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Recession behavior of Lu_2SiO_5 under a high speed steam jet at high temperatures

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Abstract

Study of recession behavior of Lu_2SiO_5 bulk was performed in high speed steam jet with a velocity of ~50 m/s temperature range between 1300 and 1500 °C for 100 h. X-ray results showed that no phase change was observed for all samples after steam exposure. Detailed scanning electron microscopy examinations showed some cracks formation was observed on the bulk surface for the samples of 1400 and 1500 °C. Also, porous structure was formed on the bulk surface for the samples of 1300 and 1400 °C. As for 1500 °C sample, the porous structure disappeared after exposure test. The high magnification images of 1300 °C sample showed the depletion of grain boundary glassy phase. However, for 1400 °C sample, boundary phase was formed again, and the grain growth can be identified for the sample of 1500 °C. The recession mechanism can be explained by a mass transfer of evaporated species from the bulk surface and the weight loss rate measured can be expressed by Arrhenius plot.

Keywords: Atmospheric corrosion; Ceramics; SEM

1. Introduction

Advanced silicon based non-oxide ceramics such as silicon nitride ceramics possess high mechanical properties and oxidation resistance at high temperatures [1,2]. However, it has been showed that the application of these materials for gas turbine hot-section components, corrosion and recession by combustion gas flow above 1300 °C becomes critical issue. Yuri et al. predicted the recession of Al_2O_3, SiC, Si_3N_4, and $Lu_2Si_2O_7$ phases under gas turbine conditions [3,4]. For instance, Si_3N_4 exhibited 6 mm recession after 10,000 h exposure test in high speed combustion gas flow with a velocity of 150 m/s at 1250 °C [4]. In the development of ceramics gas turbine for power generation, the thermal efficiency could only be improved by increasing of the turbine inlet temperature (TIT). Thus, the components of gas turbine engine must remain stable under high velocity combustion gas flow for at least 1 year (8760 h) operation. Previous studies have suggested that an environmental barrier coating (EBC) system is necessary to protect silicon based non-oxide ceramics from harsh gas turbine environments [1,3,4].

Based on the present authors suggested that the recession rate and/or the corrosion rate of $Lu_2Si_2O_7$ phase was slower than those values measured for other oxides [3–6]. The recession rate of this material was attributed to mass transfer of evaporated species under high-speed combustion gas flow, which could be described by Arrhenius plot [3]. The proposed mass transfer mechanism for the recession of ceramics by high speed and high pressure water vapor flow was supported by other studies as well [7–11].

In the recession of $Lu_2Si_2O_7$ bulk under high speed combustion gas flow with a velocity of 140 m/s, silica component was removed from the bulk surface and the phase ratio of $Lu_2SiO_5/Lu_2Si_2O_7$ increased with increasing temperature [3]. Also, in the recession study of $Lu_2Si_2O_7$ bulk under high speed steam jet with a velocity of 50 m/s, phase changes

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from $Lu_2Si_2O_7$ to Lu_2SiO_5 at 1300 °C and from $Lu_2Si_2O_7$ to $Lu_2SiO_5 + Lu_2Si_2O_7$ at 1500 °C, respectively, were observed [6]. These results suggest that Lu_2SiO_5 phase could be more stable than $Lu_2Si_2O_7$ phase under high speed combustion gas flow and the component of evaporated species could vary with gas velocity, gas pressure, and partial pressure of water vapor. Since the corrosion and recession mechanism of ceramics such as morphology and phase changes strongly depend on the component of the evaporated species, the component of the evaporated species would be governed by the velocity and partial pressure of corrosive gas and phase.

In the present paper, the recession study for Lu_2SiO_5 phase, which is more stable than $Lu_2Si_2O_7$ phase was performed under high speed steam jet with a velocity of 50 m/s, which is same level of gas velocity in common gas turbine. The phase and morphology changes after high temperature steam exposure are discussed.

