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B. Joliet, F. Cambier, L. Dapra, C. Leblud, Anne Leriche. MICROSTRUCTURAL AND MECHANI-CAL CHARACTERISATION OF MULLITE-ZIRCONIA COMPOSITES CONTAINING YTTRIA. Journal de Physique Colloques, 1986, 47 (C1), pp.C1-723-C1-728. 10.1051/jphyscol:19861109 . jpa-00225504

HAL Id: jpa-00225504 https://hal.science/jpa-00225504

Submitted on 4 Feb 2008

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MICROSTRUCTURAL AND MECHANICAL CHARACTERISATION OF MULLITE-ZIRCONIA COMPOSITES CONTAINING YTTRIA

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<u>Résumé</u> - Des composites mullite-zircone contenant de O à 10 % en masse d'oxyde d'yttrium ont été synthétisés par frittage réactif. L'augmentation de la ténacité de ces matériaux vis-à-vis des mullites classiques est discutée sur base de la microstructure et des propriétés mécaniques.

<u>Abstract</u> - Mullite-zirconia composites containing 0 to 10 W/o yttria have been prepared by reaction sintering process. The toughness enhancement of these materials in comparison with classical mullite is discussed taking into account their microstructure and their mechanical properties.

I - Introduction

It is well known that fine zirconia particles dispersion enhances the mechanical properties of ceramic materials. Several mechanisms have been identified according to the presence or not of tetragonal ZrO_2 : transformation toughening, microcracking, crack branching or deviation, grain boundary strengthening, ... In mullite-zirconia composites, reaction sintering from alumina-zircon mixtures allows us to carry out both densification of the material and dispersion of zirconia particles. However, the tetragonal zirconia, about 8 volume percent of the material). Despite several fundamental studies (1, 2, 3), the mechanisms of reinforcement are not clearly established for this kind of zirconia ceramics. The tetragonal zirconia content can be increased, for instance by adding a stabilizer (yttria) in order to favour one particular mechanism : transformation toughening.

In this paper the mechanical and microstructural properties of mullite-zirconia materials containing increasing amount of yttria (MZY composites) are described and discussed in relation to toughening mechanisms.

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II - Experimental

Alumina and zircon powders ^{*} were mixed in water in the 3 alumina-2 zircon molar ratio. 0 to 10 Wt percent of yttria ^{*} as tetragonal zirconia stabilizer, 1.8 Wt percent of magnesia ^{*} as sintering aid and polyethylene glycol (4 Wt/o) as binder were added to the mixture. The slurry was spraydried and the agglomerates were diepressed to obtain 150 x 25 x 10 mm² green samples. After debonding, the materials were fired in air at 1500° C during 15 to 240 min. The materials were analysed by XRD in order to determine the nature and the content of the different phases (method of internal standard : NaF). The microstructural characterisation was carried out by microprobe analysis for identification of the different grains and by dimensions measurement on pictures obtained by SEM according to methods described elsewhere(3). For this purpose the samples were polished and chemically etched. Elastic modulus (E) was measured by Grindosonic method (natural vibration). The microstructural from Vickers indentation according to Evans equation (4).

III - Results and discussion

It has been stated(8) that the presence of yttria favours the dissociation of zircon and consequently the formation of zirconia. However, as it can be seen in figure 1 (which presents the mullite and alumina contents as a function of yttria), the amount of mullite decreases when the yttria content is higher than 3 W/o. It is probable that in this system the boundary between the secondary crystallisation field of mullite and this of alumina is located close to 3 W/o Y_2O_3 .



Fig 1. : Mullite and alumina contents versus Y203 for various firing times

The total tetragonal zirconia content (Qr tot) was measured on fired samples (without any mechanical treatment). The chemically stabilized tetragonal zirconia content (Qr stab) was measured on ground materials. The two values (Qr tot and Qr stab) were calculated from the Porter and Heuer's relation (5). The tetragonal zirconia ratio able to transform was obtained by the difference Qr tot - Qr stab = Qrtr and generally presents a maximal value for 5 W/o of yttria (120 min as example in table 1). The addition of yttria leads to tetragonal zirconia stabilisation by formation of a solid solution $ZrO_2-Y_2O_3$ and a complete stabilisation is obtained for 10 W/o of yttria.



Picture 1 : 0 W/o $\dot{Y}_2 0_3$ ZS means non dissociated zircon

Picture 2 : 1 W/o Y_2O_3



Picture 3 : 2 W/o Y203



Picture 4 : 10 W/o Y203

Picture 5 : 10 W/o Y203



<u>Fig. 2</u> : mean diameter of each $Zr0_2$ class versus Y_20_3 content



<u>Fig. 3</u> : Mean $\rm K_{IC}$ and Qr tr values versus $\rm Y_{2}O_{3}$ W/o

Table I : Tetragonal zirconia mechanical properties and open porosity of MZY composites fired during 120 min. at 1500° C.

Y2 ⁰ 3 (W/o)	Qr st (%)	Qr tr (%)	E (GPa)	H _v (GPa)	K _{IC} (MPa √m)	P 0 (W/o)
0	3	9	150	6.0	4.7	2.4
0.5	9	4	123	5.4	4.1	5.5
1.0	9	0	119	5.2	3.8	5.6
2.0	8	4	127	5.6	4.0	3.4
3.0	7	3	116	4.5	3.5	6.0
5.0	24	40	153	6.0	3.8	0.1
10.0	97	2	163	6.0	2.5	0.1

Pictures 1, 2 and 3 present microstructural aspect of mullite-zirconia composites for respectively 0,1 and 2 W/o Y_2O_z .

