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Microstructure and Mechanical Properties of Al_2O_3-C Refractories Using Carbon Black and Multi-Walled Carbon Nanotubes as Carbon Sources

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Abstract

Low-carbon Al_2O_3-C refractories were prepared using tabular alumina and reactive alumina powder as the main raw materials and silicon powder as an additive, while nano carbon black and multi-walled carbon nanotubes (MWCNTs) were added as nano carbon sources. The low-carbon Al_2O_3-C refractories were first fired at 800 °C, 1000 °C, 1200 °C and 1400 °C in a coke bed. Then the phase compositions and microstructures of the samples coked at the above temperatures were analyzed by means of X-ray diffraction (XRD) and scanning electron microscopy (SEM). Finally, the mechanical properties and thermal shock resistance of the samples were evaluated using three-point bending methods. The results showed that: 1) The mechanical properties of MWCNTs-containing Al_2O_3-C refractories coked above 1000 °C were significantly improved, which is attributed to the strengthening effects of MWCNTs and the formation of a large quantity of SiC whiskers via transformation of MWCNTs based on the Vapor-Solid model. 2) Thanks to the thermal stress absorption effect of nano carbon black, strengthening effects of MWCNTs and SiC whiskers, the low-carbon Al_2O_3-C refractories containing 1 wt% mixture of nano-carbon-black and MWCNTs achieved a similar residual strength ratio to those containing a 2 wt% mixture of nano carbon black and flaky graphite, exhibiting a higher residual strength.

Keywords: Mechanical properties, thermal shock resistance, Al_2O_3-C refractories, nano carbon black, MWCNTs

I. Introduction

Since the late 1970s carbon-containing refractories have become one of the most essential products used in the steelmaking industries [1]. Nowadays low-/ultra-low-carbon refractories are the state-of-art for clean steel products when energy saving, emission reduction and environmental issues are concerned [2–4]. However, their thermal shock resistance deteriorates as the carbon content decreases [5–9]. So, the innovation of a new generation of low-/ultra-low-carbon-containing refractories with excellent thermal shock resistance and mechanical properties has been the paramount focus in this field.

In recent years, carbon-containing refractories have been designed as a nano-/micro-structured matrix combining flaky graphite with different nano carbon sources, such as nano carbon black, MWCNTs, nano carbon fibers and graphene. For example, low-C MgO-C refractories with a mixture of 0.9 wt% nano carbon black and 3 wt% flaky graphite as carbon sources have exhibited thermal shock resistance as good as that of refractories with 10 wt% graphite [10–12]. Meanwhile, their low thermal conductivity can also reduce the heat loss because of the low carbon content in refractories [13]. Additionally, Matsuo [14] verified that MgO-C refractories with 0.4 wt% nano carbon fibers had 2.2 times higher strength than that of the reference refractories. Luo, Aneziris *et al.* used MWCNTs to reinforce low-carbon Al_2O_3-C refractories together with *in-situ*-formed whiskers at high temperatures owing to the pull-out, crack deflection and bridging effects [15–19]. Besides 1D nano carbon sources, 2D graphene oxide nanosheets (GONs) were added into refractories to partly replace graphite flake. And better mechanical properties and thermal shock resistance can be achieved with just 0.2 – 0.5 wt% graphene [20, 21].

In this present work, new kinds of Al_2O_3-C refractories with excellent mechanical properties and good thermal shock resistance were developed by combining nano carbon black and MWCNTs as carbon sources in order to reduce the carbon content to lower than 3 wt%. In this case, nano carbon black filled the spaces between oxide particles to absorb the thermal stress during thermal cycles [22, 23]. In addition to that, MWCNTs reinforced and toughened the matrix together with *in-situ*-formed SiC whiskers via vapor reactions at high temperatures [17, 24]. In this experiment the microstructure, mechanical properties and thermal shock resistance of low-carbon Al_2O_3-C refractories containing 1 wt% mixture of carbon black and MWCNTs were studied in comparison with a refractory

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material containing 1 wt% nano carbon black and 1 wt% flaky graphite as a reference.

