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Thermo-optical properties of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ $(0 \leq x \leq 1)$ hexaaluminates for metallic thermal protection system

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$LaMg_{1-x}Ni_xAl_{11}O_{19}$ ($x=0$, 0.25, 0.5, 0.75, 1) ceramics are fabricated by pressureless-sintering method at 1700 °C for 10 h in air. The microstructure and thermo-optical properties of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics are investigated by the X-ray diffraction, scanning electron microscopy and Fourier transform infrared spectroscopy measurements. The influence of NiO doping on structure and thermo-optical properties of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics is investigated. The partial substitution of Ni^{2+} for Mg^{2+} results in a significant increase in emissivity at low wavelengths as compared with unmodified $LaMgAl_{11}O_{19}$. When the Ni^{2+} content increases to $x=0.75$ or above, $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics have a high emissivity value above 0.70 at low wavelengths at 500 °C. The measured emissivity of all $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics shows a similar trend in the wavelength range of 6 to 14 μm.

1. Introduction

The desire for higher-speed interceptor missiles, long-range fast-response strike weapons and reusable launch vehicles (RLVs) has ushered in a new, exciting era of hypersonic flight [1]. Temperature-resistant metallic materials are studied as hot structures and thermal protection systems (TPS) for hypersonic flight vehicles during ascent and reentry because of their inherent ductility, design flexibility and lower maintenance costs than competing systems [2,3]. However, metallic materials can be susceptible to damage in severe, high-temperature service conditions. When crossing the atmosphere at hypersonic speeds, aerodynamic heating produced by the passage of air over the vehicle is great and increases with its speed. Also, the recombination of oxygen and nitrogen atoms into their molecular form at the surface can result in an additional heat input to the TPS besides aerodynamic heating [4]. Since the thermal radiation energy is proportional to the fourth power of the temperature, the thermal radiative transfer effect is expected to increase rapidly with increasing operating temperature. Therefore, a high infrared emissivity (≥0.70 [4]) coating is desirable to reduce significantly the heat flux to the vehicle to maintain the metallic substrate within acceptable temperature limits.

According to the Plank blackbody function and Wien's displacement law, 95% of the radiation emitted by a blackbody at 1000 °C is below 14 μm and the largest radiation is below 3 μm. Unfortunately, common coating materials with excellent high-temperature mechanical properties and thermal stability, such as 6–8 wt.% Y_2O_3 partially stabilized ZrO_2 (6–8YSZ), have a low emissivity value below 5 μm wavelength at high temperatures [5]. Hexaaluminates have been developed as a potential candidate for laser crystals [6], luminescent materials [7], high-temperature combustion catalysts [8] and thermal barrier coating (TBC) materials [9–11]. Among these hexaaluminates, lanthanum magnesium hexaaluminate ($LaMgAl_{11}O_{19}$) is studied extensively. Consequently, $LaMgAl_{11}O_{19}$ shows a high thermal expansion, low thermal conductivity, excellent long-term sintering resistance and structural stability up to 1400 °C [12]. Such properties are controlled mainly by its magnetoplumbite structure consisting of alternative stacking of a $[Al_{11}O_{16}]^+$ spinel block in which Al^{3+} substituted partially by Mg^{2+} and a $[LaAlO_3]$ mirror plane [12–14]. However, to the best of our knowledge, thermo-optical properties (such as infrared emissivity) of hexaaluminates are not available in the open literature. The thermal radiation mechanisms of hexaaluminates still remain unclear.

In this paper, $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ($x=0$, 0.25, 0.5, 0.75, 1) ceramics were fabricated by the pressureless-sintering method at 1700 °C for 10 h in air. The microstructure and thermo-optical properties of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics were studied.

2. Experimental

In the present study, polycrystalline ceramic samples of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ($x=0$, 0.25, 0.5, 0.75, 1) were prepared by the conventional solid-state reaction technique using high-purity commercially available La_2O_3 (Grirem Advanced Materials Co. Ltd., China, purity ≥ 99.9%), NiO (Tianjin Fuchen Chemical Reagant Factory, Ltd.,

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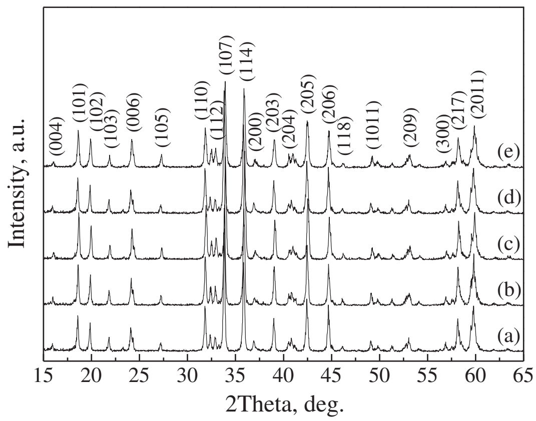

Fig. 1. XRD patterns of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics: (a) $x=0$, (b) $x=0.25$, (c) $x=0.5$, (d) $x=0.75$, (e) $x=1$.

