Conference Paper

Composite Ceramics Based on Garnet-type Oxide $Y_{2.5}Nd_{0.5}Al_5O_{12}$ and Silicon Carbide. Preparation. Properties

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Abstract

We have obtained powders of garnet-type complex oxide $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – x vol.% SiC (x = 0, 10, 20) using wet chemistry techniques. The ceramics based on the studying compounds were sintered using Spark Plasma Sintering (SPS) ($T_{sintering}$ = 1320-1350 °C, P = 80 MPa, $t_{sintering}$ = 7 min, $t_{shrinkage}$ = 2 min) with relative density ~98%. Microstructure of the obtained composites and influence of the concentration of silicon carbide on their mechanical (microhardness, fracture toughness) properties were investigated.

Keywords: Composites, Spark Plasma Sintering, microstructure, mechanical properties

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1. INTRODUCTION

Yttrium-aluminum garnet is a promising matrix for transmutation of minor actinides (MA) [1 – 5] due to the possibility of wide isomorphism (the structure can include actinide cations (including Pu (IV)) and REE [6 – 8]), and ceramics based on it has high hydrolytic and radiation stability [7 – 12].

The main drawbacks of ceramics based on mineral-like compounds (including those based on garnet) are: 1) low thermal conductivity, 2) low plasticity. This leads to the appearance of cracks and microcracks due to uneven heating of the material by radiogenic heat and a decrease in chemical stability. All these factors result in the damage to the fuel and limitations of fuel burn time-up.

To eliminate these drawbacks, additional components with high thermal conductivity and high ductility are introduced into the ceramic composition. The role of such a component can be performed by metals, oxides, carbides, metal nitrides, etc. Their choice is due to high melting temperatures and thermal conductivity, as well as a

low neutron capture cross section. Silicon carbide was used in the present work: the thermal conductivity coefficient λ (300 K) – 490 W m^{-1} K^{-1}, the fracture toughness coefficient K_{1C} – 3.5-4.5 MPa·$m^{1/2}$.

The goal of this paper are to obtain a CerCer composite based on garnet-type $Y_{2.5}Nd_{0.5}Al_5O_{12}$ oxide with SiC by Spark Plasma Sintering (SPS); to determine its microstructure and mechanical (microhardness, fracture toughness) properties.

2. MATERIALS AND TECHNIQUES

Garnet-type powder $Y_{2.5}Nd_{0.5}Al_5O_{12}$ was obtained by the coprecipitation method. Aqueous solutions of reactants ($Y(NO_3)_3$, $Nd(NO_3)_3$ and $Al(NO_3)_3$) were mixed with ammonium hydroxide NH_4OH (5% in water) to pH = 8 value. The prepared gel was dried at 90°C and baked at 300, 500, 800 and 1000°C for 10 h at each stage, followed by mechanical dispersion after each annealing treatment.

To obtain a CerCer composite, powders of garnet $Y_{2.5}Nd_{0.5}Al_5O_{12}$ and silicon carbide were ground in a Pulverisette 6 planetary ball mono mill for 2 h. To minimize abrasion contamination, tungsten carbid grinding balls (5 mm dia.) were used for grinding in isopropyl alcohol. Mass ratio powder:balls:alcohol was 2:4:1. Rotational speed was 300 rpm. After grinding, powder annealing was carried out for 8 h at 70 °C in a Nabertherm RHTC 80-230/15 circulating-air furnace.

Ceramics were obtained by SPS method using a Dr. Sinter model SPS-625 unit. Powders were loaded into a graphite compression mold of 10 mm int. dia. and heated by applying millisecond-long strong pulses (up to 3 kA). Temperature readings were taken with a Chino IR-AH infrared thermometer focused on the surface of the graphite compression mold. Sintering was carried out in a vacuum (6 Pa). The heating rate was 100 °C/min, and the applied uniaxial pressure was 80 MPa. Measurement accuracy was 10 C; pressure stability was 1 MPa.

