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Environmental, physical and structural characterisation of geopolymer matrixes synthesised from coal (co-)combustion fly ashes

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Abstract

The synthesis of geopolymer matrixes from coal (co-)combustion fly ashes as the sole source of silica and alumina has been studied in order to assess both their capacity to immobilise the potentially toxic elements contained in these coal (co-)combustion by-products and their suitability to be used as cement replacements. The geopolymerisation process has been performed using (5, 8 and 12 M) NaOH solutions as activation media and different curing time (6–48 h) and temperature (40–80 °C) conditions. Synthesised geopolymers have been characterised with regard to their leaching behaviour, following the DIN 38414-S4 [DIN 38414-S4, Determination of leachability by water (S4), group S: sludge and sediments. German standard methods for the examination of water, waste water and sludge. Institut für Normung, Berlin, 1984] and NEN 7375 [NEN 7375, Leaching characteristics of moulded or monolithic building and waste materials. Determination of leaching of inorganic components with the diffusion test. Netherlands Normalisation Institute, Delft, 2004] procedures, and to their structural stability by means of compressive strength measurements. In addition, geopolymer mineralogy, morphology and structure have been studied by X-ray diffraction (XRD), scanning electron microscopy (SEM) and Fourier transform infrared spectroscopy (FTIR), respectively. It was found that synthesised geopolymer matrixes were only effective in the chemical immobilisation of a number of elements of environmental concern contained in fly ashes, reducing (especially for Ba), or maintaining their leachable contents after the geopolymerisation process, but not for those elements present as oxyanions. Physical entrapment does not seem either to contribute in an important way, in general, to the immobilisation of oxyanions. The structural stability of synthesised geopolymers was mainly dependent on the glass content of fly ashes, attaining at the optimal activation conditions (12 M NaOH, 48 h, 80 °C) compressive strength values about 60 MPa when the fly ash glass content was higher than 90%.
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1. Introduction

Worldwide millions of tons of fly ash are generated each year by coal-fired power plants satisfying the large demand for industrial and domestic energy. The management of this by-product is always matter of concern. Only about 20–30% of the generated fly ash is used, mainly as additive in cement and concrete and as filling material, the rest is disposed of, landfilling is currently the processing technique used. Therefore, strategies to deal with this waste safely are required. Especial attention should be paid not only to prevent environmental pollution, but to treat fly ash as a valuable resource material. In this regard, the synthesis of geopolymers seems an interesting approach.

Geopolymers, also called polysialates, are amorphous to semi-crystalline three-dimensional aluminosilicate polymers considered as the analogues of certain zeolites. Thus, geopolymers consist of a polymeric silicon–oxygen–aluminium framework with alternating silicon and aluminium tetrahedra joined together in three directions by sharing all the oxygen atoms. The fact that aluminium is four coordinated with respect to oxygen creates a negative charge imbalance, and, therefore, the presence of cations is essential to maintain electric neutrality in the matrix [1]. The general formula to describe the chemical composition of these mineral polymers is $M_n[-(SiO_2)_z-AlO_2]_n \cdot wH_2O$, where z is 1, 2 or 3, M is an alkali cation (such as potassium or

sodium), and n is the degree of polymerisation. Accordingly, in order to better describe the geopolymeric structures, a terminology has been proposed: poly(sialate) (–Si–O–Al–O–), poly(sialate-siloxo) (–Si–O–Al–O–Si–O–), and poly(sialate-disiloxo) (–Si–O–Al–O–Si–O–Si–O–) [2]. The main properties of geopolymers are: quick compressive strength development, low permeability, resistance to acid attack, good resistance to freeze-thaw cycles, and tendency to drastically decrease the mobility of most heavy metal ions contained within the geopolymeric structure [3]. Such properties make them interesting structural products, such as concrete replacements in various environments, and immobilisation systems for heavy metal containment.

Geopolymerisation occurs in alkaline solutions with aluminosilicate oxides and silicates (either solid or liquid) as reactants. Geopolymerisation takes place through a mechanism involving the dissolution of aluminium and silicon species from the surfaces of source materials as well as the surface hydration of undissolved particles. Afterwards, the polymerisation of active surface groups and soluble species takes place to form a gel, generating subsequently a hardened geopolymer structure. In most cases, only a small amount of the silica and alumina present in particles needs to dissolve and take part in the reaction for the whole mixture to solidify.

Many materials containing large amounts of silica and alumina that partially dissolve in alkaline solutions have been used as reagents for geopolymerisation reactions. These include natural minerals (kaolinite, feldspar, albite, stilbite) [4–6], treated minerals (metakaolinite) [7] and waste materials (building waste, blast furnace slag, fly ash) [8–10].

Although geopolymerisation is not a new concept, the application of this technology to waste materials is relatively recent. Thus, only in the last decade it has been applied to fly ashes, remaining still quite to know for geopolymerisation to became an actual environmental and commercial approach to treat them. A typical fly ash-based geopolymer mix consists of approximately 60% mass dry fly ash, and approximately 15% mass dry additional Al–Si source (usually kaolinite or metakaolinite), the rest of the mix is the alkali or alkali silicate mixing solution [8,11–16]. However, this percentage can vary greatly depending on the characteristics of fly ash (bulk and soluble Si and Al contents). Thus, percentages of fly ash as low as 10% have been employed in some geopolymer reactions [17,18], in contrast to those geosyntheses where fly ash was the only solid source material [10]. It is evident that previous research on geopolymerisation concerning fly ash as the sole source of silica and alumina is quite limited, remaining still a lot unknown about the physical and chemical properties of derived products as well as about the factors governing the formation of these geopolymers.

