

Synthesis of microwave-sintered ceramics from lignite fly and bottom ashes

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Management of huge quantities of fly ash and bottom ash produced from lignite combustion in power generation units is nowadays of great environmental concern. The use of these industrial solid wastes in the development of value-added materials is a significant subject. Microwave sintering can be an efficient, economic and valuable approach for the processing of powdery materials for ceramics manufacturing, because microwave processes, with their selective and volumetric heating, can enhance the solidification efficiency and reduce the processing time considerably thereby leading to energy savings. In the present research, microwave sintering of Class C (high-Ca) lignite combustion fly ash and bottom ash towards ceramic microstructures development is investigated. For microwave processing, fly ash, bottom ash and mixtures of them were cold pressed to form a series of disc-shaped specimens. Microwave sintering of the ash specimens was carried out at 1000 °C for 10, 20 or 30 minutes in a special purpose microwave (2.45 GHz) system designed for rapid laboratory heating of materials. The microstructures produced were characterized by means of XRD, SEM-EDX analyses and density measurements, and the effectiveness of the sintering process was evaluated. The experimental results are encouraging, as reasonably dense ceramic microstructures mainly composed of gehlenite, quartz and kyanite are achieved after rapid sintering (30 minutes) of the compacted ash specimens using microwave radiation. Several experimental parameters, including ash particle size and shape, green density of the compressed powders, and sintering time appear to influence the consolidation efficiency by microwave heating.

Key words: Microwave sintering, Ceramics, High-Ca fly ash, Bottom ash, Valorization.

Introduction

Management of massive quantities of fly ash and bottom ash that are annually produced from lignite combustion in power generation units is nowadays of great environmental concern. Actually, only a small amount of the ash is currently used while the rest is landfilled, a situation that will possibly cause severe and irreversible long-term environmental effects [1-4]. Use of these industrial by-products as secondary raw materials in the manufacturing of value-added products can contribute to environmental protection, resource conservation as well as cost reduction [5-8]. Besides, current advances in environmental legislation encourage manufacturers to optimize industrial by-product management. Therefore, sintering of lignite ashes towards ceramics development represents a significant subject.

Microwave radiation is emerging as a novel and innovative technology, widely applied in various fields. Recent microwave applications include decomposition of cellulose for biomass exploitation, preparation and regeneration of activated carbon, as well as several applications in green chemistry, biosciences and metallurgical processes [9-15]. In particular microwave

sintering, can be an efficient, economic and valuable approach for the processing of some powdery materials [16-19]. Indeed, a growing interest in the sintering of various ceramics, glass-ceramics, nano-ceramics and bio-ceramics through microwave processing is reported [20-30]. Microwave energy offers many advantages for eco-friendlier processing of materials over conventional processing. In fact, microwave processes, with their selective and volumetric heating, can enhance the solidification efficiency and reduce the processing cycle time considerably, thereby resulting in substantial energy and cost savings. They also provide finer microstructures leading to improved mechanical properties. These characteristics provide sufficient motivation to promote the use of microwaves in “greener” materials processing. The potential use of microwave technology as an energy-efficient alternative to conventional heating technologies employed in the processing and treatment of waste, such as used tyres, plastic waste and even sewage sludge, is currently being investigated [31]. Combustion ashes in particular, specifically Class-F (siliceous) coal fly ashes [32, 33], MSWI fly ashes [34, 35] and even SiO₂-rich rice-husk ash [36] have already been considered as secondary raw materials for the development of various ceramic, glass-ceramic or zeolite crystalline structures, substituting traditional heating by microwave rapid sintering or vitrification technology.

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In the present research, microwave sintering of Class C (ASTM C 618) (i.e. high-Ca) lignite combustion fly ash and bottom ash towards ceramic microstructures development is investigated. The ashes were derived from a lignite-fed power station situated in Northern Greece (the region of West Macedonia where the main lignite deposits of the country are located). The ash composition, rich in Ca, can be expected to yield an interesting and possibly more complex mineralogy in the sintered materials. Moreover, Ca may act as a flux enabling melting to begin at lower temperatures (thus using less energy), which could provide beneficial formation of a thin liquid layer between ash particles during sintering, tending to pull them together. In a previous preliminary study, ashes of similar composition have been used by the authors for conventional furnace sintering [37]. By microwave processing, significant temperature gradients between the surface and the interior that are frequently encountered upon conventional sintering at high heating rates can be avoided, which is of particular interest especially for large-size workpieces fabrication from low thermal conductivity raw materials such as fly ash mainly consisting of hollow sphere-shaped particles. Thus, accelerated solidification, a uniform microstructure and quality of the sintered products can be expected by microwave sintering of the lignite calcareous ashes under investigation.

