



Self-propagating high temperature synthesis of SiAlON



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ABSTRACT

In this work, a new method of SHS process has been investigated for the synthesis of SiAlON under low nitrogen pressure. A gel mixture of silica and sodium oxide (sodium silicate) as a reactant and catalyst was produced by alkali melting of silica, solving the results in water and heating in auto-clave. Al powder was also used as reactants. Urea and aluminum nitrate hydrate were used as igniting agents. The addition of Mg also was necessary for reaching the combustion. Sodium azide as a good reactant addition for preparing both combustion and nitrogen supply was tested. Beta-Si₃N₄ powder acted as diluting agent and by reducing the adiabatic temperature helped the nitrogen diffusion into the melted reactants which was not agglomerated. NH₄Cl in some cases was used as a catalyst which showed good results for reaching SiAlON. For comparison possibility some experiments were made using silica powder instead of sodium silicate gel. These materials were mixed by pestle and mortar and laid into the combustion chamber cavity without pressing. The compound was heated by electrical element until the ignition happened by gases which were produced from urea and aluminum nitrate hydrate. The pressure of nitrogen in combustion chamber was kept constant near 0.1 Mpa. The products of syntheses were of agglomerated one mostly O'-SiAlON & β and β'-SiAlON. Effects of NH₄Cl were examined by XRD results. TGA and DTA graphs showed the thermodynamics of reaction which were analyzed in some parts by XRD spectroscopy. In some cases nano material of SiAlON was produced which is demonstrated by XRD and FTIR spectroscopy.

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1. Introduction

Ceramic of silicon nitride has high temperature (>1000 °C) structural applications. It has high strength, good friction and oxidation properties, low density, and high erosion and chemical corrosion resistance [1,2]. SiAlON is a general name for a big family of ceramics based on silicon nitride [3,4]. It was discovered in 1970 and actively developed [1,4]. Silicon nitride ceramics are used in aerospace and automotive industry in vast. Also due to their good tribology and low friction coefficient properties and resistance to chemical corrosion, they can be used in cutting tools, wire die extension and blast nozzles [2,5–7]. By substitution of silicon–nitrogen bond for aluminum–oxygen one in β-silicon nitride, a solid solution is made which was called primarily β'-silicon nitride, β'-SiAlON and nowadays SiAlON. Each unit cell of β-SiAlON is made by two unit cell of silicon nitride and is represented by the formula Si_{6-z}Al_zO_zN_{8-z}

wherein z varies between 0 and 4.2 [2,4,8,9]. Comparing to silicon nitride and other SiAlON phases, O'-SiAlON has the best oxidation properties because of its high oxygen content. It can be written in the form of Si_{2-x}Al_xO_{1+x}N_{2-x} (0 ≤ x ≤ 0.4). Its oxidation properties are remarkable and has no oxidation before the 1200 °C [10,11].

The self-propagating high temperature synthesis method (SHS) has investigated the production of SiAlON by mixture of nano material gel of silicon oxide as reactant and sodium oxide as catalyst in the form of sodium silicate. Gels can be obtained from metal oxide sols. The major benefit of this method is high homogeneity and good purity [12]. Combustion synthesis (CS) or self-propagating high-temperature synthesis (SHS) is an effective, low-cost method for production of various industrial useful materials [13]. This method is discovered in USSR and can be used for nitrides and oxy nitride production in very fast process times and relatively low preheating temperatures. Very high reaction temperatures are produced and the powder components produce a large range of unique materials by controlled high-temperature combustion. Desired amount of metals nitrates + fuel is locally ignited. As a result a self-sustained reaction wave steadily propagates along the media forming nanometer powder of desired composition [14–16].

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Table 1
The reactants characteristics.

Row	Reagent	Particle size	Purity (%)	Source
1	Si	Micronized	99	China, Chanlian-Dacheng SS
2	SiO ₂	Micronized	99	China, Chanlian-Dacheng
3	Al	Micronized (30–90)	99%	Merck
4	Al(NO ₃) ₃ ·9H ₂ O	Gel	Reinst	Merck
5	H ₂ NCONH ₂	Gel	Reinst	Merck
6	NaN ₃	Micronized	99%	Merck
7	Mg	30–55 mesh	Purum	Fluka,Chemica
8	N ₂	–	99%	Local source
9	β-Si ₃ N ₄	325 mesh	99%	China, Chanlian-Dacheng
10	Na OH	Gel	Reinst	Merck
11	Potassium nitrate	Gel	Reinst	Merck
12	NH ₄ Cl	–	99,8	Merck

The characteristic feature of the temperature of the wave front is reaching quite high values (2000–4000 K) [17].

However, in self-propagating high temperature synthesis of SiAlON, the purity of the products has always been low because of low reaction of Si (it can be resulted from silica), which is because of the melting and subsequent agglomeration of Si during the self-propagating high temperature synthesis process with an extremely high reaction temperature and a very short reaction time. In order to decrease the combustion temperature as well as to slow down the reaction speed, diluents such as β-Si₃N₄ are added to the starting materials [18].

