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Phase Relations in Si-Al-Y-O-C Systems

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Abstract

The present work investigated the phase relations in SiC-Al_2O_3-Y_2O_3-SiO_2 (Si-Al-Y-O-C) system. As a continuation of our previous works, the purpose of this study is to understand the high temperature reaction behaviors of SiO_2 in the system and its effect on the phase relations of the valuable system of SiC-Al_2O_3-Y_2O_3. The phase compositions of six solid-state reacted samples with different components of Y_2O_3:Al_2O_3:SiC:SiO_2 were analyzed by XRD. The phase relations of the systems were determined. The subsolidus phase diagrams of ternary Al_2O_3-SiC-SiO_2 system and the tentative phase diagram of an extended quaternary Y_2O_3-Al_2O_3-SiC-SiO_2 system were presented latter involving several coexisting regions of four phases. The high temperature reaction behavior of SiO_2 in the system and its effect on the phase relations of system were discussed.

Keywords

Phase Relation, Phase Diagram, SiC, SiO_2, YAG, YAM, YAP, $Al_6Si_2O_{13}$

1. Introduction

SiC ceramic is one of advanced structural ceramics with excellent mechanical properties, mainly in high temperature properties, high hardness, wear resistance, as well as good chemical resistance. It is widely applied in the industry. But its low strength and poor toughness restrict its more extensively applications [1] [2]. Y_2O_3-Al_2O_3 as sintering additives of SiC ceramic have been used for many years [3] [4]. The phase relationship of SiC-Y_2O_3-Al_2O_3 system has also been studied by calculation [5] and experiment [6]. However a few of SiO_2 on SiC surface caused by oxidation of SiC has the non-ignorable effect on the sintering of SiC-Y_2O_3-Al_2O_3 system at high temperature. Thus the effect of SiO_2 on phase relations of SiC-Y_2O_3-Al_2O_3 ternary systems is being concerned. For made it clear this effect, understanding the reaction of SiO_2 with other members in the system at high temperature is required. The phase relations of SiC-Si_3N_4-R_2O_3 (Si-C-N-O-R; R = La, Gd, Y) [7] and SiC-AlN-R_2O_3 (R = Nd, Gd, Yb, Y [8] systems have been reported by the authors. Within systems the effect of SiO_2 impurity on the formation of serial rare-earth nitrogen-containing silicates and silicon-aluminates had been revealed. As the continuation work of them, the present work investigates the high temperature reaction behavior of SiO_2 and the phase relations in the Y_2O_3-Al_2O_3-SiC-SiO_2 system. In this quaternary system the phase diagram of Y_2O_3-SiO_2 subsystem has been identified to form two $Y_2Si_2O_7$ and Y_2SiO_5 compounds [9] [10]. The phase relations of binary Y_2O_3-Al_2O_3 subsystem have also been reported, in which three compounds $Y_4Al_2O_9$ (YAM), $YAlO_3$ (YAP) and $Y_3Al_5O_{12}$ (YAG) were confirmed [11]-[13]. The phase diagram of Al_2O_3-SiO_2 subsystem has

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also been reported to form $Al_6Si_2O_{13}$ (Mullite), and is used in ceramic manufacture [14]. SiC is difficult to react with others oxides. SiC-SiO_2 has a simple binary phase diagram [15], but the SiC-Al_2O_3 phase relation remains unclear [16]. The phase relations of binary Y_2O_3-SiC subsystem has been identified that no any compound was formed [7] [8] [17]. Knowing the phase relations of Y_2O_3-Al_2O_3-SiC-SiO_2 quaternary system will provide help to design and manufacture of SiC ceramic. It will also provide strong evidence to know the factors of controlling the equilibrium of Y_2O_3-Al_2O_3-SiC-SiO_2system.