Phase composition of powders and ceramics was studied by X-ray diffraction analysis (XRD). XRD readings were recorded on a Shimadzu LabX XRD-6000 unit (CuK_α-filter, λ = 1.54178 Å within the range of angles 2θ = 20–60°, increment 0.02°, spot exposure 0.6 s).

The density of the ceramic was measured by hydrostatic weighing in distilled water using Sartorius CPA scales. Microstructure of samples wase studied with a Jeol JSM-6495 scanning electron microscope (SEM)

Microhardness measurements of the ceramic samples were performed on a Struers Duramin-5 installation by the Vickers method. Fracture toughness coefficient (K_{1C}) was

determined based on the length of the radial cracks from the Vickers indenter. K_{1C} values were calculated by the Palmqvist method: $K_{1C} = 0.016(P/c^{3/2})(E/H_V)^{1/2}$, where P is the load on the indenter (g), c is the average distance from the imprint center to the tip of the crack, H_V is the Vickers hardness, and E is the elastic modulus of the material. Solving for fracture toughness coefficient, elasticity modulus E for all investigated materials was assumed to equal the elasticity modulus of yttrium-aluminum garnet (E = 300 GPa [13, 14]).

3. RESULTS AND DISCUSSION

The relative densities of composites with composition $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – x vol.% SiC (x = 10, 20%) obtained by SPS were ~ 98% from theoretical (4.60 g/cm^3 (10% SiC), 4.48 g/cm^3 (20% SiC)). Sintering conditions are presented in Table 1. From the composite sintering diagrams (Fig.1), it can be seen that the shrinkage of the samples occurs in the temperature ranges 1100-1300 ° (10% SiC) and 1120-1340 ° (20% SiC), maximum shrinkage rates (0.00675 mm/s – 10 % SiC, 0.00685 mm/s -20% SiC) are achieved at 1240 ° and 1270 °, respectively. The optimal sintering temperatures corresponding to the end of the active shrinkage stage are 1280-1310 °C (10% SiC) and 1310-1330 (20% SiC). The presence of two phases in the composites obtained was established by the XRD method (Fig.2): garnet (ICDD DataBase card no. 79-1891, sp. gr. Ia3d) and silicon carbide (ICDD DataBase card no. 74-1302, sp. gr. P63mc).

Table 1: Sintering parameters of composites based on garnet-type compounds.

Sintering parameters	$Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 10% SiC	$Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 20% SiC
P, MPa	80	80
$T_{sintering}$, max, °C	1320	1350
$t_{shrinkage}$, min	2	2
$t_{sintering}$, min	<7	<7

On the SEM image collected for each ceramic sample with the backscattered electron detector (Fig. 3), residual microporosity is observed. The pore sizes are <1 μm (for composite with 10% of SiC) and <0.2 μm (for composite with 20% of SiC). The grain size of the composites is 1.5-2.5 μm. By using a relatively high rate of heating and by avoiding exposure at the maximum sintering temperature, it was possible to prevent intensive growth of composite grains.

The microhardness of H_V and the fracture toughness coefficient K_{1C} were determined by the Vickers method. The load was 100 g, the loading time was 30 s. It

