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Hot corrosion behaviour of barium-strontium aluminosilicates in a molten Na_2SO_4 environment

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ABSTRACT

The hot corrosion behaviour of barium-strontium aluminosilicates ($B_{1-x}S_xAS$) attacked by Na_2SO_4 was investigated in the temperature range from 900 to 1100 °C and the weight change was measured as a function of the corrosion time. The surfaces and cross-sections of the corroded samples were observed by scanning electron microscopy in backscattered electron mode and energy-dispersive X-ray spectroscopy. The phase composition was characterized by X-ray diffraction. The results indicate that the hot corrosion of $B_{1-x}S_xAS$ by molten Na_2SO_4 was controlled by a diffusion-reaction mechanism. The strontium and/or barium cations diffused out of their aluminosilicate network, and the vacant sites were filled by sodium cations diffusing into the structure to form a $NaAlSiO_4$ on the top. Due to their smaller radius, the strontium atoms showed a faster diffusion rate than the barium atoms. The corrosion depth significantly increased with the temperature and the strontium concentration in the $B_{1-x}S_xAS$.

1. Introduction

Advanced ceramic matrix composites (CMCs), in particular silicon carbide fiber reinforced silicon carbide ceramic matrix composites (SiC/SiC), are increasingly used in the aerospace industry for the manufacturing of hot section components due to their excellent properties, including their low weight, high strength, and high temperature resistance compared with superalloys [1–3]. In the aero-engine, these components are exposed to erosive media from the turbine air inlet and fuel, which may result in their failures. CMCs with a self-sealing matrix typically show a good oxidation resistance in dry air [4,5]. However, compared with dry air, the combustion environment is much more complex. About 5–10% of the combustion gas is water vapour resulting from the burning of hydrocarbon fuels in air [6]. Silicon-based ceramics have been demonstrated to oxidise and react with water vapour to form volatile silica hydroxide, leading to the damage of the components [7–9]. Simultaneously, the impurities sucked in from the environment or introduced by the fuel may react to form low-melting point salts such as NaCl, KCl, Na_2SO_4, Na_2CO_3, K_2SO_4, $NaVO_3$, and/or volcanic ash. These molten salts can deposit on the CMC components and dramatically accelerate the degradation of the components' performance [6,10–14].

To protect the SiC/SiC components from corrosion, environmental barrier coatings (EBCs) have been developed to increase their durability in combustion environments [15]. These EBC materials usually exhibit a good resistance to hydrothermal corrosion [16–18]. Although there are several reports on the corrosion behaviour and performance of EBCs in a hot corrosion environment [19–21], the corrosion mechanism is still not clear. Barium-strontium aluminosilicates ($B_{1-x}S_xAS$, $x = 0–1$) have been identified as an excellent protective coating material due to their stability and low CTE value which is similar to that of SiC [22], and have been successfully employed for protecting components in the hot sections of aero-engines [23]. Nevertheless, the corrosion mechanism and the effect of the addition of strontium oxide by substituting strontium for barium in $B_{1-x}S_xAS$ on the hot corrosion behaviour of $B_{1-x}S_xAS$ remain to be studied in detail.

In this study, we aimed to reveal the hot corrosion behaviour of $B_{1-x}S_xAS$ with different barium to strontium ratios in a molten Na_2SO_4 environment in the temperature range from 900 to 1100 °C. A model for the corrosion mechanism is also proposed.

2. Material and methods

$BaCO_3$ (analytical grade, Zhongxin Fine Chemical Factory, Xi'an, China), $Sr(NO_3)_2$ (analytical grade, Bodi Chemical Corp., Tianjin, China), and $Al(NO_3)_3 \cdot 9H_2O$ (analytical grade, Hongyan Chemical Reagent Factory., Tianjin, China) were used as the starting materials for the fabrication of the $B_{1-x}S_xAS$. First, the materials

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were weighed according to the intended molar ratios (Ba: Sr: Al: Si = 1−x:x:2:2), and then dissolved in an aqueous citric acid solution (40 wt%). Simultaneously, tetraethoxysilane (TEOS, analytical grade, Kemiou Chemical Reagent Corp., Tianjin, China) was dissolved in a solution of ethanol and deionized water according to a 1:4:1 ratio. Next, the two solutions were mixed and the pH value was regulated to about 3–4 by adding ammonia. A stable and transparent sol was obtained after the complete mixing of the reactants. The transparent solution was heated to form a gel at 60 °C for 12 h and oven dried at 120 °C for 24 h. The dried gels were calcined at 900 °C for 2 h and then milled into fine powders. Green disks with a diameter of 15.7 mm and a thickness of 5 mm were produced by cold pressing at 200 MPa for 1 min. Finally, the dense $B_{1-x}S_xAS$ samples were obtained after sintering the green disks at 1600 °C for 3 h in air.

