



# Application of the statistical Taguchi method to optimize X-SiAlON and mullite formation in composite powders prepared by the SRN process

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## Abstract

The formation of an X-sialon/mullite nanocomposite by a combination of mechanical activation and silicothermal reduction and nitridation is reported. The starting materials (andalusite, Si and various aluminium sources) were activated in a planetary mill and fired in N<sub>2</sub> at 1450 °C. Microstructure and phase analysis was carried out on fired and unfired samples using X-ray diffraction (XRD), scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS). Taguchi statistical experiments were carried out to optimize the formation and crystallinity of the X-sialon component of the sialon–mullite composite. The milling time is the factor of greatest influence on the formation and crystallite size of X-sialon; milling for 15 h increases the amount of X-sialon formed but longer milling times decrease the crystallinity of the products and the sialon/mullite ratio. High-energy milling of an andalusite/Si/alumina mixture produces a mullite-sialon nanocomposite powder. TiO<sub>2</sub> additive stabilises the mullite component whereas Y<sub>2</sub>O<sub>3</sub> enhances the formation of X-sialon and Al(OH)<sub>3</sub> increases the crystallinity. The optimized conditions for the production of X-sialon/mullite nanocomposite powders are the use of Al<sub>2</sub>O<sub>3</sub> as the Al source, milling time 15 h, avoiding the use of TiO<sub>2</sub> as an additive.

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## 1. Introduction

Ceramic matrix nanocomposites have attracted a great deal of attention from researchers due to their numerous technological applications. There are two general categories of nanocomposites, one type containing only nanometer sized grains and the other consisting of nanosized particles distributed with micron-sized grains [1]. Various techniques have been used to prepare nanocomposite powders, including mechanical mixing of powders, mechanosynthesis, vapour deposition, chemical

vapour condensation, hydrothermal precipitation, sol–gel methods and spray pyrolysis [2].

Oxide-nonoxide ceramic composites are of considerable interest nowadays. Previously reported nanocomposites include Si<sub>3</sub>N<sub>4</sub>–mullite–Al<sub>2</sub>O<sub>3</sub> nanocomposites synthesized by a reaction sintering method [1], mullite–SiC nanocomposites fabricated by a sol–gel route [3] and a mullite–sialon–alumina composite prepared by an infiltration process [4].

Sialons are phases in the Si–Al–O–N system and have excellent properties such as high hardness and mechanical strengths at low and high temperatures, low thermal expansion coefficients, good resistance to wear, and chemical resistance in a number of corrosive environments. One of their least favourable properties is their resistance to oxidation. X-sialon is a sialon phase with a high oxygen content; its synthesis by a

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reduction reaction does not therefore require a strong reducing agent such as carbon. The most popular route to synthesize X-sialon is by silicothermal reduction and nitridation (SRN) of aluminosilicates [5–8]. SRN is a one-step method to produce an X-sialon body [6]. SRN of an aluminosilicate which follows four separate steps; decomposition of the aluminosilicate to form mullite and amorphous silica, nitridation of the elemental Si, reaction between the aluminium source and the amorphous silica and finally the formation of a solid solution of  $\text{Si}_3\text{N}_4$  and mullite to form X-sialon. In the first step, the formation of a gas-impermeable product layer around the partially reacted Si grains causes the nitridation reaction to become markedly slower and occur in two steps. In the presence of  $\text{Y}_2\text{O}_3$ , the nitridation reactions proceed faster and in one-step. In the final step, the presence of  $\text{Y}_2\text{O}_3$  causes the mullite to be removed at lower temperatures with the formation of X-sialon. Thus, the action of  $\text{Y}_2\text{O}_3$  is to increase the reaction rate without significantly changing the reaction sequence [5]. The effect of 1 wt% of other additives such as MgO, CaO, BaO,  $\text{Fe}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{ZrO}_2$  and  $\text{CeO}_2$  on the SRN process has been investigated. It is reported that the first step (nitridation of the Si) is facilitated by all above oxides but especially by  $\text{Fe}_2\text{O}_3$  and BaO. The second step (mullite formation) is promoted by  $\text{Y}_2\text{O}_3$  and  $\text{ZrO}_2$  but delayed by CaO and MgO. The fourth step (X-sialon formation) is most marked in the presence of  $\text{Y}_2\text{O}_3$ , CaO and MgO, but is suppressed by  $\text{Fe}_2\text{O}_3$ , which enters and stabilises the mullite structure. The beneficial effects of  $\text{Y}_2\text{O}_3$  and MgO are related to their formation of liquid phases at the reaction temperature. The most favourable additives for promoting both X-sialon formation and densification are  $\text{Y}_2\text{O}_3$ , CaO and  $\text{CeO}_2$  followed by MgO [6]. The trends in the formation of mullite and subsequent X-sialon are essentially independent of the additive concentration (1 and 10 wt% MgO,  $\text{Y}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ ) whereas the density is increased by an increase in the additive content [7]. In other research, MacKenzie et al. compared  $\gamma\text{-Al}_2\text{O}_3$  with  $\text{Al}(\text{OH})_3$  as the Al source in the SRN process under mechanochemical activation, showing that substitution of  $\gamma\text{-Al}_2\text{O}_3$  by  $\text{Al}(\text{OH})_3$  facilitates the production of Al–O–Si bonds during grinding and increases the degree of crystallinity, whereas ground samples containing  $\gamma\text{-Al}_2\text{O}_3$  showed a greater tendency to convert to X phase sialon [9].

