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Recycling of microwave inertised asbestos containing waste in refractory materials

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Abstract

Asbestos is a health hazard and its removal a priority for pollution prevention. Asbestos containing wastes (ACW) can be transformed into inert silicate phases by means of microwave irradiation. The aim of this investigation was to recycle microwave inertised ACW in mullite–cordierite refractory materials. A MgO-rich talc was replaced by inertised asbestos keeping approximately equal oxide composition of the raw materials. No significant variations of water absorption, linear shrinkage and Young's modulus but a higher occurrence of cordierite phase with the change of raw material was found. This can be considered an important technological result.

Keywords: Asbestos; Microwave processing; Mullite; Refractories

1. Introduction

Asbestos has been frequently used in building materials, it is however dangerous for human health due to the fibrous asbestiform crystal habit, possibly leading to lung cancer when inhaled, as documented in the literature.[1] Even if European regulation allows the recycling of ACW after inertisation (e.g. Italian Decree no. 248 of 29/06/04), disposal in dumps after inertising treatments has been taken as the only definitive solution.

Inertisation of ACW with microwave irradiation has been applied since 1997 in Italy and numerous studies have been carried out by joint research efforts of the University of Modena and Reggio Emilia and ENEA-UTS Tecnologie Fisiche Avanzate.[2] This thermal treatment modifies the dangerous fibrous structure of asbestos and transforms it into inert magnesium oxide containing compounds in the form of solid blocks. The composition of these blocks is based on forsterite and enstatite as major crystalline phases, no hazardous minerals are present.[3]

The purpose of this research was to investigate for the first time the possible recycling of microwave inertised ACW as raw material in the production of refractory materials with compositions similar to those of Italian manufacturers. The starting formulations of the refractory materials were chosen in the cordierite–mullite system. Cordierite–mullite composites find increasing application as refractory plates in fast-firing techniques of ceramic products.[4] Cordierite supplies considerable resistance to thermal shock while mullite provides the required strength. Since few natural deposits of cordierite exist and considering the current cost of magnesium containing raw materials, a stable cordieritic material that can be obtained from an economical source is of great interest for the refractory industry from an economic point of view.

2. Experimental

2.1. Materials and sample preparation

Chemical analysis was carried out on powders of both base commercial compositions in as-received condition by ICP-AES (Model Liberty 200, VARIAN) and the results are shown in Table 1. Massive serpentine asbestos inertised by microwave heat-treatment were available in powder form from previous research.[3] Fig. 1 shows the XRD pattern of this powder evidentiating the presence of forsterite and enstatite as the main crystalline phases. A Mg-rich talc raw material was replaced by the inertised asbestos in percentages of 16 and 20 wt.%. The raw materials and their percentages were varied so that the overall

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Table 1
Cordierite–mullite base refractory and microwave inertised ACW compositions

Sample	Oxide (wt.%)								
	SiO_2	Al_2O_3	CaO	MgO	Na_2O	K_2O	Fe_2O_3	TiO_2	Total
Inertised ACW	42.30	3.78	2.09	42.36	0.05	0.18	9.07	0.17	100
Cordierite–mullite refractory base	54.22	33.19	0.48	8.89	0.14	0.9	1.47	0.71	100

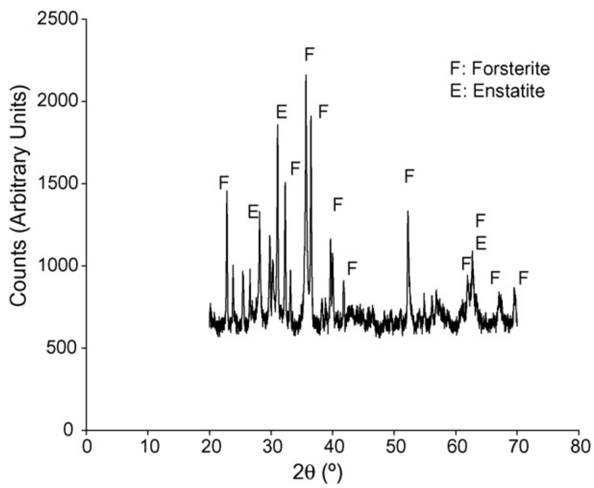

Fig. 1. XRD pattern of microwave inertised asbestos showing forsterite (F) and enstatite (E) as main crystalline phases.