2. Experimental

2.1. Materials

The starting powders used for the experiments were β-SiC with 0.5% O_2 (mass ratio, the same below) (BF 12-A, H.C. Starck), Al_2O_3 purity ≥ 99.99%, (Xuan Cheng Jing Rui New Material Co., Ltd., China), Y_2O_3 > 99.99% purity (Baotou Research Institute of Rare Earth), SiO_2 (Tianjing Fuchen Chemical Reagents factory, China) respectively. Y_2O_3 and Al_2O_3 powders were calcined at 1100˚C in air for 2 hr to remove their hydrates before being used.

2.2. Experimental Procedure

Selected compositions were marked as YASS in order of Y_2O_3/Al_2O_3/SiC/SiO_2. For example, YASS 1422 represents as the sample with composition of Y_2O_3/Al_2O_3/SiC/SiO_2 = 1/4/2/2 (molar ratio). The details are shown in **Table 1**. The 20 g powder mixture was mixed and ground in an agate mortar for 1.5 - 2 hrs by adding alcohol (analytical reagent, 99.9% purity). After dried up, the prepared powders were cold isostatic pressed under 250 MPa. The conditions used for solid-state reaction were: in Ar atmosphere, at temperature of 1450˚C - 1600˚C, holding 2 hrs, and then cooling down to room temperature freely. In order to achieve the reaction equilibrium, the holding time prolonged. None of noticeable phase composition change is observed could be the judgment of whether the system equilibrium is achieved. The phase compositions of the sintered samples were analyzed by X-ray diffraction (XRD) using equipment SHIMADZU XRD-6000 with CuKα radiation in 0.2˚ scan step, 2˚·min^{-1}. The experimental and the analysis method of phase compositions for samples are similar with our previous papers [6]-[8].

3. Results and Discussion

XRD analysis results of sintered samples are shown in **Table 1**. The effect of SiO_2 impurity on the phase relations in SiC ceramic system is paid close attention. However in our previous work of the phase relations of SiC-Al_2O_3-Y_2O_3 system [6], seemingly no such effect was found (see **Figure 1**). In order to identify SiO_2 caused by oxidation of SiC at high temperature, the SiC powder was heated to 1450˚C, hold 2 hours in Ar. **Figure 2** is XRD pattern of β-SiC starting powder after heating at 1450˚C, showing no trace of SiO_2 could be found. The experimental results also indicated that SiO_2 impurity on the surface of SiC did not participated in the reaction with other oxides in system [6]. There might be too little amount of SiO_2 to detect or the SiO_2 and other components might form a few of liquid at the grain-boundaries, because the eutectic temperature of Y_2O_3-Al_2O_3-SiO_2

Table 1. XRD analyses of sintered samples of Y_2O_3-Al_2O_3-SiC-SiO_2 system.

Sample	Compositions (mol.)				Temp (˚C)	Phases identified in sintered bodies©
	Y_2O_3	Al_2O_3	SiC	SiO_2		
YASS0133	0	1	3	3	1550	$SiO_{2(VS)}$, $SiC_{(S)}$, $Al_6Si_2O_{13(M)}$
YASS0421	0	4	2	1	1550	$Al_2O_{3(S)}$, $SiC_{(S)}$, $Al_6Si_2O_{13(M)}$
YASS1422	1	4	2	2	1550	$YAG_{(S)}$, $Al_2O_{3(M)}$, $Y_2Si_2O_{7(M)}$, $SiC_{(W)}$
YASS3211	3	2	1	1	1550	$YAP_{(VS)}$, $YAM_{(S)}$, $SiC_{(W)}$, $Y_2SiO_{5(W)}$
YASS1414	1	4	1	4	1500	$Y_2Si_2O_{7(VS)}$, $SiC_{(M)}$, $Al_6Si_2O_{13(M)}$, $Al_2O_{3(W)}$
YASS1315	1	3	1	5	1500	$Y_2Si_2O_{7(VS)}$, $SiC_{(M)}$, $Al_6Si_2O_{13(M)}$, $SiO_{2(M)}$