Mullite is an attractive oxide ceramic because of its high melting point (1850 °C), good mechanical properties, low thermal conductivity, low thermal expansion and good chemical stability. The influence of additives on the formation of mullite from aluminosilicates has previously been reported [10–13]. Since mullite formation is an inevitable step in the SRN process [5] and the crystal structures of mullite and X-sialon are similar [14], the relative amounts and morphology of these phases in powders synthesized by the SRN process could be an important factor in determining the ultimate properties of composite powders. X-sialon formation is also governed by the starting materials, preparation technique and firing temperature, and the additional formation of other phases such as  $\text{Si}_3\text{N}_4$ , mullite, and O-sialon have been reported [5–7].

The present research investigated the formation of X-sialon containing nanocomposite powders. To achieve nanosized

particles, the raw materials were ground in a planetary mill, then subjected to silicothermal reduction and nitridation to obtain X-sialon. The ease of synthesis of the resulting X-sialon containing composites and the influence of different synthesis variables was optimized by a series of experiments based on Taguchi methodology.

## 2. Material and methods

The starting materials were 50–350  $\mu\text{m}$  andalusite (Kerphalite KF, Damrec, Paris, France), 63–200  $\mu\text{m}$   $\alpha\text{-alumina}$ , < 150  $\mu\text{m}$  aluminium hydroxide and  $\sim 40$   $\mu\text{m}$  Al powder (Merck, Frankfurt, Germany), < 10  $\mu\text{m}$  Si powder (Sicomill, Kema Nord Engineering Ceramics, Ljungaverk, Sweden),  $\sim 10$   $\mu\text{m}$   $\text{Y}_2\text{O}_3$  (Alfa Aesar, Johnson Matthey, Reacton<sup>®</sup>, London, England) and 21 nm  $\text{TiO}_2$  (Degussa<sup>®</sup> P<sub>25</sub>, Evonik, Essen, Germany).

The influence of different factors on the phase composition and crystallinity of the composite samples were assessed by designing an orthogonal experimental based on the Taguchi method with three factors each of the three levels as shown in Table 1. Powder blends were prepared from appropriate mixtures to provide the samples described in Table 2. The powder mixtures were milled in a planetary mill at 600 rpm with alumina balls ( $\varnothing = \sim 2$  mm) in a Teflon container with a balls-to-powder weight ratio of 20. To prevent oxidation the milling pot was filled with high-purity nitrogen gas and the mill was stopped and allowed to cool for 15 min after each 1 h of grinding. In samples 3, 6 and 9, in which the aluminium source was metallic Al powder, the mixture was cooled slowly under nitrogen after milling to prevent aluminothermic reactions. Upon reaching room temperature the Al reacted with  $\text{O}_2$  to form  $\text{Al}_2\text{O}_3$  without oxidation of Si (this was demonstrated by XRD).

The milled mixtures were fired in a horizontal laboratory tube furnace under flowing nitrogen (50 mL/min) at a heating rate of 10 °C/min to 1100 °C, and then at 2 °C/min to 1450 °C. The furnace was cooled immediately on reaching the maximum temperature.

The morphology and elemental compositions of the powders were determined by SEM/EDS (XL30, Philips, Amsterdam, Netherlands, operating at 17 kV in the secondary electron imaging mode). The powder samples were coated with Au using a BAL-TEC SCD005 sputter coater prior to imaging.

Transmission electron microscopy and selected area electron diffraction (TEM-SAED, Philips) was carried out at 100 kV.

XRD analysis of the samples was carried out using a D8 Advance spectrometer (Bruker, Karlsruhe, Germany) with Cu radiation ( $K\alpha = 1.540598$ ) and a Ni filter. The diffraction traces

Table 1  
Factors and levels for the experimental design.

Factors	Additive	Al Source	Milling time (h)
Levels	–	$\text{Al}(\text{OH})_3$	0
	$\text{Y}_2\text{O}_3$	$\alpha\text{-Al}_2\text{O}_3$	30
	$\text{TiO}_2$	Al	15

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