Prior to the tests, the saturated Na_2SO_4 water solution was uniformly dropped onto the polished surface, followed by evaporation of the solvent. The area density of Na_2SO_4 on the surface was about 12.5 ± 2.5 mg/cm^2. The corrosion behaviour of $B_{1-x}S_xAS$ in a molten salt environment was investigated in an alumina tube furnace at

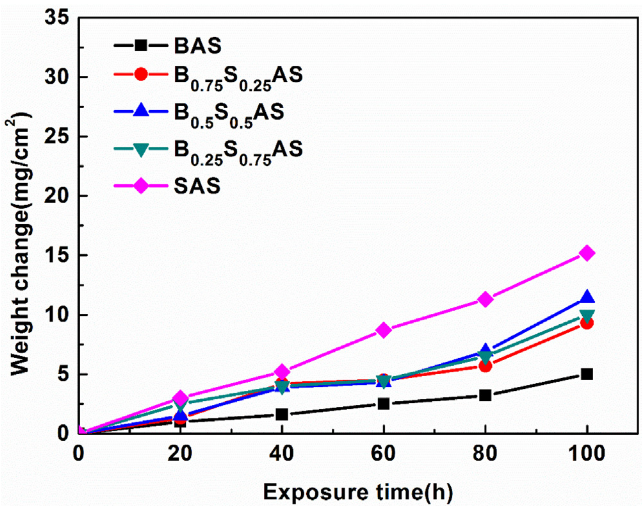

Fig. 1. Weight changes of the $B_{1-x}S_xAS$ samples subjected to molten Na_2SO_4 at 900 °C as a function of the corrosion time.

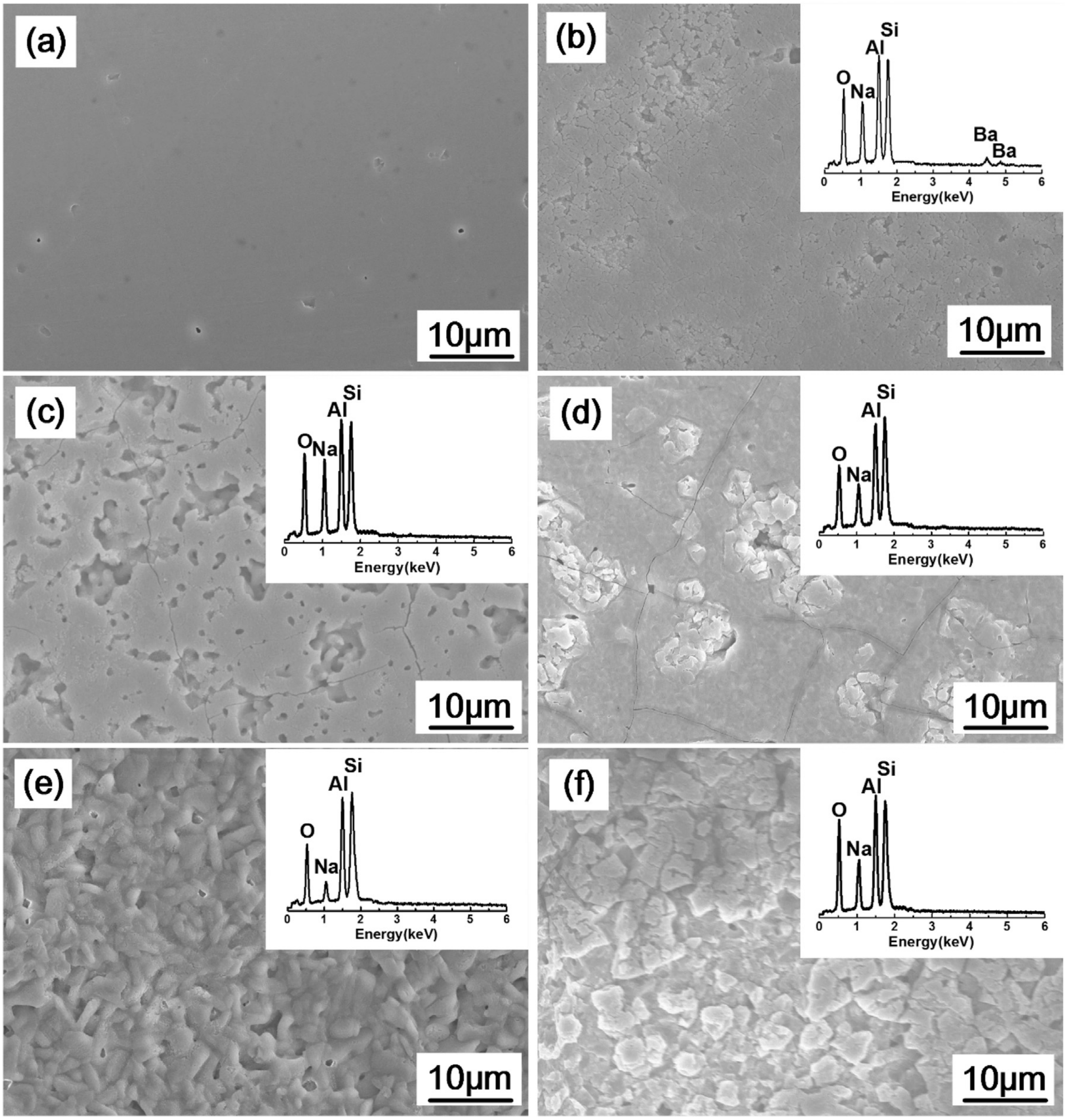

Fig. 2. (a) Typical surface morphology of the $B_{1-x}S_xAS$ samples after polishing and surface morphologies of the $B_{1-x}S_xAS$ samples after corrosion in molten Na_2SO_4 at 900 °C for 100 h and results of the EDS analysis: (b) BAS, (c) $B_{0.75}S_{0.25}AS$, (d) $B_{0.5}S_{0.5}AS$, (e) $B_{0.25}S_{0.75}AS$, (f) SAS.

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