The main objectives of this work are: (a) to study the suitability of different class F fly ashes to be used as the sole source material in geopolymer synthesis, both from a physical point of view (structural stability), aimed to explore their use as concrete replacements, and from a chemical standpoint (leaching behaviour), aimed to assess their stabilization and (b) to better understand the processes involved in this synthesis.

Table 1
Chemical composition (wt%) of fly ash samples subject of study

	FA-1	FA-2	FA-3	FA-4
SiO_2	54.1	43.4	58.1	51.3
Al_2O_3	23.3	25.9	22.7	25.5
CaO	3.5	5.3	3.5	2.9
Na_2O	0.9	0.1	0.6	0.7
K_2O	3.2	1.2	1.6	3.6
MgO	2.0	1.2	1.8	1.8
MnO	0.1	0.04	0.1	0.1
TiO_2	1.0	0.9	1.1	1.0
SO_3	0.4	0.9	0.2	0.5
P_2O_5	0.8	0.5	0.5	0.5
Fe_2O_3	8.5	19	6.1	6.9
LOI	2.0	1.5	3.5	5.0

2. Materials and methods

2.1. Fly ashes

Four fly ashes from Spanish power plants with the chemical composition given in Table 1 and with the particle size distribution indicated in Table 2 were used in the synthesis of geopolymer matrixes. Three of the fly ashes were generated from coal combustion processes (FA-1, FA-2 and FA-3 from Unión FENOSA-Narcea, ENDESA-Teruel and ENDESA-Los Barrios power plants, respectively), and the remaining one from a co-combustion process where petroleum coke is employed as co-combustible (FA-4 from ENDESA-Compostilla power plant). The coal:petroleum coke ratio employed in this co-combustion process is 96:4. The addition of petroleum coke at low proportion as a secondary fuel (as is the case of the present study) hardly modify the bulk composition or the overall leachability of the resulting fly ash.

The four fly ashes were characterised with regard to their mineralogy, structure, morphology and leaching behaviour. The mineralogy was determined by X-ray diffraction (XRD) on a Siemens D 501 diffractometer using Cu Kα radiation and scanning the unoriented powder solids from 4° to 60° 2θ with a 0.04° 2θ step interval and a 1 s per step counting time. Semi-quantitative analyses were carried out using the Reference

Table 2
Particle size distribution of fly ash samples subject of study

	Particle size distribution	Value (μm)
FA-1	90th percentile	103
	50th percentile	20
	10th percentile	5.6
FA-2	90th percentile	131
	50th percentile	21
	10th percentile	4.8
FA-3	90th percentile	147
	50th percentile	37
	10th percentile	7.5
FA-4	90th percentile	78
	50th percentile	12
	10th percentile	2.9

Intensity Method (RIM) described by Chung [19,20], where CaF_2 was employed as internal standard. The glass content was derived from such analyses after determination of the total content of crystalline phases. Structural analyses were performed by Fourier transform infrared spectroscopy (FTIR). The infrared absorption spectra of samples in the form of KBr tablets were recorded for 350–4000 cm^{-1} using a Perkin-Elmer spectrum one unit, being all spectra obtained with a sensitivity of 4 cm^{-1} and 30 scans per spectrum. Morphological characterisation was carried out using a JEOL JSM 840 Scanning Electron Microscope (SEM). Samples were studied on SEM stubs with a sticky carbon cover and then coated with carbon. The leaching behaviour of fly ash samples was studied following the DIN 38414-S4 [21] procedure which specifies the use of deionised water as leachant at a liquid to solid ratio of 10 l/kg dry matter. The leachates were analysed for pH, and for elements of environmental concern by inductively coupled plasma-mass spectrometry (ICP-MS) using a Thermo Electron Corporation ICP-MS X Series II apparatus.

2.2. Geopolymerisation

Analytical grade NaOH (Merck) was used as reagent in the geopolymerisation process. Fly ashes were activated by means of 5, 8 and 12 M NaOH solutions using a liquid/solid ratio of 0.3 for all the fly ash samples subject of study, except for the FA-3 fly ash sample for which the liquid/solid ratio was increased up to 0.4 in order to obtain a workable paste (FA-3 fly ash requires higher liquid amount to achieve a thixotropic behaviour which is desirable for geopolymerisation). Pastes were mixed by shaking for 5–10 min to give complete homogenisation. The mixtures were cast in ø30 mm × 60 mm polypropylene cylinders. The moulds were half-filled, vibrated for 30 s by ultrasonification, filled to the top, again vibrated for 30 s, and sealed with the lid. It has been found that ultrasonification enhances the dissolution of Al–Si materials in alkaline solutions [16]. The mixtures were then precured for 24 h at room temperature (this precuring time has been found to be beneficial to strength development [10]). Precuring time before application of heat induces significant dissolution of silica and alumina from fly ash and formation of a continuous matrix phase, increasing, therefore, the homogeneity of the geopolymeric materials [10,22]. After the precuring process, the mixtures were cured in an oven at a fixed temperature (40–80 °C) for a fixed time (6–48 h), and finally allowed to set at room temperature before being removed from the moulds to be subjected to their characterisation.