oxide percentages were kept close to those of the commercial base composition, as detailed in Table 2. The dry mixtures were ground in a ball-mill for 40 min. Finally, disk specimens (diameter, 40 mm; thickness, 5 mm) were prepared by uniaxial pressing at 28 MPa without addition of any binder. The green bodies were subjected to an industrial thermal cycle.

2.2. Characterization and test methods

Linear shrinkage and water absorption were measured on representative samples following standard laboratory procedures. X-ray diffraction (XRD) analyses were carried out on powdered samples (Cu Kα, Ni-filtered radiation, Model PW3710 Philips) to characterize the mineralogical phase composition of samples. ICCD files were used to identify the crystalline phases.

The Young's modulus of representative specimens was determined by the impulse excitation technique (Grindosonic® MKS, J.W. Lemmens, Leuven, Belgium). Disk-shaped specimens (diameter, 40 mm; thickness, 5 mm) prepared by uniaxial pressing as explained above were used. Each specimen was run at least five times to correctly identify the resonance frequency and the results were averaged.

3. Results and discussion

Fig. 2 reports the water absorption curve of the mullite–cordierite refractory compositions. Error bars showing minimal and maximal measured values are also reported. Minor variations of water absorption with increasing content of inertised asbestos was found, which indicates small changes in open porosity. Higher open porosity should increase the thermal insulating capacity and decrease the Young's modulus of

Table 2
Cordierite–mullite compositions investigated: commercial base composition and compositions with 16 and 20 wt.% inertised ACW

Raw materials and oxides weight percentages of the refractory base composition		Raw materials and oxides weight percentages of refractories with 16 wt.% of inertised asbestos		Raw materials and oxides weight percentages of refractories with 20 wt.% of inertised asbestos	
Raw material	wt.%	Raw material	wt.%	Raw material	wt.%
Molochite	30.33	Molochite	30.3	Molochite	30.3
Silice fusars	11.24	Silice fusars	6.73	Silice fusars	3.73
Artal	7.49	Artal	9.49	Artal	9.49
Talco HK-70 RS	14.98	Inertised ACW	15.98	Inertised ACW	20
Magnesite RS	3.75	Magnesite RS	4.75	Magnesite RS	4
Argille RR40	20.6	Argille RR40	19.1	Argille RR40	19.1
Alolt 8S	5.62	Alolt 8S	7.62	Alolt 8S	7.62
Argilla UNI-1	5.99	Argilla UNI-1	4.99	Argilla UNI-1	4.99
Oxide	wt.%	Oxide	wt.%	Oxide	wt.%
SiO_2	54.218	SiO_2	54.899	SiO_2	54.994
Al_2O_3	33.186	Al_2O_3	33.164	Al_2O_3	33.029
K_2O	0.9	K_2O	0.843	K_2O	0.804
CaO	0.488	CaO	0.37	CaO	0.348
MgO	8.864	MgO	8.904	MgO	9.028
TiO_2	0.71517	TiO_2	0.5961	TiO_2	0.59417
Fe_2O_3	1.47967	Fe_2O_3	1.6742	Fe_2O_3	1.065
Na_2O	0.149	Na_2O	0.142	Na_2O	0.137