II. Experimental

(1) Preparation of the Al_2O_3-C samples

The raw materials used for preparing the Al_2O_3-C refractories samples were tabular alumina (3.0 ~ 1.0, 1.0 ~ 0.5, 0.6 ~ 0.2, 0.3 ~ 0, ~ 45 μm, ~ 20 μm, 98 wt% Al_2O_3, Qingdao Almatis Premium Alumina Co. Ltd., China), reactive alumina powder (~ 2 μm, 98 wt% Al_2O_3, Qingdao Almatis Premium Alumina Co. Ltd., China), silicon powder (45 μm, 98.47 wt% Si, Anyang, China), nano carbon black (N220, 99.5 wt% fixed carbon, Wuhan Cobo New Materials Co. Ltd., China), multi-walled carbon nanotubes (MWCNTs, 95 wt% fixed carbon, Chengdu Organic Chemicals Co. Ltd., China) and flaky graphite (FG, λ 74 mm, 97.5 wt% fixed carbon, Qingdao, China). Additionally, thermosetting phenolic resin (liquid, ≥ 40 wt% fixed carbon, Wuhan Lifa Chemistry & Industry Co., Ltd., China) was added as the binder. Table 1 lists the compositions of the Al_2O_3-C refractories; the samples were labeled C1, T0.05, T0.1, T0.3, T0.5, T1 and C1G1. C1 was the reference sample containing 1 wt% nano carbon black. The end of the labels of T0.05, T0.1, T0.3, T0.5, and T1 mean that the MWCNTs contents were 0.05 wt%, 0.1 wt%, 0.3 wt%, 0.5 wt% and 1 wt%, respectively. For comparison, an additional sample with mixture of 1 wt% nano carbon black and 1 wt% flaky graphite was prepared and labeled C1G1.

The nano carbon sources were first ball milled with alumina powder (~ 2 μm) in order to disperse them into the matrix of refractories. The specific mixing process was as follows: 1 wt% nano carbon sources and 10 wt% reactive alumina powder were mixed manually with ethanol as the dispersion medium to obtain a slurry, then corundum balls were used as grinding media and kept the mass ratio of corundum balls: ingredients: ethanol at 1.3:1:1.5. The slurry and the corundum balls were put into milling tanks and placed in a planetary ball mill, which was run for 4 h at a speed of 300 rpm. Finally, the mixture was dried at 80 °C for 24 h, then ground finely.

Before the mixing, all the fine powders were pre-mixed for 10 min. We started with mixing the aggregates for 3 ~ 5 min, adding the phenolic resin, and running the mixer for 3 ~ 5 min, then adding the pre-mixed fine powder and mixing for another 30min. The well-dispersed mixtures were cold pressed into samples measuring 25 mm × 25 mm × 140 mm at 165 MPa and cured at 140 °C for 24 h. Finally, the as-prepared Al_2O_3-C refractories samples were coked at 800 °C, 1000 °C, 1200 °C and 1400 °C in a saggar filled with coke grit with a dwelling time of 3 h, respectively. The heating rate of 5 °C/min was adopted from room temperature to 1000 °C and 2 °C/min afterwards.

(2) Tests and characterization methods

The phase compositions of the coked samples were analyzed by means of X-ray diffraction (XRD, x'Pert Pro, Philips, Netherlands). Microstructures of the samples were observed with scanning electron microscopy (SEM, Quanta 400, FEI Company, USA) and transmission electron microscopy (TEM, 2000F, Jeol Ltd., Japan), both of them equipped with energy-dispersive X-ray spectroscope (EDS, Noran 623M-3SUT, Thermo Electron Corporation, Japan). Cold modulus of rupture (CMOR) was measured in the three-point bending test at ambient temperature with a span of 80 mm and a loading rate of 0.5 mm/min using an electronic digital control system (EDC 120, DOLI Company, Germany). The force displacement curves were recorded simultaneously during the test. All the preceding measurements were conducted with three samples of each composition.