2.3. Geopolymer characterisation

Geopolymer samples were characterised with regard to their structural stability, mineralogy, structure, morphology and leaching behaviour. Structural stability was determined for all the synthesised geopolymer samples by compressive strength measurements. Such determinations were performed at the age of 28 days using a Toni Technik compressive strength testing unit. Three cylinders of each sample were tested, with the experimental values being averaged. Finely ground geopolymer samples synthesised at the optimal conditions of temperature and time (in relation to their compressive strength) were also analysed for their mineralogy, structure and morphology by XRD, FTIR and SEM, respectively, employing the same conditions and equipments mentioned before. Geopolymer samples displaying the highest compressive strength values were also studied concerning their leaching behaviour following the DIN 38414-S4 [21] and NEN 7375 [23] procedures, and analysing the pH, and the content of elements of environmental concern by ICP-MS. The former leaching test is performed on ground materials, while the latter one on monolithic materials. This test was conducted on monolithic cylindrical specimens (ø58 mm × 93 mm, dimensions meeting the leaching test requirements regarding the leachant volume to solid volume ratio and the height of leachant level surrounding the solid). Thus, according to this tank test such specimens were subjected to leaching in a closed tank using deionised water as leachant, being renewed at certain time intervals (0.25, 1, 2.25, 4, 9, 16, 36 and 64 days). The leachant volume to solid volume ratio employed in the study was 3.

3. Results and discussion

3.1. Compressive strength measurements

Fig. 1 represents the compressive strength development of synthesised geopolymers at the different concentrations of activation medium. The compressive strength shown by the geopolymers synthesised using the most concentrated alkaline solution (12 M NaOH) was the highest for all the fly ashes subject of study, except for the FA-3 sample for which a less concentrated medium (8 M NaOH) appeared as the most suitable for this aim. The compressive strength developed in the optimal NaOH concentrations can be considered intermediate for geopolymers arising from the FA-2 and FA-3 fly ash samples (29 and 33 MPa, respectively), and high for those from the FA-1 and FA-4 samples (52 and 66 MPa, respectively). The former values are similar to that obtained for mortars (29 MPa) prepared with high-resistance cement CEM 52.5 R (compressive strength 52.5 MPa at 28 days) using a sand:cement:water ratio of 6:2:1.
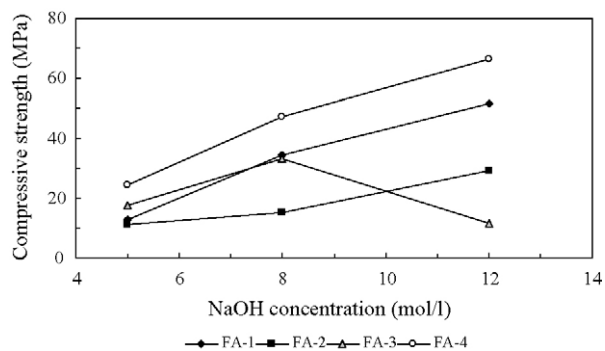

Fig. 1. Compressive strength development of fly ash samples subject of study after the activation process (curing time, 48 h; curing temperature, 80 °C) at different NaOH concentrations.
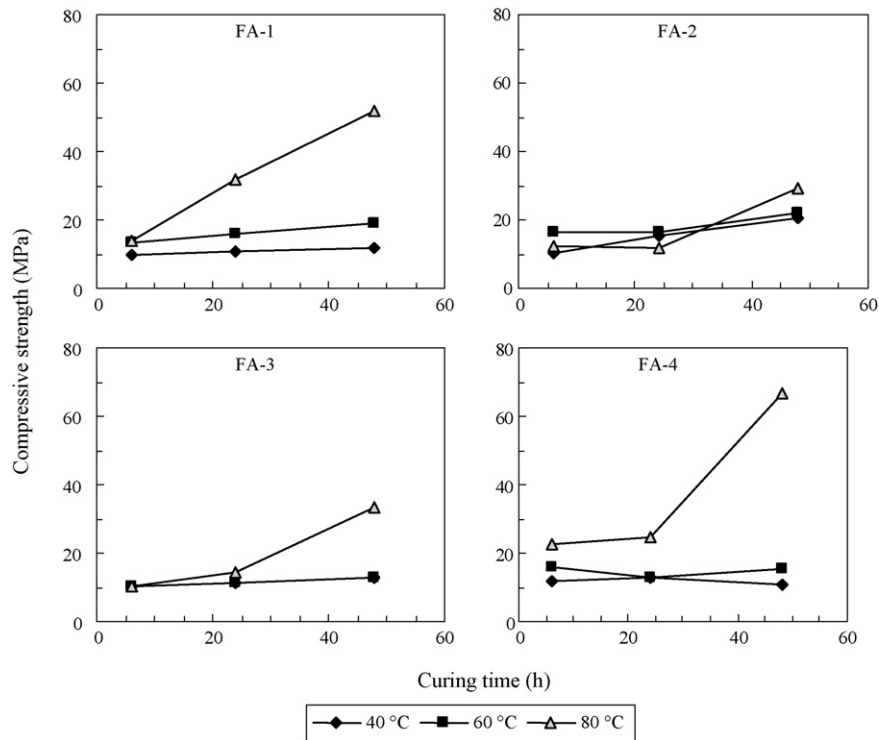
Fig. 2. Compressive strength development of fly ash samples subject of study at different times and temperatures of curing after the activation process at the optimal NaOH concentrations for compressive strength development.