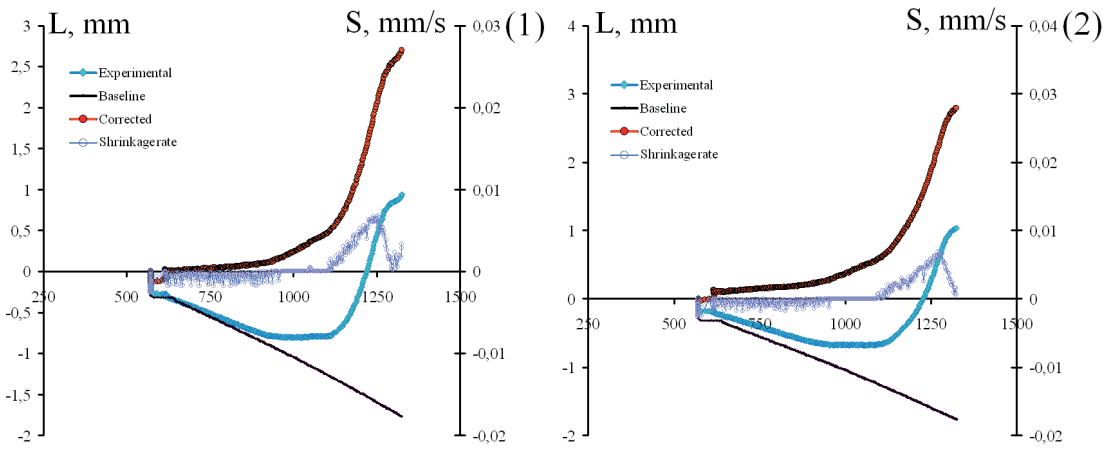

Figure 1: Graphs of shrinkage (L) and shrinkage rate (S) of $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 10% SiC (1), and $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 10% SiC (2) ceramic samples depending of sintering temperature: Experimental (shrinkage) – observed shrinkage; Baseline – the observed displacement represents overall characterization of shrinkage as it also includes contributions from the die and systems; Corrected (shrinkage) – valid shrinkage.