The thermal shock resistance of the samples pre-coked at 1400 °C was tested. The test procedure can be described as follows: i) heating the samples in a coke bed to 1100 °C with a heating rate of 5 °K/min and then soaking at this temperature for 30 min; ii) quickly quenching them into flowing water to avoid severe oxidation. Then after drying at 110 °C, the mechanical properties were assessed in a three-point bending test. The residual strength ratio of CMOR was calculated based on the change in CMOR before and after the thermal shocks, i.e. the residual strength ratio of CMOR = 100 %·$CMOR_{TS}$/CMOR, in which the CMOR and $CMOR_{TS}$ represented the CMOR before and after the thermal shock test, respectively.

Table 1: Batch compositions of Al_2O_3-C refractories.

	C1	T0.05	T0.1	T0.3	T0.5	T1	C1G1
Alumina	95	95	95	95	95	95	94
Silicon	4	4	4	4	4	4	4
Carbon black	1	0.95	0.9	0.7	0.5	-	1
Carbon nanotubes	-	0.05	0.1	0.3	0.5	1	-
Graphite flake	-	-	-	-	-	-	1
Phenolic resin (liquid)	+4	+4	+4	+4	+4	+4	+4

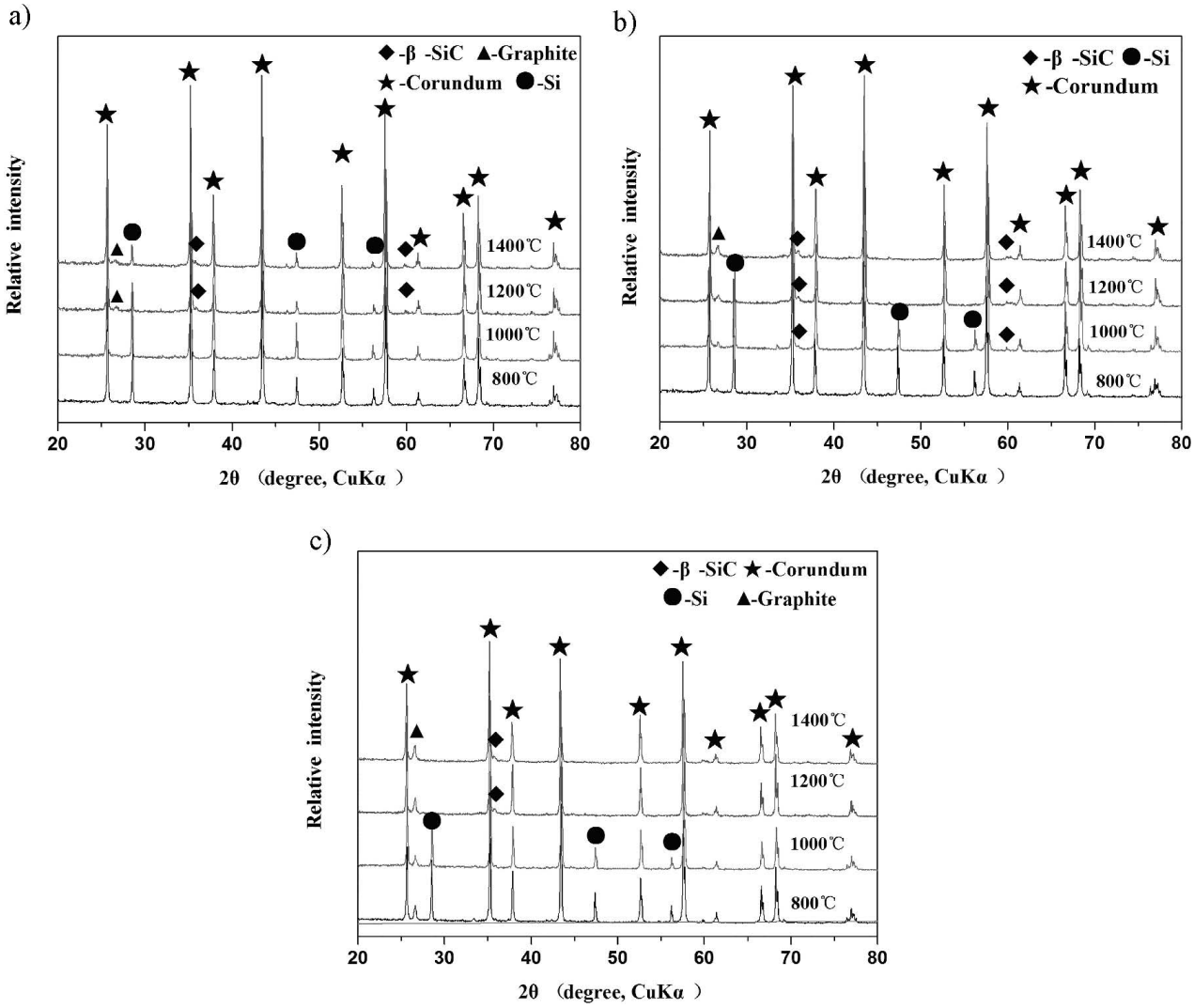

Fig. 1: XRD patterns of samples. (a) C1, (b) T1 and (c) C1G1.

III. Results and Discussion

(1) Phase composition

The phase compositions of the selected low-carbon Al_2O_3-C samples (C1, T1 and C1G1) coked in the range of 800 °C to 1400 °C are examined in Fig. 1. As shown in Fig. 1(a), there is no new phase except corundum and silicon phases in sample C1 fired at lower than 1200 °C. At the temperature of 1200 °C, the SiC and graphite phase appeared. Correspondingly, the peak intensity of silicon decreased. In fact, the graphitization of carbon black was observed to initiate at 1000 °C and became much dominant at higher temperatures [24]. Up to 1400 °C, higher