Fig. 2 represents the compressive strength values achieved at different times and temperatures of curing for the fly ash samples activated using the optimal NaOH concentrations for compressive strength development. In general, the activation at 40 °C at any curing time resulted in quite low compressive strength values (about 10 MPa), except for the FA-2 sample for which compressive strength rose up to about 20 MPa when cured during 48 h. Activation at 60 °C, although enhancing in general the performance of the geopolymer matrixes synthesised at 40 °C, gave also poor compressive strength values, being again FA-2 the sole sample attaining a value about 20 MPa when cured during 48 h. Conversely, activation at 80 °C provided a high compressive strength increase for any of the studied samples when the curing period lasted for 48 h. Curing time showed generally a positive effect in the compressive strength performance of geopolymers, though this effect was much more marked at the optimal curing temperature (80 °C), and when curing time was increased beyond 24 h. Fly ash dissolution is an endothermic process requiring, therefore, energy to progress, such energy can be supplied either by thermal treatment or by incorporating in the geopolymerisation process materials of exothermic dissolution. The latter option has been greatly practiced, using preferably metakaolinite due to its strongly exothermic dissolution [24]. Nevertheless, this is an expensive material for large-scale geopolymer manufacture. From the light of the present results, it could be derived that the energy needed for the required extent of fly ash dissolution for the geopolymerisation process to proceed can be supplied by thermal treatment at 80 °C during 48 h.

3.2. X-ray diffraction analysis

Fig. 3 shows the XRD patterns of fly ash samples as well as those of geopolymers synthesised from them. The XRD patterns of fly ashes displayed peaks due to quartz, mullite and iron oxides either magnetite or hematite or both, also other crystalline phases such as anhydrite, calcite, anorthite-albite, illite and microcline were also detected in some samples, but always at trace level. In addition to these crystalline components, a broad peak in the region 20–32° 2θ arising from the glassy phase of fly ashes could be observed in all the patterns. For all the studied fly ashes the intensity of this peak was progressively decreased after the geoplymerisation processes with the activation medium concentration increase. This decrease is related to the high glassy phase content of fly ashes. Thus, it was more evident for the FA-1 and FA-4 samples, those displaying higher glass content. The alkalinity raise also provoked a progressive intensity increase of a broad peak appearing in the region below 8° 2θ, except for the FA-3 sample for which the intensity increase of this peak was similar for the different activation media. The occurrence of such a peak suggests the formation of a mesoporous material (a material with pores in the range 20–50 nm in diameter) of poorly crystalline nature. In addition, reflections of phases such as zeolite X, zeolite 4A, chabazite, sodalite and zeolite NaP1 were present in the XRD patterns of geopolymer samples. Chabazite and sodalite occurred in most samples, while zeolite X and zeolite 4A were only present in geopolymers synthesised from the least alkaline activation medium (5 M NaOH), and zeolite NaP1 in those synthesised from the most alkaline activation media (8 and 12 M NaOH). It is quite frequent the presence
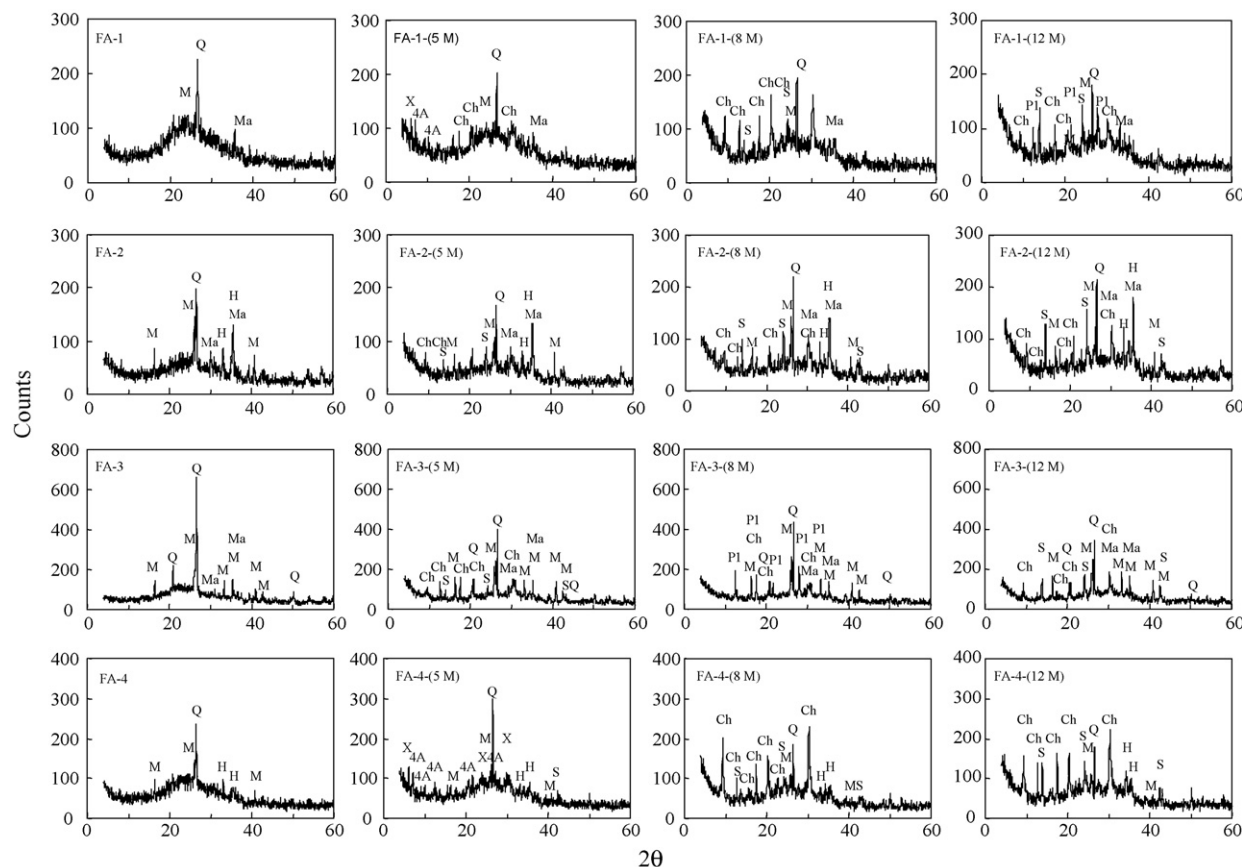
Fig. 3. XRD diffraction patterns of original fly ashes and geopolymer samples arising from them after the activation process (curing time, 48 h; curing temperature, 80 °C) at different NaOH concentrations (Q, quartz; M, mullite; Ma, magnetite; H, hematite; Ch, chabazite; S, sodalite; X, zeolite X; 4A, zeolite 4A; P1, zeolite NaP1).

