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Original Article

Effect of the phase transformation on fracture behaviour of fused silica refractories

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ABSTRACT

In this paper, the influence of phase transformation on the properties and fracture behaviour of fused silica refractory was investigated. The virgin fused silica refractory is amorphous, and possible failure is attributed to the propagation of a single crack in the structure. Due to the crystallization and phase transformation of low-/high- temperature cristobalite subpolymorphs occurring during the heat treatment, microcracks are formed especially in the matrix and at the grain boundary. This microcracking enables the development of sizable fracture process zone, which is responsible for the increase of specific fracture energy even with the decrease of strength. Therefore, the heat-treated specimens exhibit lower brittleness and higher strain tolerance before failure compared with the virgin fused silica refractory. All of these properties represent a better thermal shock resistance. Furthermore, microcracking causes a characteristic temperature dependence of Young's Modulus due to phase transformation and partial crack closure at increased temperatures.

1. Introduction

Silica refractory is a special category of non-basic refractory products containing more than 93 wt% silica [1]. Silica is rather unique and complex because of its diverse polymorphs and subpolymorphs [2,3]. Thereinto, cristobalite and tridymite are the mineral silica phases in silica refractories. Due to the low- and high- temperature subpolymorphs of these two phases and the induced microstructure change, silica refractories often exhibit temperature dependent elastic and inelastic properties [4,5]. The thermal expansion coefficient of cristobalite, which becomes relatively small or even slightly negative at temperature higher than 800 °C, is predominant for silica refractories [6]. It was also proved by impulse excitation investigation that the damage accumulation is more crucial for silica materials heated to lower temperatures [7]. Nevertheless, its good performance at high temperature, remarkable refractoriness and good chemical resistance to certain acidic components enable silica refractories specially used for constructing coke ovens, glass melter roof, nonferrous metallurgical furnaces etc. [8,9].

In service, they undergo high temperature, thermal shock, mechanical stress, corrosion and erosion. The thermo-mechanical stress induced failure is one of the predominant factors limiting longevity of refractory linings. The compressive creep behaviour and the cyclic fatigue of silica refractories were investigated [10–12]. Two principal types of silica refractories are commercially available for the users: one is the conventional silica refractory produced from quartz raw materials and mainly consisting of cristobalite and tridymite; the other is the fused-silica based refractory [5,10,13]. The latter one is amorphous and shows smaller thermal expansion compared to the crystalline silica refractories at least in the low temperature region, which is believed to cause better thermal shock resistance. Although the interest in its application has grown, the fracture behaviour of fused silica refractories was not studied yet. The crystallization of cristobalite from amorphous silica will gradually take place during their service lifetime. The crystallization as well as the further possible phase transformation cause a critical effect on the thermo-mechanical performance of fused silica refractories. The influence of sintering temperature and additives (i.e. zircon and alumina) on the crystallization of fused silica based ceramics have been investigated [14,15]. Furthermore, the phase transformation of low- /high- temperature cristobalite was simulated and its auxetic behaviour was also studied to extend the understanding of this crystalline silica [16,17]. The strength of fused silica refractories reduces due to the devitrification of amorphous fused silica to cristobalite, and is attributed to microcracking of the cristobalite caused by the phase transformation between the high-temperature cubic cristobalite and low-temperature tetragonal cristobalite (about 2.8%–5% volume

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contraction in the temperature range 200–250 °C during cooling) [18,19]. The strength of fused silica is dependent on cristobalite content [20]. Besides, by acoustic emission technology and fatigue test, Andreev et al. [10] have proven that the presence of initial microcracks in silica refractories reduces the brittleness and limits the severe damage.

In the present paper, the temperature dependent Young's Modulus and dilatation as well as the fracture behaviour of fused silica refractories is investigated. The main purpose is to receive insight into the temperature dependent fracture properties and to understand the influence of crystallization and phase transformation on the fracture behaviour of fused silica refractories.

2. Experimental

2.1. Materials

The fused silica refractory investigated in this work is a commercial product from an industrial producer. The received fused silica refractory is unfired and consist of 98.63 wt% SiO_2 and trace amount of impurities (mainly Al_2O_3, CaO, Fe_2O_3 and TiO_2). The fused silica refractory was heated up to 1400 °C with a heating rate of 3 °C/min and held at the target temperature for 0.5, 2, 5 and 10 h, respectively. The cooling rate was also preset to 3 °C/min. However, the cooling rate below 300 °C slowed down due to the heat capacity of the furnace and prolonged the dwell time at low temperature range. Normally, the working temperature of fused silica refractory is approximately 1100–1200 °C. In order to imitate the crystallization degree in the first several weeks of service, a higher temperature was adopted to accelerate the crystallization of the fused silica. The virgin fused silica refractory specimens were named as 0H, which was amorphous as shown in Fig. 1. The other specimens were successively named 0.5H, 2H, 5H and 10H, according to their dwell time at 1400 °C. The crystallization of fused silica to cristobalite took place during the aforementioned heat treatment and the crystallization degree increased with the dwell time.