Experimental

Microwave heating

Microwave irradiation (usually at the ISM - Industrial Scientific and Medical - frequency of 2.45 GHz) can produce efficient internal heating. With microwave processes, it is possible to achieve rapid and uniform heating of thick materials. Actually, microwave energy is delivered directly to the material through molecular interaction with the electromagnetic field, as microwaves can penetrate the material and supply energy.

The power absorbed per unit volume, P (W/m^3), is expressed as follows (Eq. 1) [12]:

$$P = \sigma |E|^2 = 2\pi f \epsilon_0 \epsilon_r \tan\delta |E|^2, \quad (1)$$

where E (V/m) is the magnitude of the internal field, σ the total effective conductivity (S/m), f the frequency (GHz), ϵ_0 the permittivity of free space ($\epsilon_0 = 8.86 \times 10^{-12}$ F/m), ϵ_r the relative dielectric constant and $\tan\delta$ is the loss tangent.

From Equation 1, the dielectric parameters (dielectric constant and loss tangent) determine the effect of a given microwave electromagnetic field on the material. In particular, the dielectric constant measures the ability of a material to store microwave energy. The loss tangent provides an indication of how well a material can be penetrated by the electric field and how it dissipates energy into heat [15].

Since energy transfer occurs at a molecular level, the interaction of microwaves with a dielectric material results in translational motions of free or bound charges and rotation of the dipoles. The resistance of these induced motions due to inertial, elastic, and frictional forces causes heat losses, and thereby heat can be generated throughout the volume of the material resulting in volumetric heating, thus reducing processing time and enhancing product quality. Thus, the thermal gradient in a microwave-processed material is the reverse of that in a material processed by conventional heating, where slow heating rates are normally selected to reduce an abrupt thermal gradient possibly leading to process-induced stresses.

Moreover, rapid heating that is possible upon microwave sintering usually favors densification while limiting excessive grain coarsening. Therefore, high densities can be attained with a fine grain structure.

Materials

The highly-calcareous fly ash (FA) (Class-C fly ash according to ASTM C 618) utilized as a secondary raw material in the present research, a fine powder, was obtained by the electrostatic precipitation of dust-like particles from the flue gases of the Ptolemais lignite combustion power plant situated in Northern Greece (the region of West Macedonia).

The bottom ash (BA) used, a granular material much coarser than FA, also formed during lignite firing, was removed from the bottom of dry boilers of the same power plant.

The chemical characterization results for these ashes are given in Table 1.

According to the chemical analysis results, the main components of FA and BA are calcium, silicon, aluminum and iron compounds. The SiO_2 and Al_2O_3 contents of these ashes are due to the aluminosilicate components of lignite. FA in particular, is characterized by a high % CaO content (similarly to other FAs from power plants of Northern Greece belonging to class-C, i.e. calcareous FAs), mainly attributed to the intense presence of limestone ($CaCO_3$) in feedcoal (lignite). Actually, limestone is frequently co-extracted with lignite, and then its thermal disruption in the combustion chamber leads to high free lime percentages (approximately 5%) in the fly ash produced. BA is less abundant in Ca, but contains a higher residual carbon content. Mo, Ba, Cr, Cu, Ni and even Pb, V and Sr constitute the most important trace elements found in the ashes.

Table 1. Chemical analysis of Greek (Ptolemais) fly ash (FA) and bottom ash (BA).

Com- pound	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	Na ₂ O	K ₂ O
FA	30.16	14.93	5.10	34.99	2.69	6.28	1.01	0.40
BA	48.63	21.62	7.29	6.83	2.75	2.78	0.89	2.97

Preparation of ash specimens

FA, BA and 50-50 wt% mixtures of them were uniaxially cold pressed in a stainless steel die using a hydraulic press (SPECAC, 15011) to form a series of 13 mm diameter disc-shaped green compacts (pellets). A lubricant was applied on the die wall. The strength of the green compacts increased with increasing compacting pressure, due to a reduction in porosity as a result of filling of empty spaces existing between particles by the ash powders. All compacts obtained at 500 MPa had a reasonable green strength for the subsequent fabrication step.

Microwave sintering of the green specimens was carried out at 1000 °C for 10, 20 or 30 minutes in a special purpose microwave (2.45 GHz) system designed (PHOENIX, Microwave Muffle Furnace) for laboratory rapid heating of materials. Finally, the sintered specimens were gradually cooled to ambient temperature in the furnace.