of zeolite phases in geopolymers proceeding from the activation of fly ashes with alkali hydroxides [10,25]. It is considered that the geopolymer compressive strength could be increased due to increased crystallization contacts between zeolite and amorphous phase. However, there will be a certain limit to the fraction of crystalline phases that can be tolerated by the matrix, after which increased crystallinity will serve to weaken it [8].

According to the semi-quantitative mineralogical composition of fly ash samples subject of study (Table 3), the content of glass in the FA-1 and FA-4 samples attains values close to

Table 3
Mineralogical composition (wt%) of fly ashes subject of study as determined by XRD analysis

	FA-1	FA-2	FA-3	FA-4
Glass	94	80	85	93
Quartz	0.8	2.5	4.6	1.6
Mullite	4.4	9.3	10	4.6
Hematite	<0.1	3.4	0.5	<0.1
Magnetite	<0.1	3.4	<0.1	<0.1
Anhydrite	0.4	0.7	0.2	0.7
Calcite	<0.1	<0.1	<0.1	0.1
Anorthite-albite	<0.1	<0.1	<0.1	<0.1
Illite	<0.1	<0.1	<0.1	<0.1
Microcline	<0.1	0.2	0.1	<0.1

95%, while for the FA-2 and FA-3 samples the corresponding glass content displays values in the range 80–85%. Such differences in the amorphous content could explain the different compressive strength values displayed by fly ashes after being subjected to the activation process. The FA-1 and FA-4 samples, those with the highest glass content, yielded geopolymers with the greatest compressive strength. The higher the content of the glassy constituent in the fly ash, the higher the degree of reaction is. When fly ashes are mixed with the alkaline solution, the vitreous component is quickly dissolved, while if high contents of mullite or quartz are present in the fly ash, the reactivity of the Al–Si bearing fraction decreases drastically [12]. This is probably why the FA-3 sample, although having the highest SiO_2 and Al_2O_3 content, did not yield geopolymer samples with the best structural stability. In the case of the FA-2 sample, the relatively high content of iron oxides may contribute also to the obtained relatively low compressive strength [26]. On the other hand, the lower particle size of FA-1 and FA-4 fly ash samples could also contribute to the good performance of geopolymers arising from them.

3.3. Scanning electron microscopy

Fig. 4 shows morphological features of the original fly ashes and derived geopolymers. Different degree of reaction was
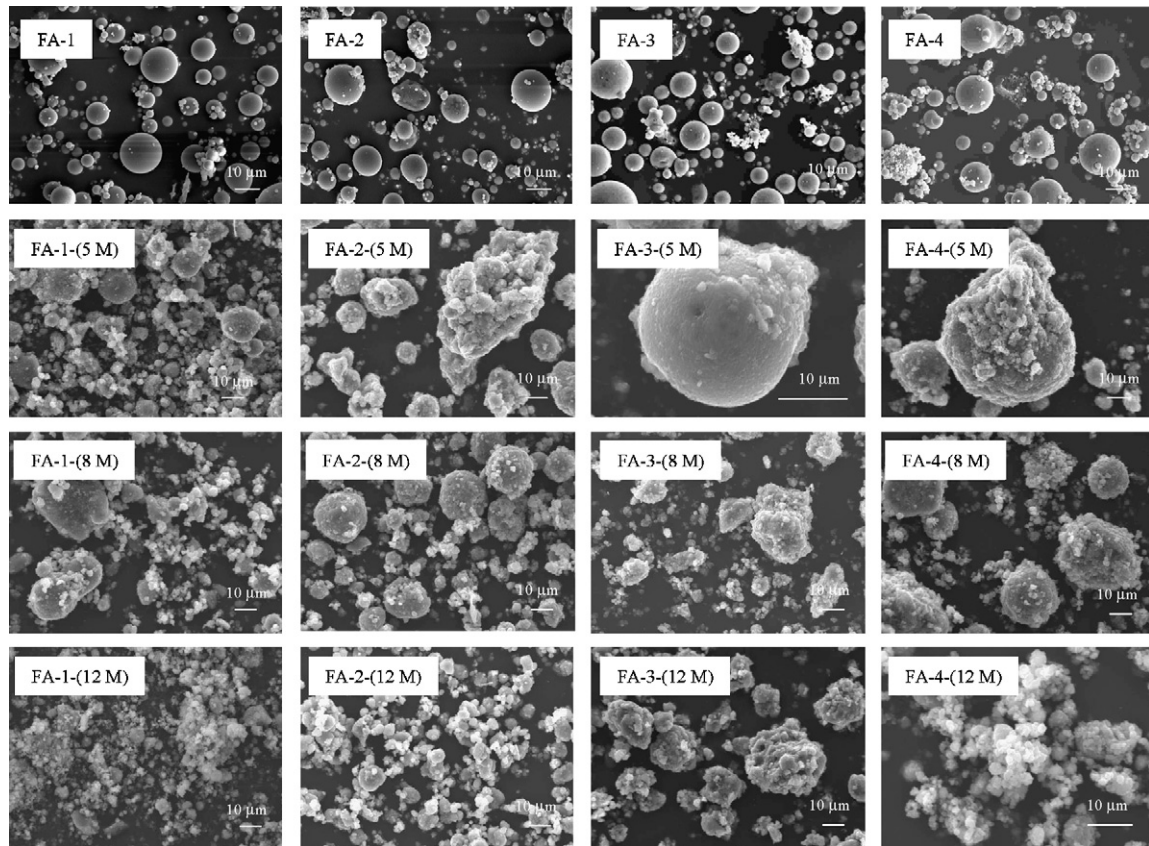
Fig. 4. SEM images of original fly ashes and finely ground geopolymer samples arising from them after the activation process (curing time, 48 h; curing temperature, 80 °C) at different NaOH concentrations.