2. Experimental procedures

The Lu_2SiO_5 bulk sample was prepared by hot pressing method. High purity Lu_2O_3 (99.99% purity, Shin-Etsu Chemical Co. Ltd., 4 μm particle) and SiO_2 (99.9% purity, High Purity Chemicals Co. Ltd., 0.8 μm particle) powders are used as starting materials. A stoichiometric molar ratio of Lu_2O_3 and SiO_2 (Lu_2O_3:SiO_2 = 1:1) powders were mixed by ball milling in ethanol for 24 h and then, dried in air at 100 °C for 24 h. This mixed powder was sintered at 1500 °C in Ar atmosphere with 0.3 MPa for 12 h. The sintered bulk was cut into test pieces with dimensions of 3 mm × 4 mm × 40 mm.

The steam jet exposure test was performed at 1300, 1400, and 1500 °C for 100 h using a water injection facility at Oak Ridge National Laboratory (ORNL) as shown in Fig. 1 [12]. The sample was positioned with an oxide fixture in the furnace. The steam was pre-heated up to 250 °C and injected onto the sample surface with 50 m/s in velocity. The exposure area is ~4 mm × 10 mm as shown in the figure.

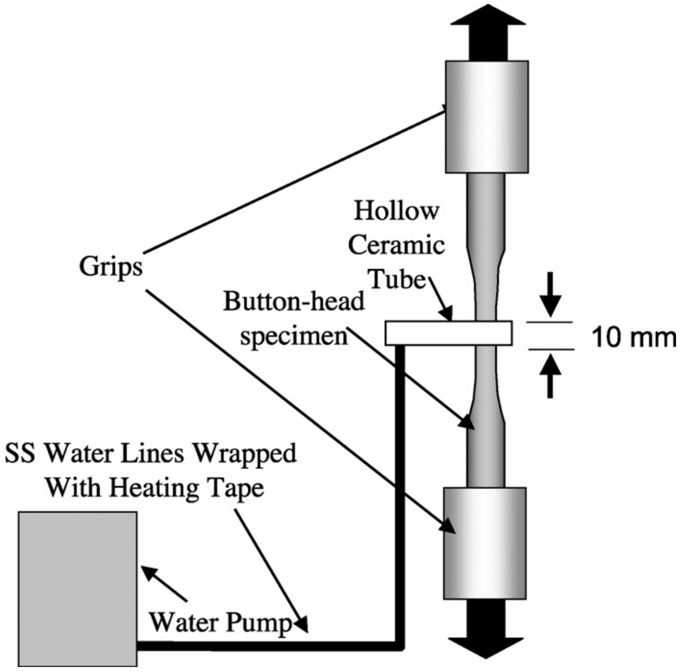

Fig. 1. The configuration of the machine for steam jet test [1].

The X-ray diffraction pattern for each sample was obtained from the exposure area. Scanning electron microscopy examination was also performed in details for the sample surface after steam exposure.

3. Results and discussions

3.1. Kinetics of the recession mechanism

The weight loss rate for the samples can be calculated from the weight changes before and after the test. Fig. 2 shows the Arrhenius plot of the weight loss rate. Since a linear relation is obtained in the Arrhenius plot, the recession mechanism of Lu_2SiO_5 sample can be explained by a model of material transfer of evaporated species from the surface to a high speed gas flow and the same recession mechanism can be applied for test carried out 1300, 1400 and 1500 °C.

Yuri et al. reported the recession of $Lu_2Si_2O_7$ under high speed combustion gas flow with a velocity of 150 m/s can be explained by a mass transfer mechanism and the weight loss rate k_w can be expressed by Eq. (1) [3].

$$k_w = c \cdot \exp\left(-\frac{E}{RT}\right) \cdot (P_{H_2O})^n \cdot \frac{Re^{0.8}}{P} \tag{1}$$

where c, E, R, T, P_{H_2O}, Re, and P denote a constant, apparent activation energy (kJ/mol), gas constant (8.31 kJ/mol), absolute temperature (K), water vapor partial pressure (MPa), Reynolds number, and total pressure, respectively. In this model, the kind of evaporated species remains constant during the steady state of the recession tests. In the reference [3], a porous structure is formed and the phase ratio of $Lu_2SiO_5/Lu_2Si_2O_7$ increased with increasing temperature. Hence, it could be assumed that the main component of the evaporated species is silica hydroxide and the ratio of silica component/lutetia component in the specie is constant, where Si/Lu is larger than 1.