Adding the ammonium chloride to the reactants as catalyst has the priority of high reaction with silicon which produces silicon chloride and the later reacts with nitrogen to produce silicon nitride [19]. For the synthesis of nano material by SHS method, the metal salt of alkali melting is used. In this method the reducing metals like magnesium react with liquid melted alkali oxide and small metal particles are produced without agglomeration [17,20].

Using sodium azide as a solid nitrogen supply can improve the SHS process and help in adding amounts of stagnant nitrogen. However, it is also used as a catalyst for the growth of Si₃N₄ whiskers [13, 21–23]. Thermal reduction of oxides by magnesium will enhance the product yield and also enhance the self-propagating high temperature synthesis by heat generation of reduction reactions [14, 21].

Use of aluminum nitrate nona hydrate and urea is possible for the ignition that can cause high heat generation activated by an external element. As it is known nitrogen has the nominal valence of minus three in urea and plus five in aluminum nitrate that causes them reducing and oxidation agent respectively. In the heating process, the urea will dissociate to biuret and ammonia and in higher temperature to trimmer. Aluminum nitrate dissociates to alumina and nitrogen oxide. These gases will react and have in explosive exothermic reaction in controlled condition [24,25]. NH₄Cl is a good catalyst for converting the silica to silicon nitride by intermediate product of SiCl₄ [26].

Achieving a good corrosion and oxidation resistance properties via micro structure is an important goal in the synthesis and manufacturing of SiAlONs. For mass production of Ceramics with complex shapes, it is beneficial to use low cost precursor's combustion synthesized SiAlON powders. Besides, fabrication of SiAlON powders with low cost initial compound is possible. The purpose of this article is to study the current state in the field of fabrication of SiAlON by SHS of low cost powders. This article discusses properties of SHS powders with the general composition of SiAlON.

2. Experimental

2.1. Sample preparation

Specimens were prepared from commercial powders as precursor according to Table 1. The flow chart of process leading to production of SiAlON is shown in Fig. 1. A gel mixture of silica and sodium oxide as a reactant and catalyst was produced by alkali melting of silica, solving the results in water and heating in auto clave.

Al powder was used as reactant and β-Si₃N₄ powder was added as diluting agent in some experiments. Urea and aluminum nitrate hydrate were used as igniting agents. Also, the addition of Mg was necessary for reaching the combustion. Sodium azide is a good reactant addition for preparing both combustion and nitrogen supply which is used in some experiments. Using the ammonium chloride increased the product yield of nano properties.

The above mentioned powders were mixed completely by mortar and pestle in selected amounts according to Table 2, and laid into combustion chamber according to Fig. 2a. Nitrogen supply was injected to the combustion chamber by inlet valve from high pressure capsule regulated pressure. The schematic view of combustion chamber used in this investigation is shown in Fig. 3. It is made of an inner part (stainless steel cylinder and cap) which is also protected by an outer cylinder thick enough to protect the inner one. Using an internal ceramics electrical element (300 W, 220 V, 30 R CE), the mixed powder could be heated on top to ignition temperature rapidly. The nitrogen gas pressure could be controlled supplementary by an inner gage which was located on outlet valve on combustion chamber. The nitrogen gas was injected by inlet valve and flushed the air out and then the nitrogen had continuous flow in contact with the reactants. The temperature variation was noticed by an internal macro thermocouple type K Japan (0–1200 °C) made of Ni—Cr (diameter about 5 mm) connected to a monitor.

By starting the process this element heated the chamber one centigrade per second approximately and after reaching the combustion temperature, ignition happened thoroughly through the chamber and its contents. A sudden increasing of temperature happened in macro scale about 250 °C for the whole chamber mass which means high temperature in scale of micro. Control of nitrogen pressure was continuously made about 0.1 Mpa and by increasing the temperature, the pressure of gas products was controlled by outlet valve and nitrogen gas pressure while entering the chamber. At ignition point the temperature increased which caused sudden increment of pressure and flowing backward the nitrogen supply. By shutting down the electrical current, gas line and also blocking inlet gas, the combustion chamber was left to cool to room temperature wherein cooling happened about 10 °C per second. After reaching the room temperature, the chamber was opened and products were noticed to have a foamy shape and full of porosity according to Fig. 2b. These products had two layers; outer was harder than inner and was agglomerated. The products were easily separated from the cylinder and by using the mortar and pestle were milled. Experiments Series S1 and S2 were made in atmospheric furnace according to Table 2.

2.2. Characterization technique

For determining the unit cell lattice parameters and phase composition of the samples, X-ray powder diffraction patterns (X'Pert. P W 3710 Philips, Cu-K_α radiation) in the range of 10–90° (2θ) were collected. The crystallite size was calculated from FWHM of XRD patterns using Scherer equation. The TGA–DTA experiments were made by Mettler Toledo apparatus. FTIR were made by apparatus Nicolet 800.

3. Results and discussion

The resulted powders were determined by x-ray diffraction (XRD) analysis. The mechanism of heat generation leading to combustion

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