observed at the different concentrations of activation medium for the different fly ashes subject of study. When the least concentrated alkaline solution (5 M NaOH) was employed in the activation process this always yielded the lowest degree of reaction, although great differences between fly ashes were apparent, with the FA-1 and FA-3 samples being the most extreme examples. Thus, whereas the FA-1 sample displayed a relatively advanced stage of reaction with numerous fly ash particles being involved in the alkaline activation, and coexisting with some partially reacted or unreacted particles, the FA-3 sample was characterised by a much lower degree of reaction (slight surface activation of particles). When the most concentrated medium (12 M NaOH) was employed in the geopolymerisation the alkaline activation was more effective, especially for the FA-1 and FA-4 samples. Conversely, in the case of the FA-3 sample, a state similar to that found when an 8 M NaOH solution was used as activation medium in the geopolymerisation process was observed. Such observations agree well with the compressive strength measured for fly ashes after the activation process: the higher was the degree of reaction, the higher were the compressive strength values. It is worth noting that although the presence of zeolite phases was found by XRD analysis in all the samples at any activation medium, morphologically no zeolite crystals were detected by SEM, suggesting that these may occur as very small crystals.

3.4. Fourier transform infrared spectroscopy analysis

Fig. 5 shows the infrared spectra of the original fly ashes and geopolymer samples synthesised from them. Original fly ash samples showed two main spectral peaks at about 480 and 1110 cm^{-1}, the former has been assigned to Si–O–Si and O–Si–O bending, and the latter to Si–O–Si and Al–O–Si asymmetric stretching [27,28]. In addition to these two bands, the spectra of untreated fly ashes showed three other peaks associated with their Al–Si framework, generally of much lesser intensity, at about 810, 750 and 590 cm^{-1}, being attributed to Si–O–Si and Al–O–Si symmetric stretching [27,28]. The stretching modes are sensitive to the Si–Al composition of the framework and may shift to a lower frequency with increasing number of tetrahedral aluminium atoms [29]. Activation of fly ashes caused structural reorganisation as could be derived by the spectral shifts displayed