Phase characterization of both the green and sintered specimens was carried out via X-ray diffraction (XRD), employing identical equipment (Siemens, Diffractometer D-5000) and analytical conditions in order to enable a reliable comparison of the X-ray patterns. Microstructural examination of the specimens was realized by scanning electron microscopy (SEM - Jeol, JSM-6400). The density was determined by means of a specific apparatus (Shimadzu, SMK401-AUW220V) according to the Archimedes principle. The effectiveness of the sintering process was evaluated as a function of the sintering time and specimen composition.

Results and Discussion

Photographs of green and microwave sintered specimens are presented in Fig. 1.

From Fig. 1, the compressed green pellets prepared from the 50-50 wt.% FA-BA mixture (a) are integral and brown (due to the dark brown color of BA). After microwave sintering at 1000 °C for 30 minutes, successfully consolidated earth-yellowish materials are obtained.

In Fig. 2, the main mineral phases present in the green specimens prepared of FA or BA as well as in the microwave (Micro: 30 minutes) sintered ones, as



Fig. 1. Photographs of specimens (diameter: 13 mm) made of 50-50 wt.% FA-BA mixture:

(a) green and (b) microwave sintered at 1000 °C for 30 minutes.

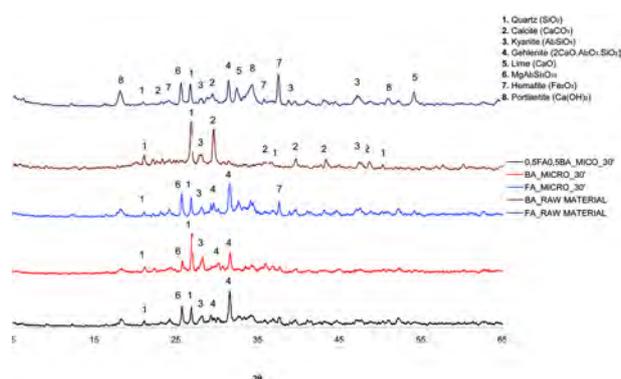
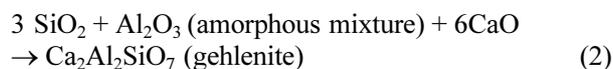


Fig. 2. Typical XRD spectra of green and microwave sintered ash specimens.

determined by means of XRD, are shown.

From Fig. 2, among the major crystalline mineral compounds identified in the green specimens prepared of FA and of BA are quartz, gehlenite, lime, calcite, portlandite, hematite and kyanite. The existence of calcareous compounds such as calcite and portlandite is attributed to the interaction of free CaO (see the materials section) with humid air, and the subsequent formation of Ca(OH)₂ and CaCO₃. Quartz, a hard and crumble resistant mineral, the most unreactive component of ash, is frequently found on the ash particle surfaces, probably in relatively unchanged conditions, as found before combustion. The hematite presence is due to the ferrous compounds that occur in the lignites. The ash is also constituted of an amorphous phase of aluminosilicate glass indicated by a broad pattern or “hump” registered in the diffractogram. It should be noted that the amorphous components of a given material, such as the Al-Si glassy matrix of fly ash, produce not only such humps in the XRD spectra, increasing the XRD background, but also a reduction of the peak intensities derived from the crystalline components, due to dilution and/or mass absorption effects [38].

Sintered specimens of FA, BA and a 50-50 wt% mixture of them (Fig. 2) exhibit generally similar mineralogical compositions between them: the major crystalline phases identified are gehlenite, quartz and kyanite. Apparently, the thermal treatment of the green specimens lead to some changes to the mineralogical structures of the ash compositions tested. Certainly, gehlenite, quartz and kyanite, which are detected after microwave sintering, were also present originally in the ashes. Obviously, the intensity of the peaks associated with gehlenite predominates as a result of reactions (Eq. 2) occurring during heating [39]:



On the other hand, lime, calcite and portlandite tend to disappear upon sintering at 1000 °C for 30 minutes. Quartz is generally stable up to 1100 °C, while calcite