intensity of SiC phase emerged with residual silicon in samples. In comparison, the SiC phase was detected in the T1 sample coked at 1000 °C, while the silicon phase disappeared completely at 1200 °C. Interestingly, the SiAlON phase was also detected at 1200 °C and 1400 °C. Based on factsage calculation, silicon nitride could be stable in Si-N_2-CO system at high temperatures [24], so SiAlON phase as a solid solution was speculated to occur in Si-Al_2O_3-N_2-CO system. However, no SiAlON phase appeared in the C1G1 sample even though it had mostly the same phase composition evolution at all elevated temperatures as the T1 sample.

(2) Microstructures

The microstructures of the Al_2O_3-C refractories samples (C1, T1 and C1G1) coked at 800 °C, 1000 °C, 1200 °C and 1400 °C are shown in Figs. 2, 3 and 4. For the sample C1, no whiskers were observed after coking at lower than 1200 °C. At 1200 °C and 1400 °C, a certain amount of curved nano whiskers appeared in the matrix of samples (Fig. 2). Analyzed with a combination of XRD and EDS analysis, these whiskers could be SiC ones, as stated elsewhere [25,26]. For sample T1 as shown in Fig. 3, MWCNTs were easily observed in the matrix at 800 °C. SiC whiskers measuring a micrometer in length began to appear at 1000 °C. And a larger quantity of the tangled SiC whiskers appeared with rising temperature, which exhibited a higher length/diameter ratio (L/D ratio) than those in sample C1. Similar to sample T1, a small number of SiC whiskers was observed in sample C1G1 at 1000 °C and many more SiC whiskers around residual flaky graphite at 1200 °C [25,26]. At higher temperature, the quantity and L/D ratio of whiskers increased in the matrix (Fig. 4).