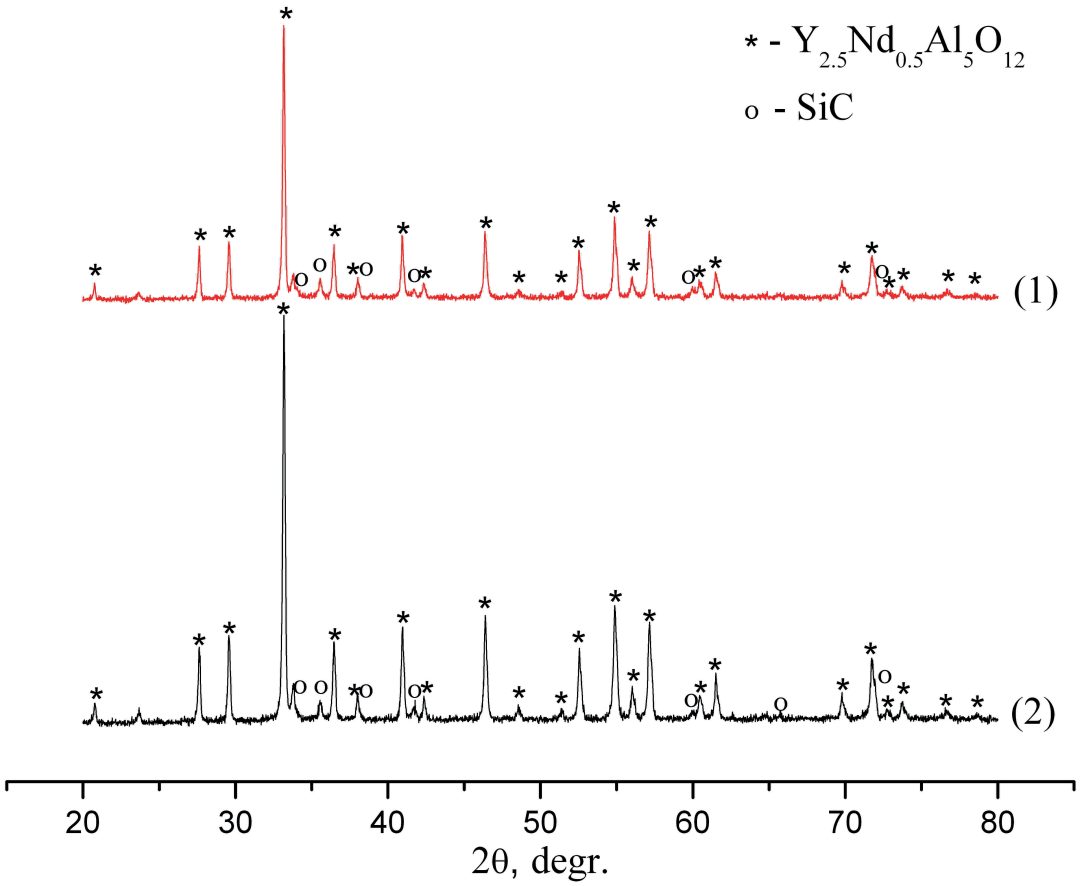

Figure 2: XRD patterns of composites based on $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 10% SiC (1), $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 20% SiC (2).

is established that H_V and K_{1C} are increased in comparison with ceramics based on $Y_{2.5}Nd_{0.5}Al_5O_{12}$ (H_V = 13.2 GPa, K_{1C} = 1.5 MPa·$m^{1/2}$) when silicon carbide is introduced into the composition of ceramics. In addition, as the concentration of silicon carbide

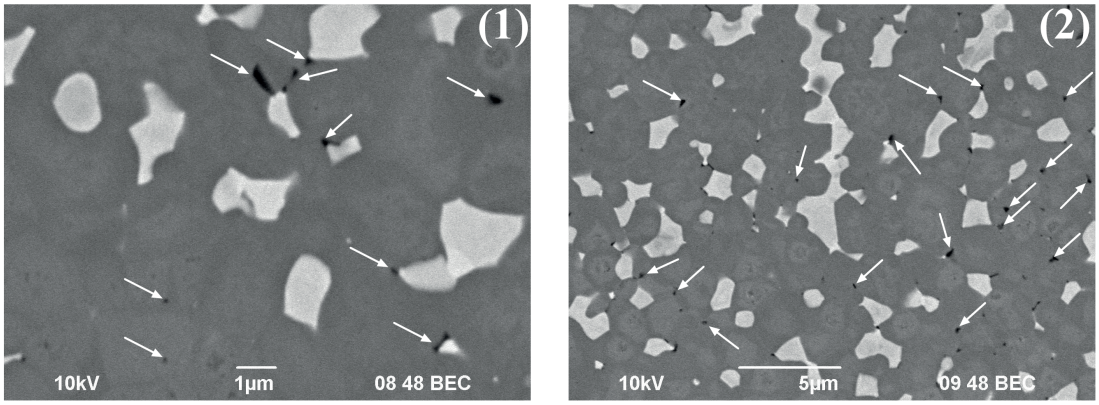

Figure 3: Backscattered scanning electron micrographs of ceramics based on $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 10% SiC (1), $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – 20% SiC (2). Light areas are silicon carbide. Pores are indicated by arrows.

increases, the fracture toughness coefficient increases in the composites (Table 2), which indirectly indicates an increase in their plasticity.

Table 2: Microhardness and fracture toughness coefficient of composites.

Sample	Microhardness H_V, GPa	Fracture toughness coefficient, MPa·m$^{1/2}$
$Y_{2.5}Nd_{0.5}Al_5O_{12}$-10% SiC	13.9	2.0
$Y_{2.5}Nd_{0.5}Al_5O_{12}$-20% SiC	13.9	2.3

4. CONCLUSION

Crystal matrices based on natural garnet minerals are promising IMFs, suitable for both transmutation of MA and direct disposal. Composites with composition $Y_{2.5}Nd_{0.5}Al_5O_{12}$ – x vol. % SiC (x = 10, 20) were characterized by XRD and SEM. Pellets with a relative density of ~98% were obtained by the SPS method at 1320-1350 °C and 80 MPa for <7 min. Mechanical properties (microhardness and fracture toughness) of the properties of composites have been studied. With an increase in the concentration of silicon carbide in composites, their fracture toughness coefficient increases. This may indicate an improvement in the plasticity of the ceramics.

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