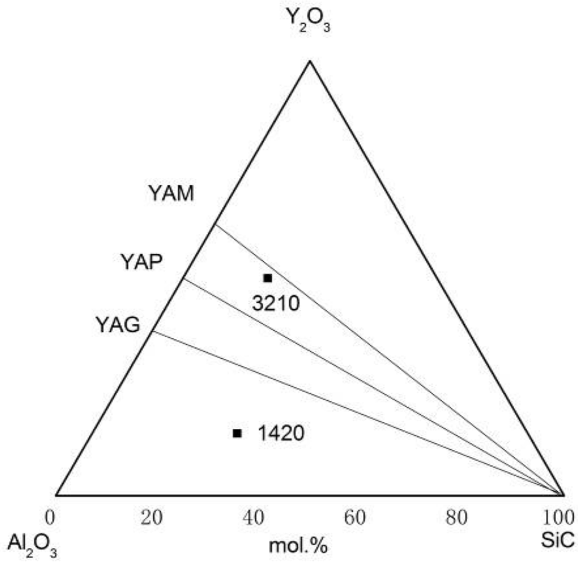

Figure 1. Phase relations of SiC-Al_2O_3-Y_2O_3 system.

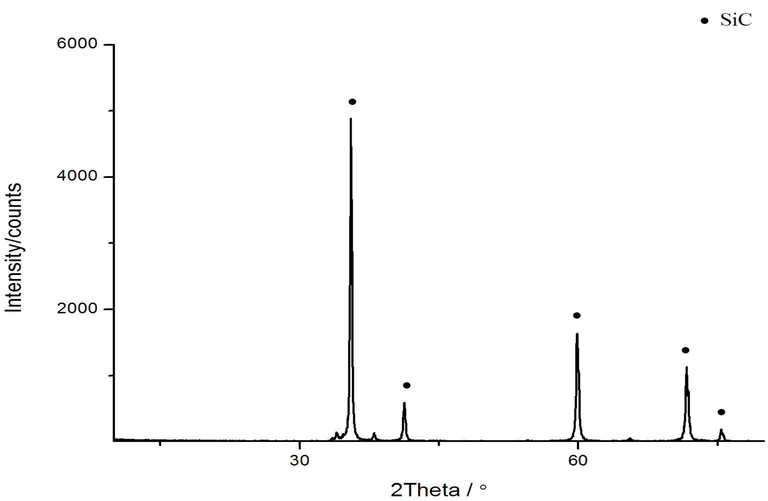

Figure 2. XRD pattern of starting powder β-SiC at 1450˚C/2 hrs in Ar.

system is very low (1370˚C) [18]. Even so, understanding the high temperature react behavior of SiO_2 and its effect on the phase relations in the present system, as well as, further on the manufacture of SiC ceramic is required. For this reason, SiO_2 was chosen as a member of the quaternary Y_2O_3-Al_2O_3-SiC-SiO_2 system. Samples were sintered at 1500˚C - 1600˚C/2 hrs in Ar. **Figure 3** is XRD patterns of YASS0133 and YASS0421 sintered samples. From **Figure 3**, it could be found that both samples have $Al_6Si_2O_{13}$ (mullite) and SiC formed. The different is YASS0133 contains SiO_2, butYASS0421 contains Al_2O_3. It can be confirmed that SiC can form a tie-line with mullite. Three phases could be identified in YASS0133 as SiC, SiO_2 and mullite. While in YASS0421 three phase coexistence of SiC, Al_2O_3 and$Al_6Si_2O_{13}$ could be found. Therefore the subsolidus phase diagram of Al_2O_3-SiC-SiO_2 system can be presented as **Figure 4**. **Figure 5** is XRD patterns of specimens of YASS 1422 and YASS 3211. In **Figure 5** it was found that the two samples both have SiC and yttrium silicates phases. The different is YASS1422 have Al_2O_3 and YAG ($Y_3Al_5O_{12}$), while YASS3211 have YAM ($Y_4Al_2O_9$) and YAP ($YAlO_3$). It indicates two points, one is mulite can not has a tie-line with SiC. The other is SiO_2 was almost used up to form yttrium silicates. Four phases could be identified as YAG, Al_2O_3, SiC and $Y_2Si_2O_7$ in YASS1422. And four phases of YAM, YAP, SiC and Y_2SiO_5 were found in YASS3211 specimen. **Figure 6** is XRD patterns