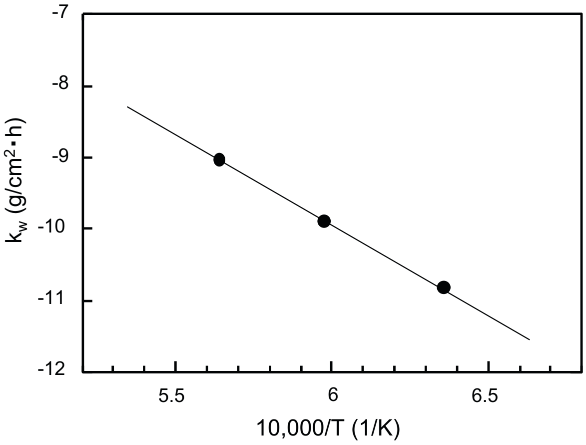

Fig. 2. Arrhenius plot of the weight loss by the steam jet tests.

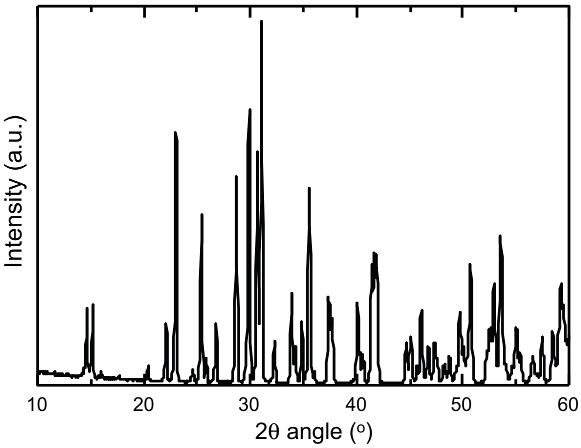

Fig. 3. The X-ray diffraction pattern of the sample before the test. All of peaks can be indexed as Lu_2SiO_5 phase.

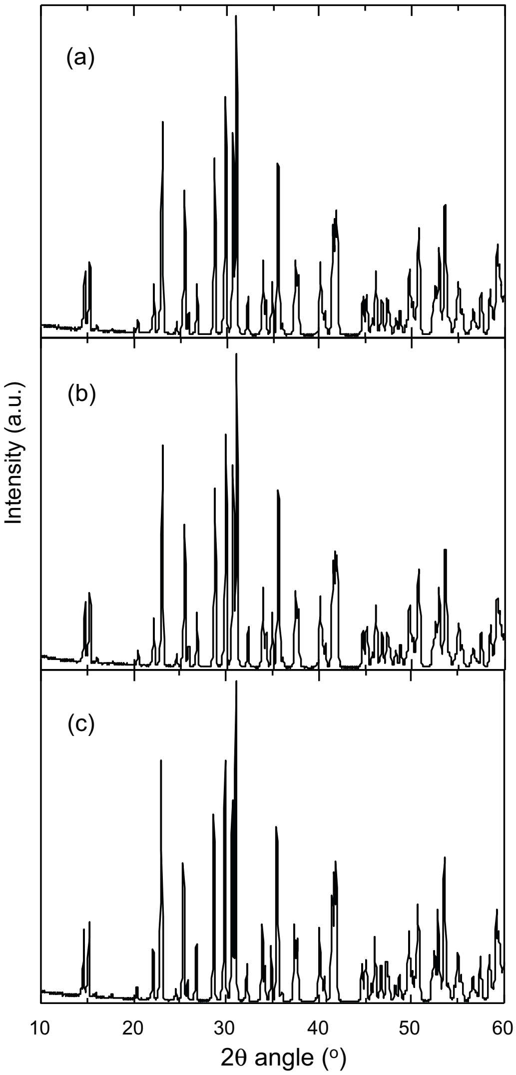

Fig. 4. The X-ray diffraction patterns of (a) 1300 °C, (b) 1400 °C and (c) 1500 °C test samples. No phase changes can be recognized.