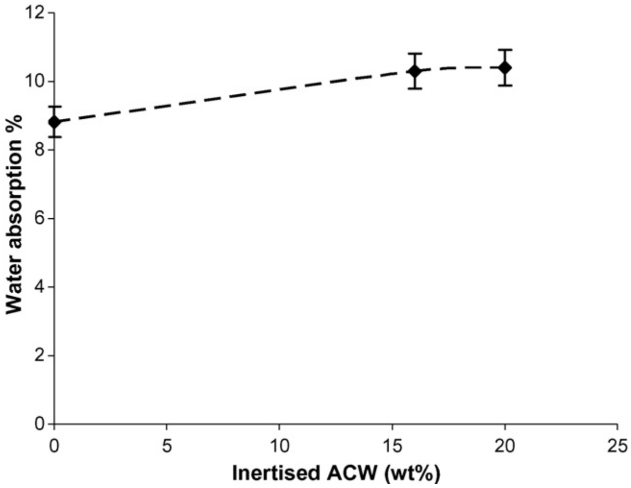

Fig. 2. Water absorption curves of the mullite–cordierite refractory composition with addition of 16 and 20 wt.% of inertised ACW (line is drawn for eye help only).

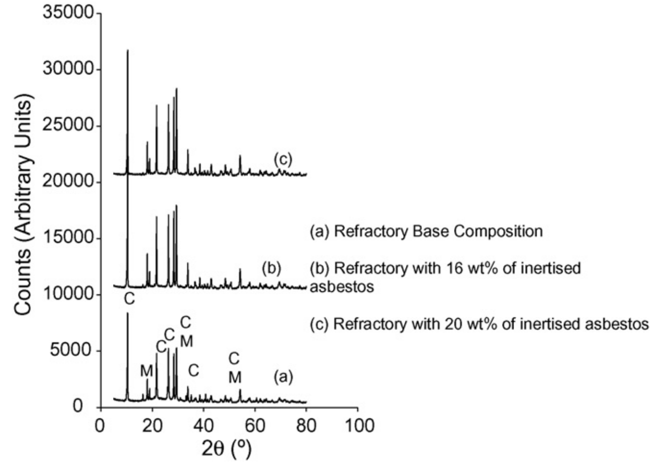

Fig. 4. XRD patterns of the mullite–cordierite refractory base composition and for compositions with 16 and 20 wt.% of inertised ACW, collected in the same 2θ range at room temperature, showing mullite (M) and cordierite (C) as main crystalline phases.

the refractory plates, which could potentially lead to improved thermal shock resistance. However, high porosity can also lead to lower resistance to chemical attack and low fracture strength of the materials. These facts should be taken into account considering future applications of these materials in the refractory industry.

The results of the linear shrinkage measurements as function of specimen composition is plotted in Fig. 3 with error bars showing minimal and maximal measured values. The permanent linear shrinkage was determined by measuring the relative diameter variation of the cylindrical specimens before and after the thermal treatment using Vernier callipers. A significant decrease of linear shrinkage values with increasing weight percentage of inertised asbestos was found which should lead to a better thermal shock behaviour.

Fig. 4 reports the XRD patterns of the mullite–cordierite refractory base composition and of samples with addition of 16 and 20 wt.% of inertised asbestos. As expected, mullite (M) and cordierite (C) are the main crystalline phases present. As observed in Fig. 4, no significant variation of the main phases present occurs with the change of raw materials. These results demonstrate that addition of inertised asbestos instead of talc as source of MgO has no effect on the final mullite–cordierite crystalline structure, at least under the detection capability of XRD.

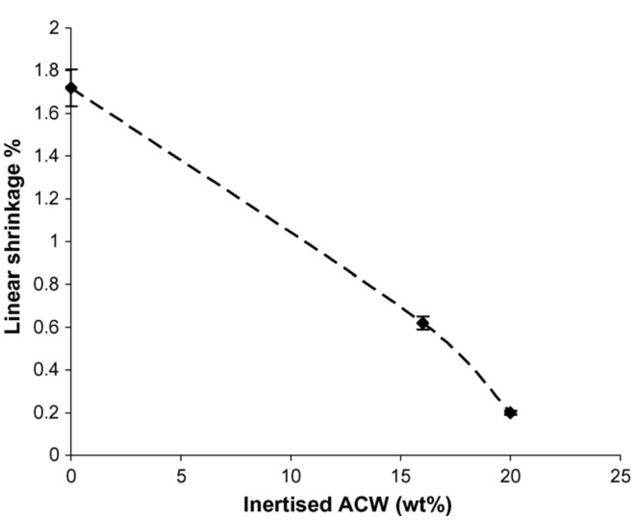

Fig. 3. Shrinkage curves of the mullite–cordierite refractory compositions with increasing wt.% of inertised ACW added (line is drawn for eye help only).