by the infrared spectra of geopolymer samples synthesised from them. The main spectral band appearing originally at about 1110 cm^{-1} shifted to lower frequencies, this decrease was comprised between 45 and 110 cm^{-1} depending on the fly ash involved in the geopolymerisation process. Important variations in the band intensity were also observed at the different activation media for a given fly ash sample. Generally, the intensity of this band rose with the alkalinity increase of activation medium, except for samples synthesised from the FA-3 fly ash for which the highest intensity of this
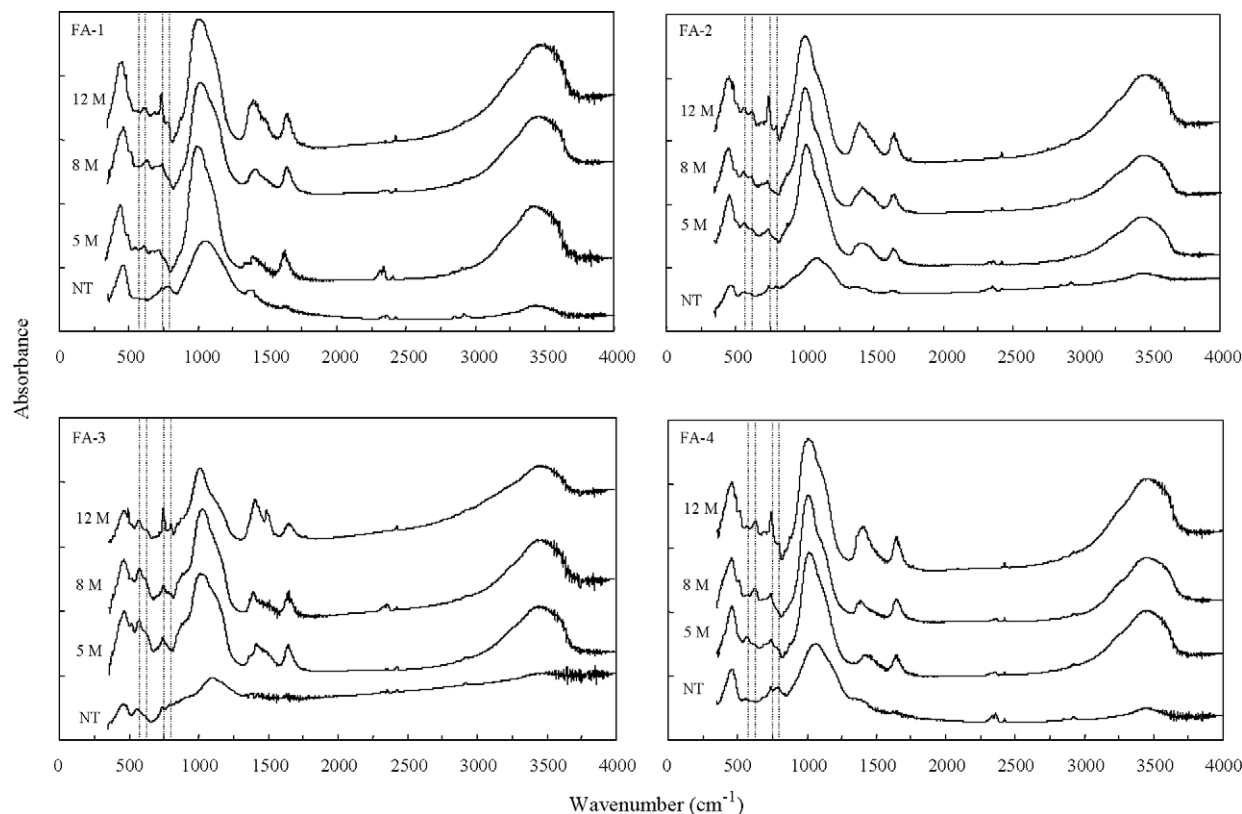
Fig. 5. FTIR spectra of original fly ashes (NT) and geopolymer samples arising from them after the activation process (curing time, 48 h; curing temperature, 80 °C) at different NaOH concentrations (dashed lines indicate positions at 590, 635, 750 and 810 cm^{-1}).

band was observed at the 8 M NaOH activation medium. The intensity increase of this band is indicative of an increase of a mean chain length of aluminosilicate polymers [10]. This finding is consistent with the compressive strength values displayed by geopolymer samples at the different activation media as well as with the observations derived by SEM. Other spectral changes were also manifested after the geopolymerisation process: the peak at about 810 cm^{-1} decreased or disappeared at low-intermediate activation media, the peak at about 750 cm^{-1} increased and a new peak at about 635 cm^{-1} associated to Si–O–Si and Al–O–Si symmetric stretching [27,28] was generally manifested. After the fly ash activation, besides spectral changes related to the Si–Al framework, also broad bands at about 3485 and 1655 cm^{-1} were present in the spectra of geopolymer samples, these bands are associated to –OH stretching and H–O–H bending, respectively, being characteristic of weakly bound molecules of water either surface adsorbed or trapped in large cavities. Other spectral band at about 1435 cm^{-1} appeared in all the geopolymer samples. This band is characteristic of carbonate asymmetric stretching, what suggests the presence of sodium carbonate due to the atmospheric carbonation of alkaline activation media.

3.5. Leaching studies

Fig. 6 shows the leachable content of elements of major environmental concern derived from the DIN 38414-S4 [21] leaching test both in original fly ashes and in ground geopolymer