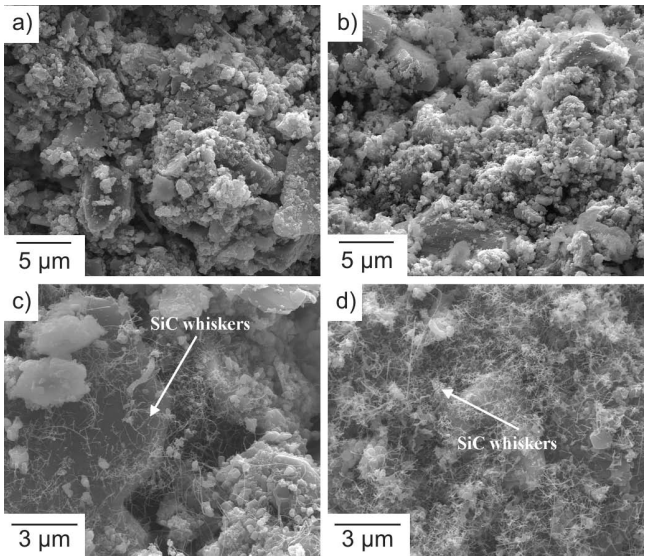

Fig. 2: SEM micrographs of sample C1 coked at 800 °C (a), 1000 °C (b), 1200 °C (c) and 1400 °C (d).

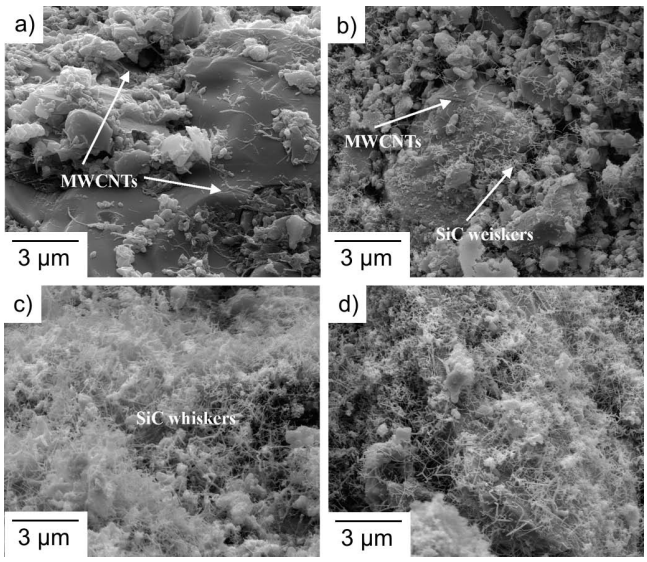

Fig. 3: SEM micrographs of sample T1 coked at 800 °C (a), 1000 °C (b), 1200 °C (c) and 1400 °C (d).

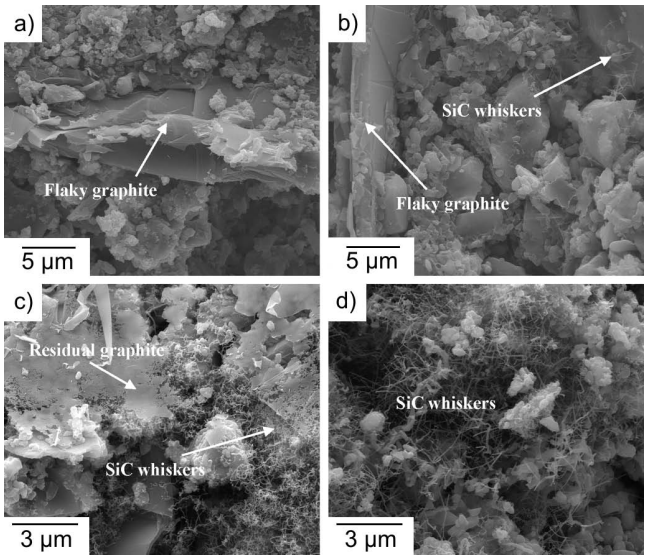

Fig. 4: SEM micrographs of sample C1G1 coked at 800 °C (a), 1000 °C (b), 1200 °C (c) and 1400 °C (d).