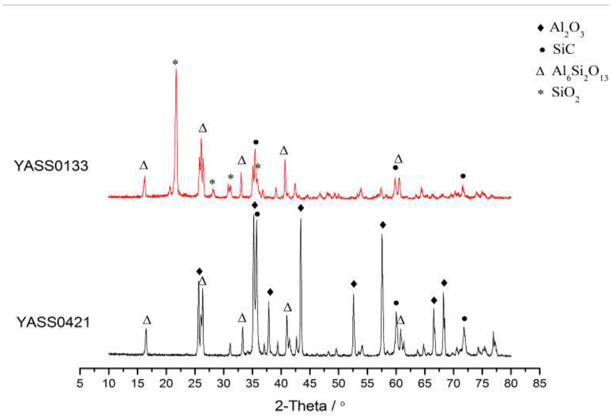

Figure 3. XRD patterns ofYASS0421 (a) and YASS0133 (b) specimens.

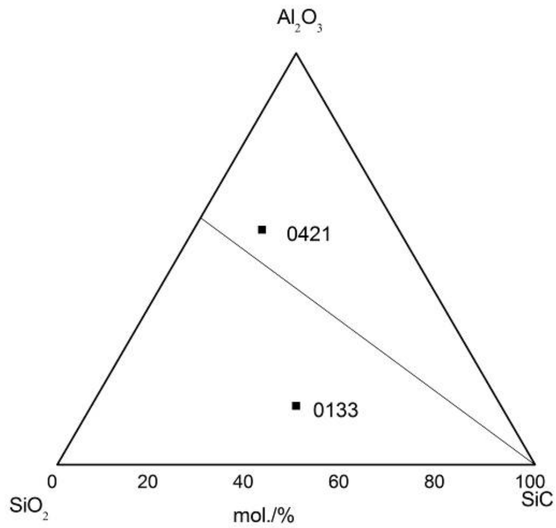

Figure 4. Subsolidus phase diagram of Al_2O_3-SiC-SiO_2 system.

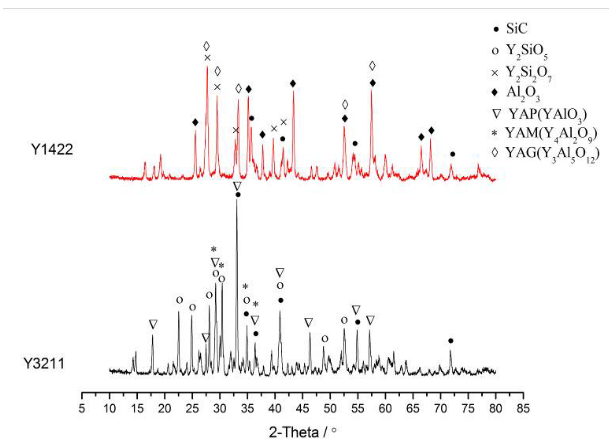

Figure 5. XRD patterns of YASS1422 and YASS3211 specimens.