samples synthesised at the optimal conditions for compressive strength development (leaching contents in geopolymers have been weighed to the fly ash proportion applied in each geopolymer sample). In general, three groups of elements can be established according to their leaching behaviour. One group is made up of elements being released at similar level from the fly ash samples and from the corresponding synthesised geopolymers (Co, Ni, Cu, Zn, Cd, Pb), in this case leaching remained always very low (<0.2 mg/kg). A second group is composed by elements occurring as oxyanions (As, Mo, Se, Sb, Cr), such elements displayed higher releases from the geopolymer samples than from the corresponding fly ashes, specially high release increases were found for arsenic for any of the samples subject of study. This behaviour seems to be related to the higher alkaline character of leachates arising from geopolymer samples (pH range: 11.0–12.5) in comparison with those from fly ashes (pH range: 8.8–10.7), and so to the elevated sodium levels relative to calcium in the geopolymer samples. Generally, oxyanions form much more soluble compounds with sodium than with calcium. The opposite behaviour was shown by barium which displayed higher releases from the fly ashes than from the corresponding synthesised geopolymers. The higher alkalinity of geopolymer leachates or/and the high replacing power of this cation could explain its lesser mobility after geopolymerisation. Fig. 6 also shows the cumulative leachable content of elements of major environmental concern derived from the NEN 7375 [23] monolithic leaching test. Although it could be expected that immobilisation of oxyanions might occur by phys-
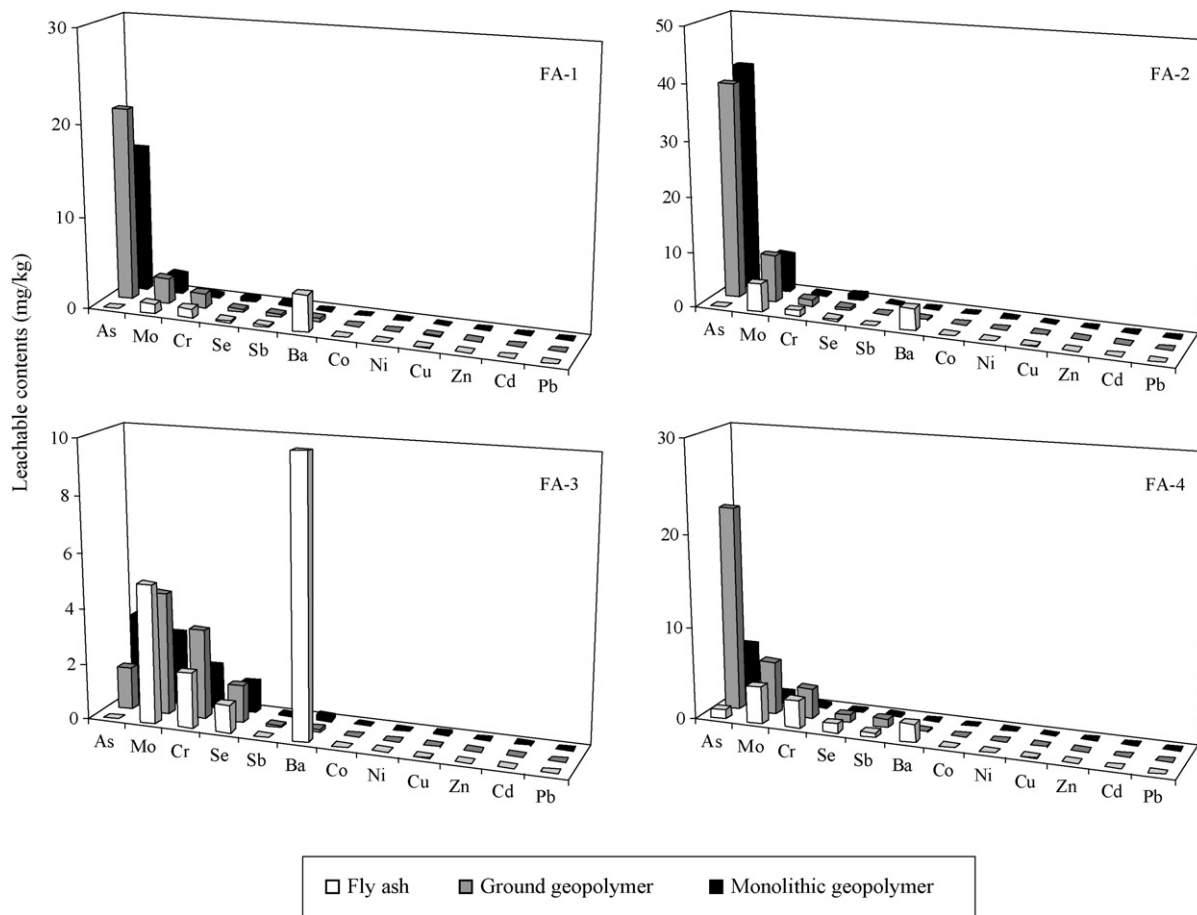
Fig. 6. Leachable contents of elements of environmental concern in fly ashes and in geopolymers synthesised from them at the optimal conditions for compressive strength development (leaching contents in geopolymers have been weighed to the fly ash proportion applied in each geopolymer sample: %FA in geopolymers: 100/1.3 for geopolymers arising from FA-1, FA-2 and FA-4 and 100/1.4 for those from FA-3) (leaching tests: DIN 38414-S4 [21] for fly ash and ground geopolymer and NEN 7375 [23] for monolithic geopolymer).

ical entrapment, rather than chemically, due to the low porosity of geopolymers, from the light of results derived from the NEN 7375 [23] monolithic leaching test it could be concluded that such a mechanism only seems to contribute slightly for some elements, and specially in the case of geopolymers synthesised from the FA-4 sample. This behaviour should be related to the lower porosity of geopolymers arising from this fly ash sample.

4. Conclusions

Compressive strength development of geopolymers synthesised using coal (co-)combustion fly ashes as the sole source of silica and alumina was greatly dependent on both the synthesis conditions and the characteristics of fly ashes subjected to the activation process. Higher compressive strength values were attained at curing time of 48 h, curing temperature of 80 °C and 8 and 12 M NaOH solutions as activation media. Such conditions were necessary for the required extent of fly ash dissolution for the geopolymerisation process to proceed, as no materials of exothermic dissolution or/and additional sources of silica and alumina were incorporated in the synthesis process. Fly ashes with glass content higher than 90% appeared as the

most suitable to synthesise geopolymer matrixes of high structural stability, values about 60 MPa were achieved from them at the optimal activation conditions. It was found that synthesised geopolymer matrixes were only effective in the chemical immobilisation of a number of elements of environmental concern contained in fly ashes, reducing (especially for Ba) or maintaining their leachable contents after the geopolymerisation process, but not for those elements present as oxyanions. Physical entrapment does not seem either to contribute in an important way, in general, to the immobilisation of oxyanions at the high pH and sodium levels of the synthesised geopolymers.

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