To further understand how carbon sources of nano carbon black or MWCNTs affected the microstructures of the silicon-containing Al_2O_3-C refractories, TEM was used to observe the evolution of carbon sources in their matrix. For nano carbon black, its original particle size is around 20 – 30 nm. But it transformed into spherical SiC phase of a smaller size during heating-up [25]. Some of the particles had a core-shell structure in samples coked at 1200 °C, namely, the outside is SiC and the inside is residual carbon black (in Figs. 5a, b). For as-received MWCNTs, they measured 10 – 30 nm in diameter and one micron in length (Fig. 5c). At high temperatures, MWCNTs gradually transformed into SiC phase via reaction deposition of SiO(g) or Si(g) on the surface of the MWCNTs (Fig. 5d). And these SiC whiskers seemed to remain the morphology of MWCNTs (in Figs. 5e, f). Their diameters grew up to 50 nm larger than those of original MWCNTs, which implies that the growth mechanism of SiC whiskers belongs to the V-S model [24, 27 – 29]. In summary, MWCNTs and flaky graphite of the hexagonal structure produced benefits for the formation of SiC whiskers at high temperatures [18, 25, 26] and nano carbon black with amorphous structure is more likely to lead to a core-shell or spherical SiC phase.

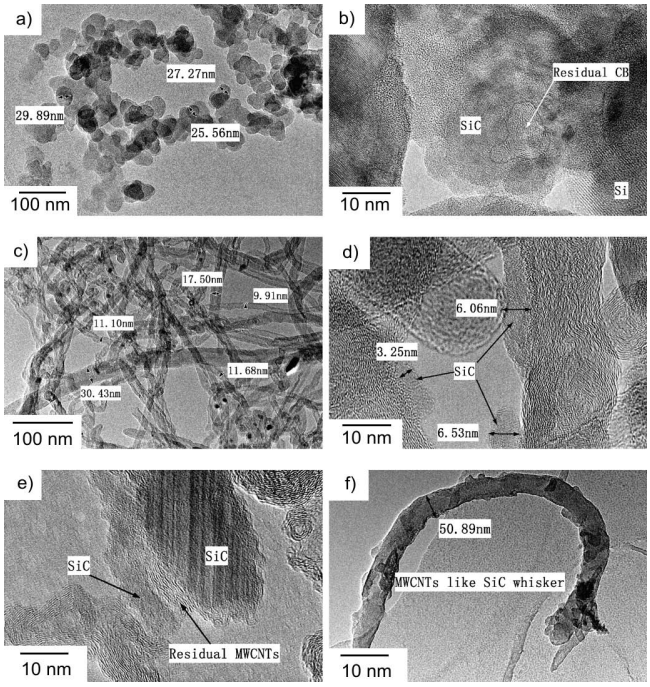

Fig. 5: TEM images of samples C1 (a- as-received CB, b- core-shell structure of CB treated at 1200 °C) and T1 (c-as received MWCNTs; d- SiC formed on MWCNTs at 1000 °C; e, f-residual MWCNTs and MWCNTs like SiC whisker at 1400 °C).

(3) Mechanical properties

Three-point bending test was employed to measure the cold modulus of rupture (CMOR) and the results are summarized in Table 2. For sample C1, its CMOR declined from 5.19 MPa to 5.08 MPa as the heating temperature increased from 800 °C to 1000 °C. However, the CMOR increased to 10.30 MPa and 12.76 MPa at 1200 °C and

1400 °C, respectively, because of *in-situ* formation of SiC whiskers in the matrices (Fig. 2). In contrast, the addition of MWCNTs had significant influence on the strengths of low-carbon Al_2O_3-C refractories. The CMOR of samples containing MWCNTs was mostly improved at all the evaluated temperatures. For example, sample T0.1 increased from 5.76 MPa to 31.44 MPa for its CMOR, which was attributed to the reinforcement of MWCNTs and SiC whiskers in the samples (Fig. 3). For sample C1G1, it had a higher CMOR than that of the samples containing only carbon black (C1), but lower than that of MWCNTs-containing refractories over 1200 °C.

