

Structural evolution of ZTA composites during synthesis and processing

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Abstract

We have studied the structural properties of industrial Zirconia Toughened Alumina (ZTA) composites for different compositions of zirconia and yttria at the main stages of the process. Influences on phases stability, lattice parameters, homogeneous and non-homogeneous strains, particles and crystallites sizes, have been considered and discussed. The structural states are explained from a very delicate balance of three competing effects: yttria-doping, strain and size effect. The first and third one have been shown to stabilize the tetragonal phase relatively to the monoclinic phase, and to destabilize the tetragonal phase relatively to the cubic phase whereas the second one is doing the opposite. In addition to this competition, the most prominent local effects have also been considered to fully understand the microscopic situation: i.e. the micro-strains and the inhomogeneous distribution of yttria which both increases the metastable coexistence of phase. At high yttria content a core-shell structure of the grains is evidenced.

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1. Introduction

The need for biocompatible materials with ultra-high performances and sufficient lifetimes to avoid further medical intervention is becoming tremendous. In particular ceramic–ceramic hip implants have been introduced in orthopaedics since they show decreased wear rates and they lower the risk of aseptic loosening. They are today considered as the most promising material to improve implant lifetime. This is why most of research and industrial development are now focused on ZTA. The use of zirconia particles inside alumina leads indeed to a toughening of alumina, while preserving a priori a better stability *in vivo* when compared to zirconia alone.^{1–4} The remarkable mechanical properties of these composites are associated to the metastable retention of the high temperature

tetragonal structure of zirconia down to room temperature. Indeed in standard conditions zirconia exhibits a sequence of phase transformations from room temperature to high temperature: the room temperature monoclinic phase (m) is stable up-to 1170 °C and transforms into a tetragonal phase (t) which remains stable until 2370 °C, temperature above which the zirconia is in (c) cubic phase. Upon cooling the inverse transformations take place. The t–m transformation induces a substantial increase in volume (about 4%) and consequently dramatic failures occur.⁵ Stabilization of the t-ZrO₂ at room temperature allows toughening by phase transformation.⁶ Indeed if any external stress is applied, this metastable phase returns to the stable monoclinic phase, and opposes via its survolume to the cracks propagation. This phenomenon is the origin of the outstanding mechanical properties of zirconia. Several ways (which may compete) exist to stabilize the tetragonal zirconia down to room temperature: chemical and doping composition (e.g. MgO, CaO, Y₂O₃, CeO₂, or some rare-earth), grain size below a critical size R_c ,^{7,8} strains effect due to alumina matrix, etc. There is however a delicate balance between these different effects which must be precisely known. Indeed the tetragonal phase must be stabilized, but in

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such a way that the t–m transition can happen when mechanical solicitations happen: if the t-ZrO₂ is too highly stabilized, this will suppress the cracks-induced phase transformation and the prevention of cracks propagation.

The stability conditions of the different phases of ZrO₂ in monolithic zirconia and in ZTA composites, in particular the t-ZrO₂ have been the subject of numerous studies, see for instance references,^{6,9–11} but most of the fundamental studies show different results depending on process conditions. In particular the mechanical properties and the ageing resistance of these composites are also strongly sample-dependent due to different routes of elaboration leading to different microstructure and different structural properties of the composites. Indeed the ZTA production includes many steps of heat treatments, milling, spray-drying, shaping, sintering etc, all those steps affecting the stability of t-ZrO₂ and the final mechanical properties. Therefore this paper aims to study step-by-step the influence of the production process on the structural properties of ZTA composites massively produced at industrial level, paying special attention to the stability of the t-ZrO₂. In a further publication, correlation of the stability of the t-ZrO₂ with the mechanical properties, as well as the influence of ageing and thermal evolution will be reported. In particular we will use the result of the present study to demonstrate that the toughening by phase transformation operates in the most efficient way when the amount of transformable tetragonal phase is maximum: if the expected mechanical properties are high bending strength and fracture toughness, high zirconia content will be preferred. On the contrary, if a high hardness is expected, the zirconia content will be limited. The present study aims in particular at evidencing the role played by the composition in yttrium and zirconia on the t-ZrO₂ stability.