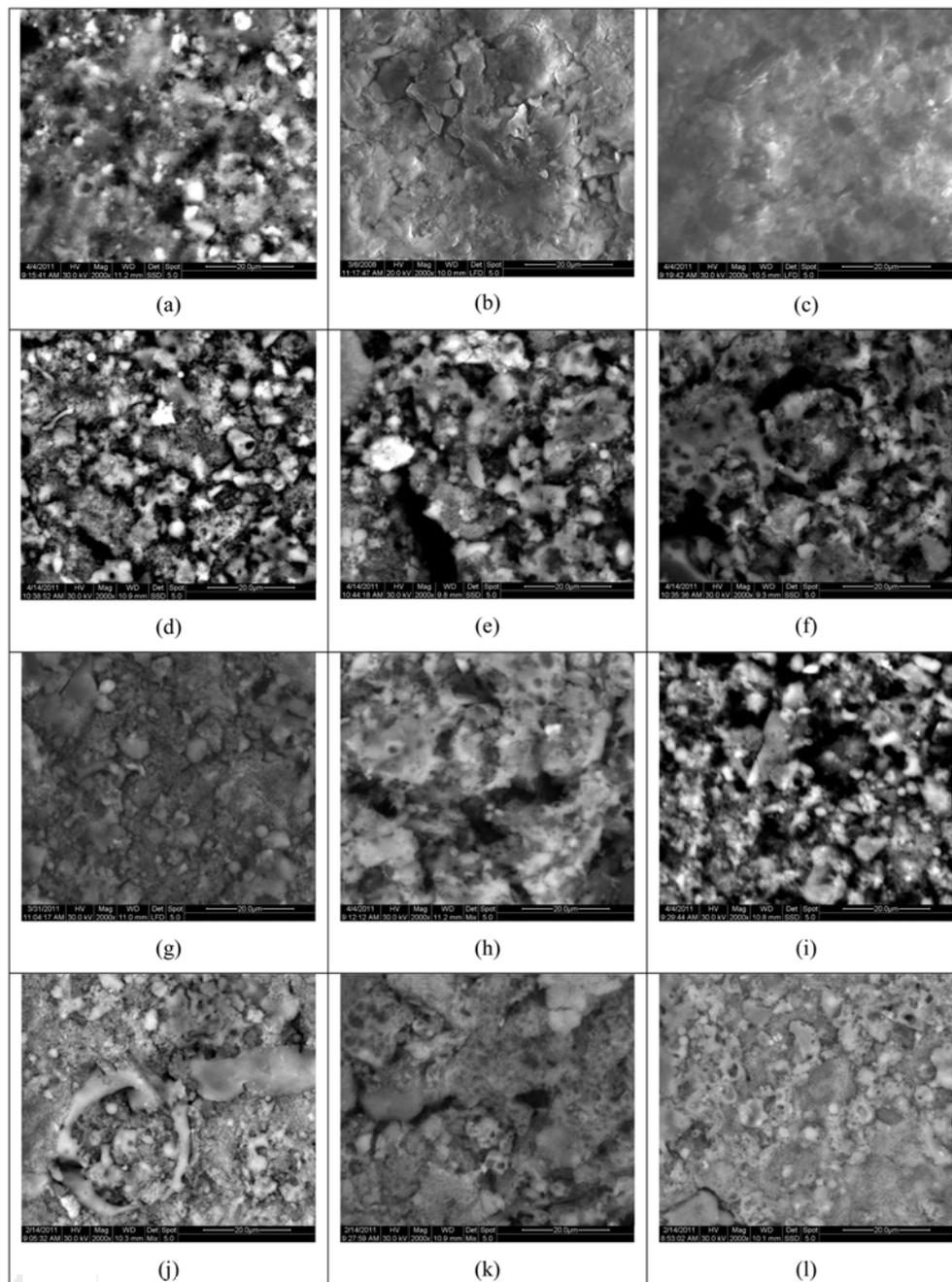


Fig. 3. Micrographs of specimens made of FA (a, d, g, j), BA (b, e, h, k) and 50-50 wt.% FA-BA mixture (c, f, i, l), green or microwave sintered at 1000 °C for 10, 20, 30 minutes, respectively (from top to bottom).

decomposes at temperatures lower than 900 °C. A magnesium aluminosilicate compound ($\text{MgAl}_2\text{Si}_3\text{O}_{10}$) is also identified in both the green and sintered specimens. Furthermore, the background levels shown in the XRD patterns after microwave sintering indicate that the final products should contain some amorphous aluminosilicate glass (as was also detected in the green specimens).

The effectiveness of the consolidation process can be evaluated upon microstructural observation of green and sintered specimens via SEM analyses (Fig. 3).

In the green specimen made of FA (Fig. 3a), several

separate micro-spheres (cenospheres, 1-3 μm) appear. The hollow-sphere shape that these ash particles assume was obtained during instantaneous cooling of the lignite combustion gases upon contact with air, and caused by the surface tension forces acting on the melt to minimize the surface free energy [40]. Particles with an irregular shape are also found. Rupture of FA cenospheres into pieces during the compaction process could not be excluded. Larger angular particles are revealed in the BA green specimen (Fig. 3b). It is worthy of note that in the case of the compressed ash mixture (FA-BA: 50-50 wt.%, Fig. 3c), a more efficient

particle packing is clearly seen that should be attributed to the difference in particle size and shape between FA and BA powders, which is generally considered beneficial for compaction.