The fractural behavior of Al_2O_3-C samples C1, T0.1 and C1G1 was characterized based on the force displacement curves in Figs. 6(a), (b) and (c), respectively. Obviously, samples C1 and T0.1 enjoyed large displacement before they fractured in comparison with sample C1G1 coked at lower than 1000 °C. In that case, carbon sources (such as particle size and morphology, etc) directly produced the impact on fracture process because there were hardly SiC whiskers occurring in all samples. For nano-sized carbon sources, e.g. most of the carbon black and nanotubes in samples C1 and T0.1 occupied the tiny space among micron or submicron alumina particles. So, spherical particles of carbon black slide against each other to produce the larger displacement during the fracture process. Instead, most of micro-sized flaky graphite in sample C1G1 was located between alumina particles with a less pronounced slide mechanism compared with the spherical carbon black. In addition, carbon nanotubes together with SiC whiskers formed in sample at 1000 °C, being conducive to the large displacement for sample T0.1.

After heat treatment at temperatures above 1200 °C, the force and displacement of the all samples have significantly increased owing to the *in-situ* formation of SiC whiskers in the matrices (Fig. 2 – 4). Especially, sample T0.1 containing 0.1 wt% MWCNTs exhibited the largest force-displacement of all the samples, which was attributed to the strengthening effects of SiC whiskers transformed from MWCNTs.

(4) Thermal shock resistance

The thermal shock resistance was evaluated by measuring the CMOR of the samples coked at 1400 °C before and after thermal cycle (Table 3, Fig. 7). With regard to the series of samples with addition of 1 wt% C, each of the samples containing a mixture of MWCNTs and carbon black had better residual strength than that of samples containing only carbon black, some of them even exceeding sample C1G1 with addition of 2 wt% C. The $CMOR_{TS}$ of samples C1G1 with 2 wt% mixture of nano carbon black and flaky graphite remained as 8.76 MPa, with a higher residual strength ratio of 44.04 % than that of sample C1. In comparison, sample T0.1 exhibited residual strength ratio of 42 % similar to that of sample C1G1, however, the $CMOR_{TS}$ is 9.77 MPa. This is mainly attributed to that fact that nano carbon black can fill the spaces between the oxides to absorb and/or suppress the thermal stress [11, 12], and MWCNTs in combination with SiC whiskers increased the strength of samples before and after thermal shock.

