439	0.412	0.5
Zn	0.030	0.029	50
Chloride	341	273	15,000
Fluoride	144	134	150
Sulphate	19,510	18,570	20,000

values (those derived when the FGD gypsum drying process was performed at 60 °C). The corresponding leachable contents (mg kg^{-1}) in FGD gypsum derived from the concentrations present in such leachates are also shown in Fig. 2.

Leaching from FGD gypsum mainly depends on the solubility of their chemical components. Ideally, solubility-controlled conditions result in concentrations in the leachates independent of the L/S ratio [20], but dealing with complex systems,

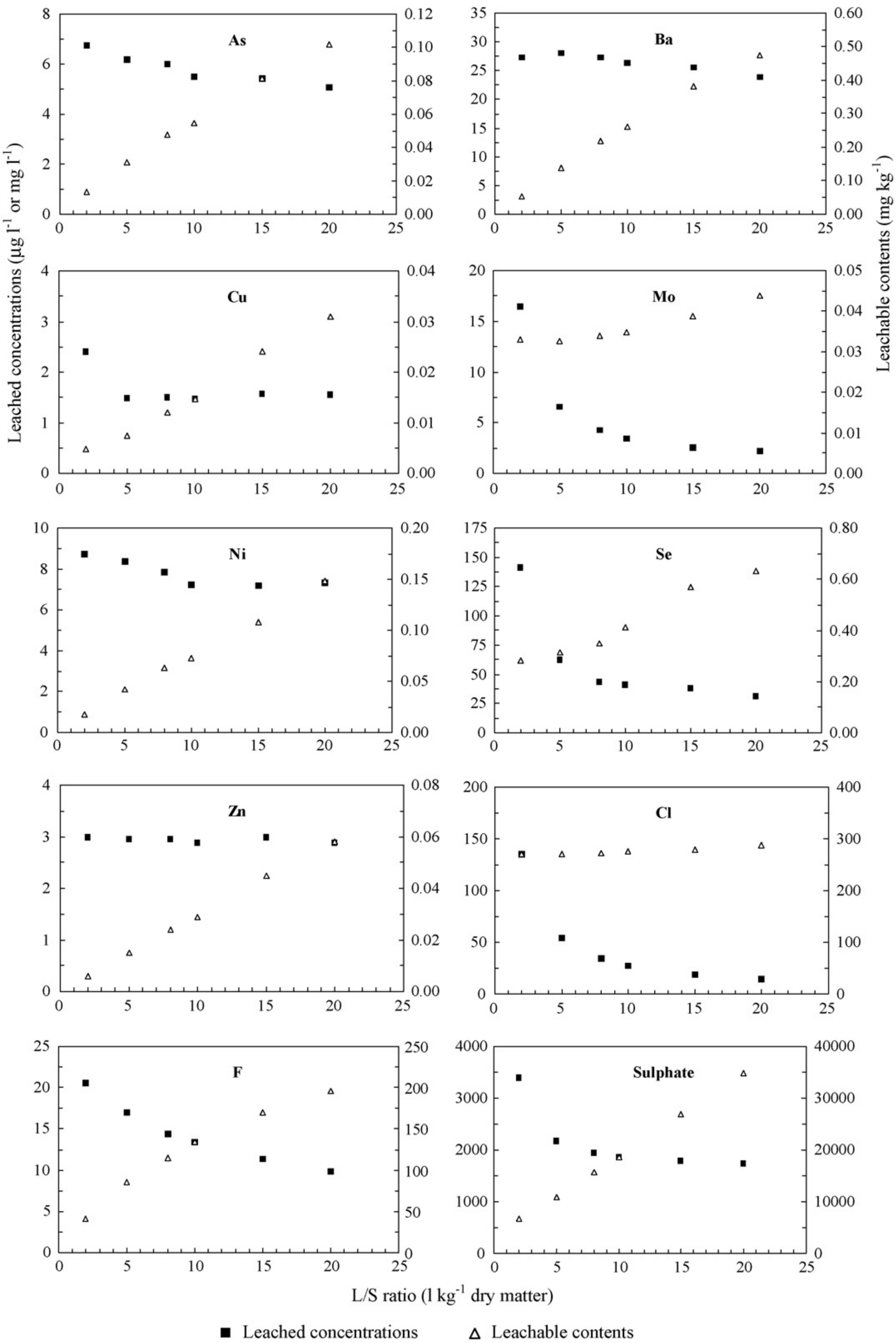

Fig. 2. Leached concentrations from FGD gypsum (μg l^{-1} for As, Ba, Cu, Mo, Ni, Se and Zn and mg l^{-1} for Cl, F and sulphate) and leachable contents in FGD gypsum (mg kg^{-1}) at different L/S ratios considering a drying temperature of 60 °C (leached concentrations of Cd, Cr, Hg, Pb and Sb were below the detection limit (0.1 μg l^{-1})).

as is the case of the by-product subject of study, and with concentrated systems, as those here, the actual behaviour differs from that expected for ideal conditions. Thus, in general, the element concentrations in the FGD gypsum leachates are raised with the increasing solid content (or with the decreasing L/S ratio). Therefore, as was suggested in the previous section, the L/S ratio influences the leaching behaviour of FGD gypsum. Nevertheless, in spite of the fact that the concentrations leached ($\mu g\,l^{-1}$ or $mg\,l^{-1}$) from FGD gypsum decrease with the L/S ratio increase, the leachable contents ($mg\,kg^{-1}$) in FGD gypsum become higher with the L/S ratio increase, as a consequence of the dilution of waste material with the leaching solution. Hence, lower leachable contents are found at a L/S ratio of $8\,l\,kg^{-1}$ dry matter than at L/S ratio of $10\,l\,kg^{-1}$ dry matter. However, when leaching is performed following strictly the standard EN-12457-4 higher leachable contents are derived, even if the actual L/S ratio employed in this leaching test is $8\,l\,kg^{-1}$ dry matter. This happens because when following the standard EN-12457-4 the leachable contents of constituents in FGD gypsum are calculated from their concentrations in the eluates considering a factor of $10\,l\,kg^{-1}$, being actually $8\,l\,kg^{-1}$. Therefore, the wrong FGD gypsum MC and DR determination at 105 °C, as established