Fig. 3 shows the X-ray diffraction pattern of the sample before the steam exposure test. All of the peaks can be readily indexed as Lu_2SiO_5 phase. Fig. 4 shows the X-ray diffraction pattern of (a) 1300, (b) 1400 and (c) 1500 °C test samples. No phase changes can be observed. These results suggest that (1) the recession of Lu_2SiO_5 bulk by a high speed steam jet with a velocity of 50 m/s can be explained by a mass transfer model as reported in previous studies [3,7–11] and (2) the composition of the evaporated species is consisted of $Si(OH)_4$ and $Lu(OH)_3$ with the ratio of $Si(OH)_4/Lu(OH)_3$ is 1/2 in the temperature range of 1300 and 1500 °C.

3.2. Corrosion mechanism of bulk surface

In a thin laminar flow region at solid–gas interface, the formation of gas species, due to the corrosion of bulk surface occurs. The corrosion of bulk surface sometimes could induce changes in structure and phase. Note that phase and structure changes were observed for the recession test for $Lu_2Si_2O_7$ bulk under high speed steam jet with a velocity of 50 m/s as reported in our previous study [6]. Since the laminar flow region thin down with the velocity of gas flow, the corrosion and phase change was observed for samples tested under a combustion gas flow with a velocity of 150 m/s [3]. In the reference of [6], Lu_2SiO_5 phase was formed at bulk surface at 1300 °C and $Lu_2Si_2O_7$ phase was regenerated at 1500 °C [6]. Since the phase and structure (material volume) changes led to a stress state generated on the bulk surface, it is thus important to know the corrosion of bulk surface by water vapor in a thin laminar flow region on the development of EBC.

Fig. 5 show the bulk surface for (a) before the test, (b) after 1300 °C test, (c) after 1400 °C test and (d) 1500 °C test, respectively. Observations showed that cracks formed on the bulk surface for the samples of 1400 and 1500 °C. Also porous structures are formed for the samples of 1300 and 1400 °C. However, the porous structure changed in the sample of 1500 °C. Fig. 6 show high magnification images for Fig. 5. In the sample of 1300 °C test (Fig. 6(a)), the grain boundary glassy phase was removed by water vapor and each grain is stand out in clear. However, the boundary phase was present in the sample of 1400 °C, even though no phase change occurred between 1300 and 1400 °C tests. The number of porosity decreased in 1500 °C sample and small holes were formed. Since the small holes were mostly located in triple points of the surface grains as shown in Fig. 6(e), the holes could be formed due to the grain growth process.

Results obtained in the present study suggest that the boundary glassy phase and the surface of Lu_2SiO_5 grains were corroded by water vapor at 1300 °C. Also, the boundary glassy phase was regenerated and the surface of Lu_2SiO_5 phase and boundary phase were corroded at 1400 and 1500 °C even though the composition of the evaporated species is almost constant of Si/Lu = 1/2. Since the grain growth and reduction of porous structure occurs at 1500 °C, it is considered that the cracks found in Fig. 4(c) and (d) induced during the cooling step after exposure tests.