2. Experimental procedures

2.1. Synthesis of materials and microstructures

The alumina and zirconia raw powders were produced by the Nanoe company (France) via an industrial process close to a method of co-precipitation at low temperature. Those raw powders were then mechanically milled in aqueous media. The alumina was doped with 1000 ppm of MgO as a sintering additive. Zirconia with $\times$ content of Yttrium (Z $\times$ Y) ranging between 0% and 8 mol% was milled at high rates to form composites containing 2.5–50 wt% of zirconia. Hereafter we use the conventional notation: e.g. A50Z8Y is a composite with 50 wt% of zirconia doped with 8 mol% of yttrium; also wt% and mol% is abbreviated in %. The spray-dried granules were uniaxially pressed (320 MPa) to obtain pellets with 20 mm diameter and 2 mm thickness. The pellets were sintered at 1500 °C during 7 h. In order to analyze the microstructures of the sintered composites, the pellets were mirror-polished with diamond paste until 1 μ m, thermally etched at 1450 °C for 1 h and gold-coated to avoid charging effects. The analyses were carried out using scanning electronic microscopy (SEM) Leo Gemini 1530.

2.2. X-ray and neutron diffraction

Room temperature high resolution X-ray diffraction patterns using Cu K α radiation were collected on a highly accurate two-axis diffractometer with an 18 kW copper Rigaku rotating anode generator, using the Bragg Brentano geometry. Neutron powder diffraction patterns were collected at room temperature on the 3T2 high-resolution diffractometer on a thermal source at 1.225 Å by using the Orphée reactor facilities at Laboratoire Léon Brillouin at Saclay (France). Rietveld refinements of XRD and neutron diffraction data have been performed. This method uses the full diffraction patterns (i.e. all diffraction peaks observed at the corresponding wavelength, typically between $2\theta = 5^\circ$ and 150°) and allows a precise and quantitative refinement of not only the volumic proportion of different phases that could be observed in a given compounds (cubic, tetragonal, monoclinic) but also complete sets of structural data (atomic positions and thermal parameters), lattice parameters, even though we show in this paper only a few part of these sets. Although it is a rather lengthy method, its precision is far better than working only with a reduce set of Bragg peaks, its advantages in particular in these materials are also described in references.^{12,13} These refinements were performed using Jana2006 package.¹⁴ All patterns could be satisfactory fitted using a combination of one, two or three among the monoclinic, tetragonal and cubic phases, whatever the composition is. In particular we did not find any trace of possible transformation of the tetragonal phase into other tetragonal phases with different tetragonality or coexistence. Indeed in the literature^{12,15–17} under some specific ageing or heating treatments the so-called t'-ZrO₂ of the Y-rich compound can eventually transform into a t-ZrO₂ corresponding to a Y-lean part of the samples and an untransformable t''-ZrO₂ corresponding to at Y-rich part of the sample, with possible modulated microstructure, and possible phase coexistences. We observed in our Y-rich sample only one tetragonal phase (that according to the literature we should have named t' but as there is no ambiguity we will call t-ZrO₂), probably because we were not in situation to create the other tetragonal phases.

2.3. Spontaneous, homogeneous and inhomogeneous strains

Several types of strains have been considered in this study. First of all, as zirconia undergoes a phase transition from its high temperature prototypal cubic phase, a so-called spontaneous strain appears. For instance the tetragonal phase is characterized by a spontaneous strain which arises from the distortion of the cubic phase occurring below the critical temperature.

Other strains may appear during the different steps of the synthesis of ZTA composite, as lattice parameters of a given phase may change; they superimpose on spontaneous strains. These strains are often called “homogeneous strains” or “macro-strains”. Comparison of lattice parameters before and after a given step of the process gives a measurement of the macro-strains appeared during this step.

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