Pictures reveal a composite micróstructure characterised by acicular crosslinked mullite grains (M). White zirconia grains are dispersed inside mullite grains (A) and in the boundaries (B). A third class of zirconia grains (C) partly located inside the mullite and in the boundary grains can be noticed. The small white yttria grains (~ 0.2 μ m-see picture 3) are agglomerated as chains around intergranular zirconia grains. These intergranular ZrO₂-Y₂O₃ zones become larger as the yttria content increases and lead for 10 W/o YO₂ to a completely heterogeneous and porous microstructure (pictures 4 and 5).

For each class of zirconia grains, a mean diameter was calculated from the distributions in number. The data are presented versus yttria content in figure 2. This figure shows that the size of intragranular zirconia is smaller than this of other grain classes and is the same for each yttria content, whereas boundary and intergranular grain sizes decrease for increasing amount of Y_2O_3 . This can be explained on one hand by the slow diffusion rate of species through crystalline mullite (A type zirconia grains) explaining their insensible behaviour, and on the other hand by the fact that yttria particles agglomerate around B and C type zirconia particles (picture 3) leading to the formation of a diffusion barrier making slower the zirconia crystal growth during the firing.

Mechanical properties (E, Hv, K_{IC}) and open porosity (Po) of MZY composites are also reported in table I (120 min of firing).

As shown by the table, E and Hv present a minimum value for 3 w/o Y_2O_3 which corresponds to a maximum open porosity and a maximum mullite content (figure 1). This behaviour is probably related to heterogeneities in the powder mixtures appearing when Y_2O_3 is added and leading to pore formation when reaction occurs (7). K_{IC} and Qr tr mean values (calculated from data obtained for all the firing times) are presented as a function of Y_2O_3 content at figure 3.

 $\rm K_{IC}$ values obtained for MZY composites are smaller (~4.0 MPa /m) than those obtained without $\rm Y_2O_3$ (~ 4.5 MPa /m). They are in the same range from 0.5 to 5.0 W/o $\rm Y_2O_3$ and decrease drastically for 10 W/o $\rm Y_2O_3$ (2.2 MPa /m). In comparison with classical mullite toughness (1.8 MPa /m) (6) it is obvious that reinforcement occurs in MZY composites (to 5 W/o $\rm Y_2O_3$), however transformation toughening cannot be considered as the main mechanism: indeed mean Qr tr varies from ~ 10 % to a maximum value of ~30 % for 5 W/o without any increase of $\rm K_{IC}$ (fig. 3). Two hypothesis can be proposed : the first one is related to other zirconia toughening mechanisms, the influence of which being balanced as a function of $\rm Y_2O_3$ addition ; the second one is an improvement of fracture energy in reaction sintered mullite-zirconia composites due to their particular microstructure (acicular crosslinked mullite grains) : indeed this microstructure becomes heterogeneous when 10 W/o $\rm Y_2O_3$ is added (see pictures 4 and 5).

IV - Conclusion

 $Y_2 0_3$ addition in mullite-zirconia composites allows us to increase the amount mechanically transformable tetragonal zirconia. However $K_{\rm IC}$ values remain almost constant to 5 W/o $Y_2 0_3$ and then decrease excluding a main influence of transformation toughening mechanism. Incidence of other mechanisms or of particular microstructure effects cannot be differenciated due to microstructural modification and absence of mechanically transformable t-Zr0_ occuring simultaneously in 10 W/o yttria MZY composites.

ACKNOWLEDGEMENT

This paper is based on 2 B. Sc. Thesis presented at the University of Mons -Belgium (A. Biernaux and B. Joliet). One of us (A.L.) thanks IRSIA for a bursary.

REFERENCES

/1/ J.S. WALLACE - Gefügeaufbau und Buchwiderstand von in situ-reagiertem

Mullit-ZrO₂. Ph. D. Thesis Stuttgart - 1983. /2/ M.I. OSENDI MIRANDA - Materiales Ceramicos Tenaces Compuestos De Mullita-Circona - Ph. D. Thesis - Madrid - 1984.

/3/ A. LERICHE - Caractérisation Microstructurale de Composites Mullite-Zircone /3/ A. LENTCHL - Caracterisation Microsofracturate de Composites Mainte-Zirca obtenus par Frittage Réactif - Ph. D. Thesis - Mons - 1985.
/4/ A. EVANS and E. CHARLES - J. Am. Ceram. Soc. <u>59</u> (7-8) 371-372 (1976).
/5/ D.L. PORTER and A.H. HEUER - J. Am. Ceram. Soc. <u>62</u> (5-6) 298-305 (1979).
/6/ T. MAH and K.S. MAZ DIYASNI. J. Am. Ceram. Soc. <u>66</u> 699-703 (1983).
/7/ F. CAMBIER, C. BAUDIN, P. PILATE and A. LERICHE, Br. Ceram. Trans. J. <u>83</u>, 106 200 4004

JOURNAL DE PHYSIQUE

196-200, 1984.

/8/ B. JOLIET - B. Sc. Thesis - Mons University, Belgium (1984).