by the standard EN-12457-4, makes overestimate leachable contents in FGD gypsum in relation to those determined taking into account the actual moisture content (that derived at 60 °C). Concerning the elements risking or susceptible of risking the characterisation of FGD gypsum as a waste acceptable at landfills for non-hazardous wastes, namely fluoride and selenium, an overestimation in their leachable contents about 7% is found. This could lead to more strict environmental requirements for FGD gypsum for its disposal in landfills when compared with other by-products not losing crystalline water during the moisture content determination at 105 °C.

4. Conclusions

The moisture content determination at 105 °C, as the standard EN-12457-4 establishes, turned out to be inappropriate for FGD gypsum, since at this temperature gypsum transforms into bassanite, and so, in addition to moisture content, crystalline water is removed. This wrong determination leads to overvalue the moisture content of FGD gypsum (about 16%), in correspondence with the crystalline water loss produced when gypsum is dried at 105 °C. As a consequence, in the leaching test performed following strictly the standard EN-12457-4 two mistakes are included, these are, the L/S ratio employed in the leaching test is not $10\,l\,kg^{-1}$ dry matter, but actually $8\,l\,kg^{-1}$ dry matter, and when calculating the leachable contents of constituents in the FGD gypsum from their concentrations in the leachates, a factor of $10\,l\,kg^{-1}$ is being considered when it is actually $8\,l\,kg^{-1}$. The conjugation of these two factors makes overestimate in general the leachable contents in FGD gypsum. Particularly the leachable contents of elements risking or susceptible of risking the characterisation of FGD gypsum as a waste acceptable at landfills for non-hazardous wastes, namely fluoride and selenium, are overestimated about 7%. This could lead to more strict environmental requirements for FGD gypsum for its disposal in landfills when compared with other materials not losing crystalline water at 105 °C.

Acknowledgements

The authors thank to ENDESA for supplying the FGD gypsum samples employed in this study. The authors also acknowledge M. Cabañas, S. Martínez and J. Elvira for their technical assistance during the development of this work.

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