The degree of consolidation after microwave sintering strongly depends on both the sintering time and the starting material used. Hence, the apparent density of the sintered specimens varies in the range of 2.3-2.7 g/cm³. The heating conditions appear significant for the sintering process and therefore for the quality of the final product. Similarly, differences in chemical and mineralogical composition of the by-products that were used as the raw materials should be taken into consideration.

Specifically, when sintering specimens for only 10 minutes, densification is relatively poor, and porous ceramic microstructures appear (Figs. 3 d, e and f). It should be noted however, that porous microstructures could offer advantages regarding specific properties of the materials. Residual porosity may to a certain degree be attributed to the thermal decomposition of calcite contained in both the FA and BA starting materials. Decarbonization as well as dehydration reactions are important factors, as they release CO₂ and H₂O at different temperatures. Porosity is even more intense in both specimens prepared from the BA and FA-BA mixture (Figs. 3e and f) compared to that made solely of FA (Fig. 3d), probably due to burnout of the residual carbon content (which is much more pronounced in the as-received BA), which normally occurs during sintering at a temperature range of 700-750 °C.

Sintering for 20 minutes, improves the degree of solidification, especially for the FA specimens. However, reasonably dense ceramic materials are achieved after 30 minutes sintering for all three compositions. In these microstructures (Figs. 3j, k and l), the development of sintering necks that bind the materials produced can be observed. Since microwave radiation can be efficiently absorbed from silica and alumina in an amorphous state [33], the existence of the aluminosilicate glass phase in the secondary raw materials under consideration in the present study should contribute to facilitating microwave energy absorption, thus enhancing sintering.

The phase diversity in the starting materials may count for the formation of some glassy structures (see Fig. 3j) indicative of localized viscous flow that can be associated with hot spot or localized thermal runaway phenomena reported for microwave sintering [41]. Actually, when ceramic systems are processed in non-uniform electromagnetic fields, the local temperature varies within the bulk of the material. If a local area reaches the critical temperature before the rest of the material, it begins to become heated more rapidly, the temperature of that region beginning to locally increase even more, possibly above the glass transition temperature. In heterogeneous systems such as the lignite combustion ashes used containing a mixture of crystalline and amorphous components, there are

particles with higher and lower melting temperatures, thus behaving differently during sintering, and particularly microwave sintering. Normally, the alkalis and alkaline earth oxides are the most reactive components present in the ash composition that provide an intergranular thin liquid phase, involving mass transport phenomena, and locally wetting the solid grains, thus providing a capillary force that pulls the grains together. Particularly, the high Ca content of the lignite fly ash used possibly acts as a fluxing agent enabling melting to begin at lower temperatures thus accelerating sintering. The presence of such glassy structures is also confirmed by the amorphous phase already identified in the sintered specimens via XRD analysis (Fig. 2). In this case, both thermally activated processes, mainly diffusive but also restricted viscous processes coexist in the bulk of the materials [42].

From Fig. 3, the best 30 minutes microwave sintering result among the three compositions tested is obtained for the FA-BA mixture (Fig. 3l), where quartz crystals are located in a densified matrix mainly composed of gehlenite. This finding should be attributed to both the aforementioned more efficient particle packing stated in the case of the compressed ash mixture and a synergistic effect between FA and BA components upon microwave sintering improving consolidation.

Conclusions

The experimental results are encouraging, as reasonably dense ceramic microstructures are achieved after sintering lignite high-Ca fly ash and bottom ash in a short period of time (30 minutes) at 1000 °C using microwaves.

Microwave sintering causes noticeable changes in the amount of the mineralogical phases originally present in the industrial by-product mixtures used as the starting materials, resulting in ceramic materials mainly composed of gehlenite, quartz and kyanite.

Experimental parameters such as the ash particle size and shape, the green density of the compacted powders and the sintering time influence the consolidation efficiency by microwave heating. The highly calcareous nature of fly ash does not hinder the synthesis of ceramics. The distribution of the electromagnetic field inside the microwave cavity, microwave-material interaction, material transformations and heat transfer mechanisms are critical for optimizing the process. Further research is currently underway for a broader assessment of the potential of microwave sintered lignite combustion ashes for utilization in marketed ceramic final products.

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