It was also found that in the cordierite–mullite refractory samples, the replacement of a Mg-rich talc by the inertised asbestos as raw material did not introduce significant variations of the Young's modulus, as shown in Fig. 5. Moreover, Fig. 6 shows the ratio of the cordierite peak intensities (counts) of samples containing 16 and 20 wt.% inertised asbestos related to those of the refractory base composition taken from the XRD patterns in Fig. 4. This figure reveals that the amount of cordierite phase in the specimens is higher when inertised asbestos is used as raw material in place of the Mg-rich talc. This replacement was performed maintaining the overall oxide percentages close to those of the refractory base composition (see Table 2). The presence of a mineral, forsterite, with higher MgO content (42 wt.%) with respect to the commonly used one, Mg-rich

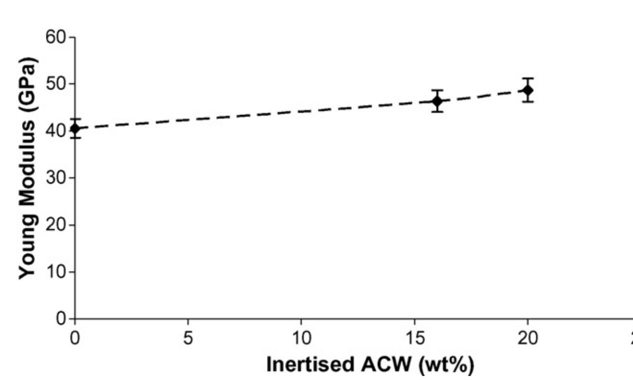

Fig. 5. Young's modulus variation with increasing addition of inertised ACW for the mullite–cordierite based refractory samples (line is drawn for eye help only).

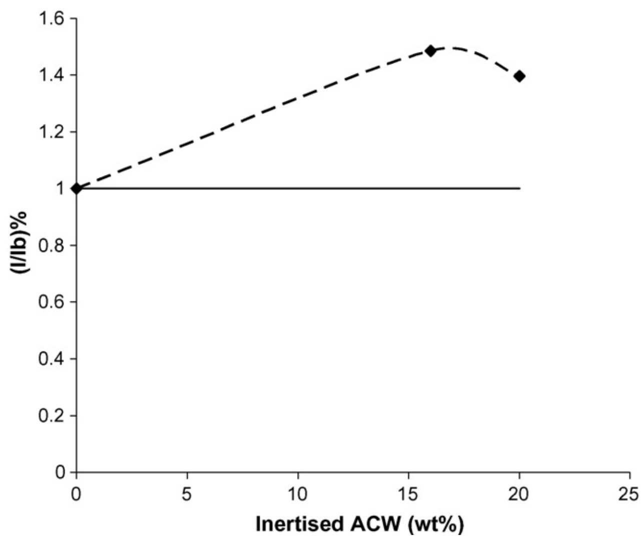

Fig. 6. Relative peak intensity ratio of the cordierite phase in samples made with microwave inertised ACW addition in relation to the base commercial composition in the mullite–cordierite system (line is drawn for eye help only).

talc (nominal 32.11 wt.%) may help cordieritic phase formation. This finding can be considered a significant technological result with potential economic implications, demonstrating that inertised asbestos represents a more convenient cordierite source raw material than the cost-intensive talc usually employed in the refractories industry.

4. Conclusion

In the case of mullite–cordierite refractories, inertised asbestos could be used as raw material to replace the currently used talc. Considering approximately equal oxide composition of the raw materials employed for sample preparation, the use of inertised asbestos led to a higher concentration of the cordierite phase. There was no significant variation of the Young's modulus of samples made with inertised asbestos compared to the commercial based composition, indicating that refractory materials containing microwave inertised asbestos should behave similarly to commercial refractories in terms of their mechanical properties.

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