of YASS1315 and YASS1414 samples. Phases identified by XRD analysis in YASS1315 are SiC, $Al_6Si_2O_{13}$, SiO_2 and $Y_2Si_2O_7$, for YASS1414 they are SiC, Al_2O_3, $Al_6Si_2O_{13}$ and $Y_2Si_2O_7$. In **Figure 6** we found that the two samples both have $Y_2Si_2O_7$, $Al_6Si_2O_{13}$ and SiC. The different is, YASS1315 has SiO_2, YASS1414 has Al_2O_3. It is indicated that SiO_2 plays the role of forming mullite and yttrium silicates and leading to establish the phase equilibria with yttrium aluminates and SiC. The formation of above several four phase coexistence involving both yttrium silicates and mullite extends the ternary system SiC-Al_2O_3-Y_2O_3 into the quaternary system included SiO_2. Combing the phase diagrams of Al_2O_3-Y_2O_3-SiC system with Al_2O_3-Y_2O_3-SiO_2 system [18] the tentative phase diagram of quaternary SiC-Al_2O_3-Y_2O_3-SiO_2 (Si-Al-Y-O-C) system can be presented as **Figure 7** which shows that the triangle equilibrium relations of SiC with three yttrium aluminates extend to the tetrahedral equilibrium relations with three yttrium silicates $Y_2Si_2O_7$, Y_2SiO_5 and mullite, respectively. Besides, it should be indicated that the tie-line YAP-Y_2SiO_5 in present work is better stead of YAG-YAMss tie-line in the phase diagram of Y_2O_3-Al_2O_3-SiO_2 system [18]. The possible YAMss solid-solution is not measured in present work.

4. Conclusion

The subsolidus phase diagram of the Al_2O_3-SiC-SiO_2 system was given. The phase relations in the SiC-Al_2O_3-

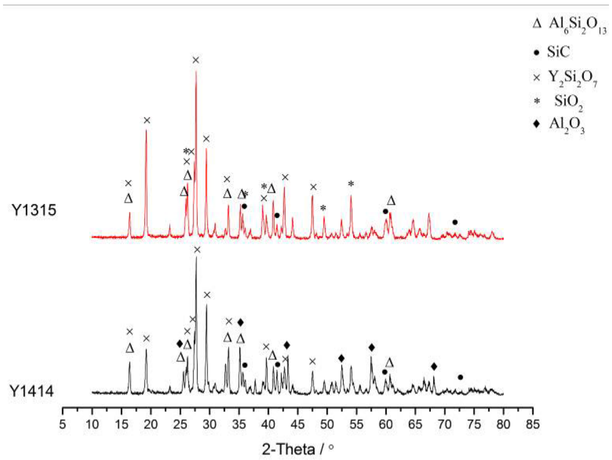

Figure 6. XRD patterns of YASS1414 and YASS1315 specimens.

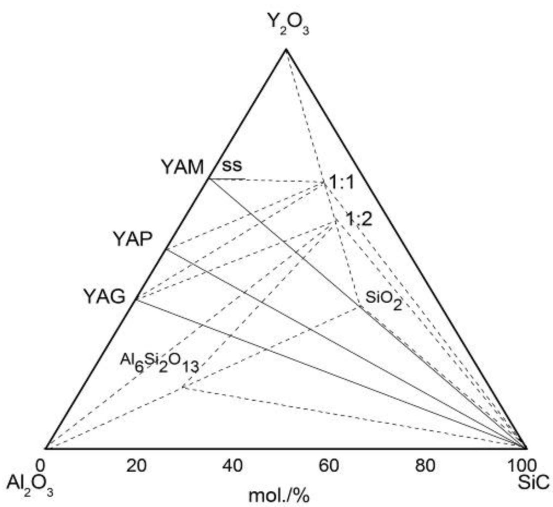

Figure 7. Tentative phase diagram of quaternary system SiC-Al_2O_3-Y_2O_3-SiO_2 (Si-Al-Y-O-C).

Y_2O_3-SiO_2 system were established. The tentative phase diagram of this quaternary system was presented in which SiO_2 plays the role of forming mullite and yttrium silicates and leading to establish the phase equilibria of yttrium silicates with yttrium aluminates and SiC.

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