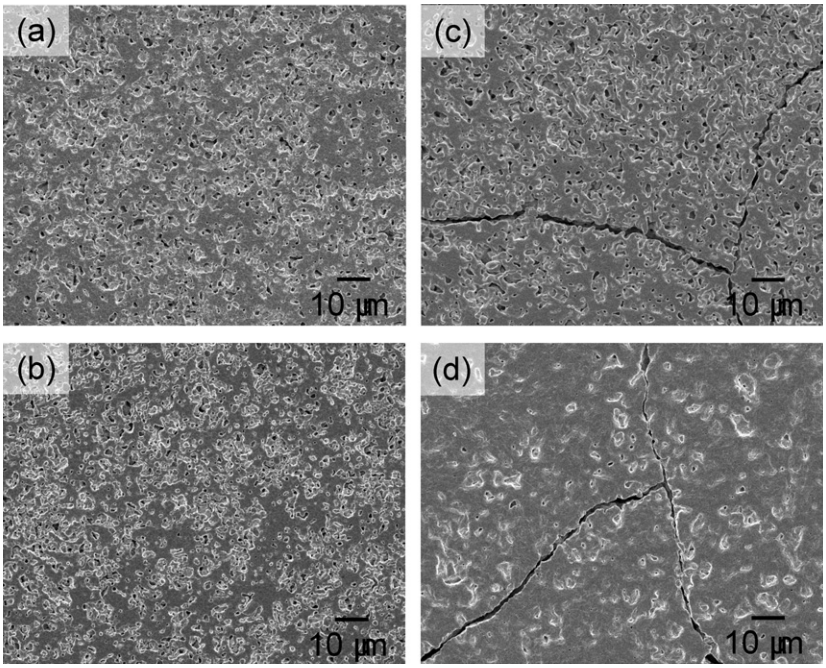

Fig. 5. The bulk surface for (a) before the test, (b) after 1300 °C test, (c) after 1400 °C test and (d) 1500 °C test, respectively.

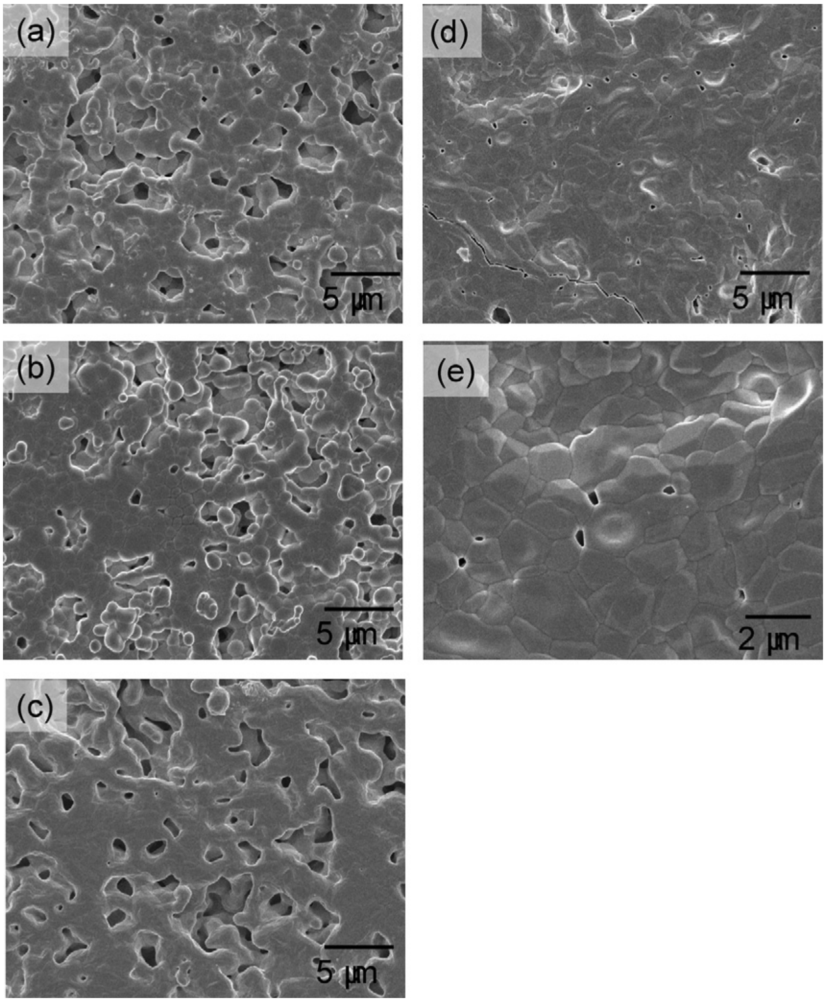

Fig. 6. (a–e) High magnification images for Fig. 5.

4. Conclusions

The recession behavior of Lu_2SiO_5 bulk was examined by an exposure test using high speed steam jet with a velocity of 50 m/s in a temperature range of 1300 and 1500 °C. The recession mechanism of this phase can be explained by a mass transfer model and the composition of the evaporated species is constant with Si/Lu = 1/2. In laminar flow region, boundary glassy phase was removed from the bulk surface at 1300 °C. The corrosion of Lu_2SiO_5 grain was observed for 1400 and 1500 °C tests. In steady state, the removal of boundary phase, regeneration of boundary phase, corrosion of silica and lutetia component from Lu_2SiO_5 grains remained stable resulting in a constant ratio of Si/Lu in evaporated species in the temperature range of 1300 and 1500 °C.

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