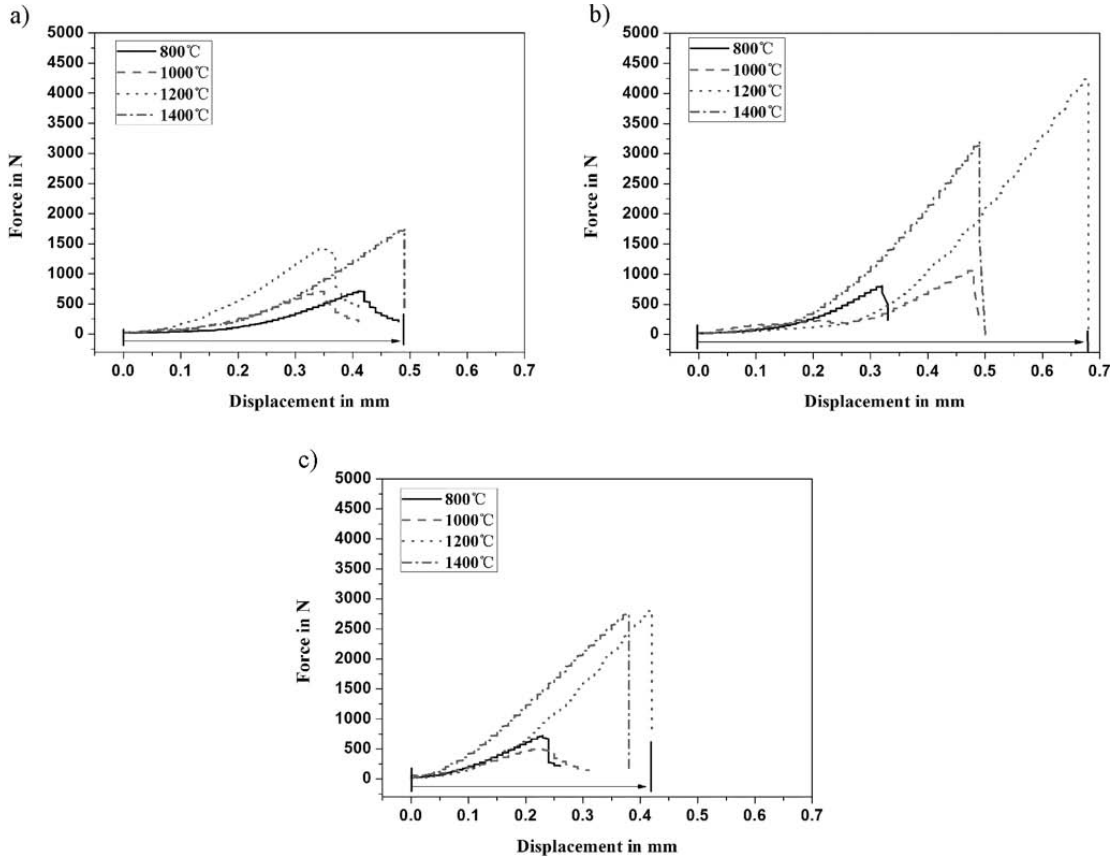

Fig. 6: Force displacement curves of Al_2O_3-C samples C1 (a), T0.1 (b) and C1G1 (c) coked at 800 – 1400 °C.

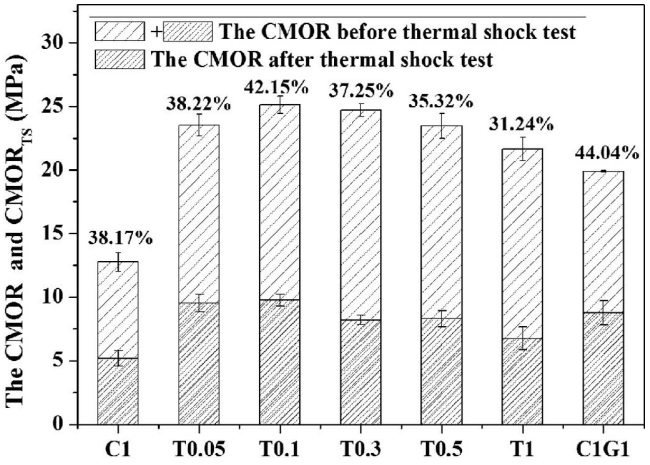

Fig. 7: CMOR, $CMOR_{TS}$ and their residual strength ratio of samples coked at 1400 °C.

IV. Conclusions

Low-carbon Al_2O_3-C refractories were developed by combining nano carbon black and MWCNTs at 800 – 1400 °C. The conclusions can be drawn as follows:

1) The mechanical properties of MWCNTs containing Al_2O_3-C refractories coked above 1000 °C were significantly improved mainly due to the strengthening effects of MWCNTs and their structural transformation into large quantities of SiC whiskers.

2) Most of the low-carbon Al_2O_3-C refractories containing 1 wt% mixture of nano carbon black and MWCNTs not only exhibited comparable residual strength ratio to that of the sample with 2 wt% mixture of nano carbon black and flaky graphite, but also provided higher residual strength.

Acknowledgments

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