The selected physical properties of each specimen are shown in Table 1. The bulk density and apparent porosity were measured via Archimedes method. The bulk density reached a minimum value for specimen 2H, which has the highest apparent porosity. The theoretic density of low-cristobalite and glass phase are 2.32 g/cm^3 and 2.20 g/cm^3, respectively [2]. As seen from Table 1, the true density of virgin fused silica refractory was 2.27 g/cm^3 and increased due to the crystallization of fused silica. For the specimen 2H, 5H and 10H, the true density is rather close to the theoretic density of low-cristobalite. Meanwhile, the Young's Modulus at room temperature decreased significantly because of the devitrification of fused silica refractory. Less

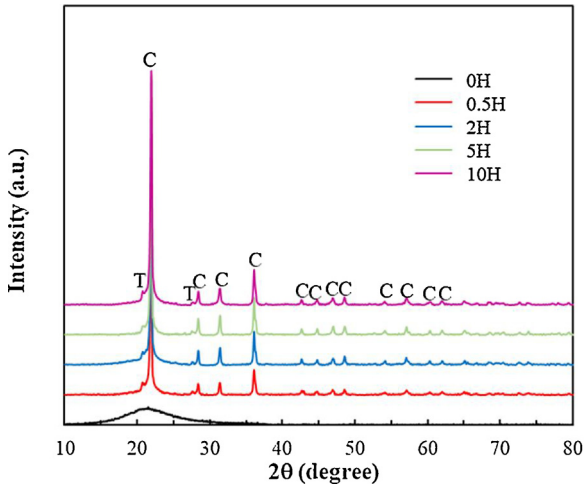

Fig. 1. XRD patterns of the studied silica refractory specimens (C: cristobalite; T: tridymite).

Table 1
Physical properties of silica refractory specimens.

Specimen	0H	0.5H	2H	5H	10H
Bulk density (g/cm^3)	1.81	1.79	1.71	1.78	1.78
Apparent porosity (%)	17.8	21.1	24.4	21.7	22.1
True density (g/cm^3)	2.27	2.29	2.31	2.31	2.31
E_0 (GPa)	26.6	1.8	1.7	1.8	1.4

than 10% of the original stiffness remains after the heat treatment.

2.2. Investigation methods

The chemical and physical characterization of the studied materials was determined by established measurement techniques. The phase composition was investigated using X-ray powder diffractometer (X'pert Pro, PANalytical, Netherland), set to copper radiation (CuK_α, $\lambda = 1.5418$ Å) at 40 kV/40 mA. The micrographs were obtained by scanning electron microscope (Nova 400 Nano-SEM, FEI Company, USA). True density measurements were carried out using automatic True Density Analyzer (AccuPyc II 1340, Micromeritics Instrument Corporation, US) according to GB/T 5071-2013 (Chinese standard of refractory materials—determination of true density). The dilatation curves up to 1400 °C were measured using connecting rod dilatometer (DIL 402 Supreme, NETZSCH, Germany) for cylindrical specimens with diameter of 8 mm. Room temperature and high temperature Young's Modulus were measured via impulse excitation technique, using a resonant frequency damping analyzer (RFDA-HTVP1600, IMCE, Belgium). The impulse excitation measurements were performed up to 1200 °C with a heating and cooling rate of 3 °C/min, the dwell time at 1200 °C for each specimen was 30 min.

Wedge splitting tests (WST) were performed at room temperature to investigate the fracture behaviour of fused silica refractory specimens. The experimental set-up is shown in Fig. 2: the applied vertical force F_V is transformed to a higher horizontal force F_H with the help of a load transmission equipment (wedge, rollers, load transmission pieces) [21]. The specimen size used is 100 mm × 100 mm × 65 mm, the initial notch length a_0 equals to 12 mm, and the wedge angle is 10°. The loading rate was 0.5 mm/min. Due to the application of the load transmission components and a high ratio of fracture area to specimen volume, the WST enables stable crack propagation for brittle or quasi-brittle materials, such as refractory, concrete, rock etc. [22]. A contacting extensometer was used to measure the horizontal displacement at the load points. A load-displacement curve was recorded by the load cell for each specimen and the fracture parameters of the fused silica refractory specimens. The specific fracture energy G_F' is determined by integrating the area under the load-displacement curve divided by the ligament area A, as shown in Eq. (1). The ligament area A is calculated as b × h, which are the width and the height of the ligament, respectively. δ_H is the horizontal displacement, δ is the horizontal displacement at 15% of the maximum load ($F_{H,\ max}$). The wedge splitting tests are terminated before the force reaches zero to avoid the contact between the wedge and the specimens.

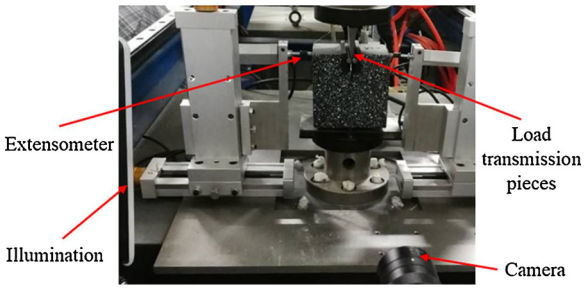

Fig. 2. Experimental set-up of the wedge splitting test with a digital camera.