China, Analytical pure), MgO and α-Al_2O_3 (Dalian Luming Nanometer Material Co. Ltd., China, purity≥99.99%) powders without further purification. All oxide powders were heat-treated at 900 °C for 2 h in air before further use. A stoichiometric mixture of these oxides was mechanically mixed for 24 h using zirconia balls and analytically pure alcohol. The mixed powders were dried and then uniaxially pressed into pellets at 20 MPa. The pellets were further compacted by cold isostatic pressing with a pressure of 400 MPa for 5 min, and then pressureless-sintered at 1700 °C for 10 h in air atmosphere.

The phase constituents of the as-sintered ceramics were determined by X-ray diffraction (XRD) analysis on an X-ray diffractometer (Rigaku D/Max 2200VPC, Japan) with Cu Kα radiation. XRD patterns were recorded in a scanning range of 10–70° at room temperature, and a scan rate of 4°/min was employed. The bulk density of cylindrical specimens was measured by the usual volume and weight measurement technique. Microstructures of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics were observed with scanning electron microscope (FEI Quanta 200F, The Netherlands). SEM specimens were prepared by polishing the surface to 1 μm finish, and then by thermally etching at 1550 °C for 1 h in air.

Fourier transform infrared spectrometer (JASCO FTIR-6100, Japan) was employed for the normal spectral emissivity and transmittance measurements. The resolution and scanning range were 4 and 4000–400 cm^{-1} (2.5–25 μm wavelength), respectively. The normal spectral emissivity is the ratio of the radiance of a given object to that of a blackbody at the same temperature level and for the same spectral and normal directional conditions. In emissivity measurements, the samples with dimensions of Φ30mm×2 mm were heated up to 500 °C by an electric heating furnace. The Landcal R1500T blackbody furnace (Land Instruments International Ltd., UK) was used as the near-blackbody source. For transmittance measurements, the solid

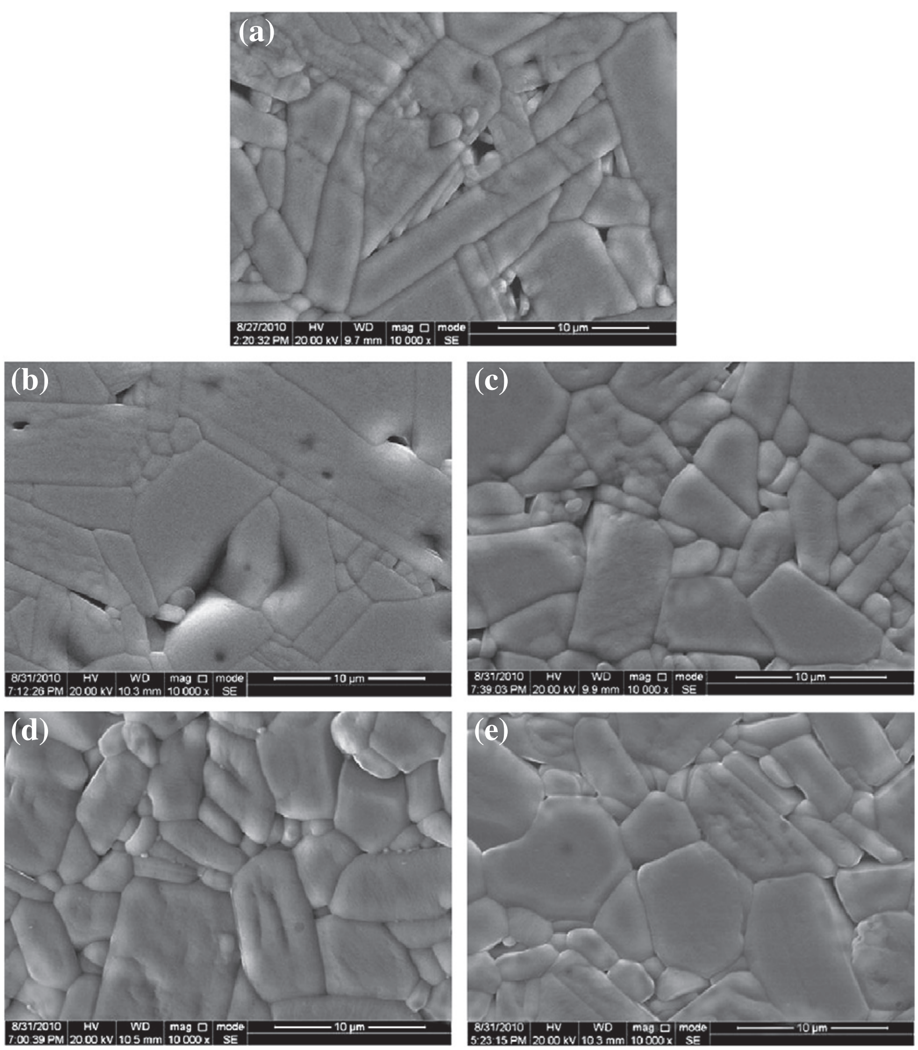

Fig. 2. Microstructures of $LaMg_{1-x}Ni_xAl_{11}O_{19}$ ceramics: (a) $x=0$, (b) $x=0.25$, (c) $x=0.5$, (d) $x=0.75$, (e) $x=1$.