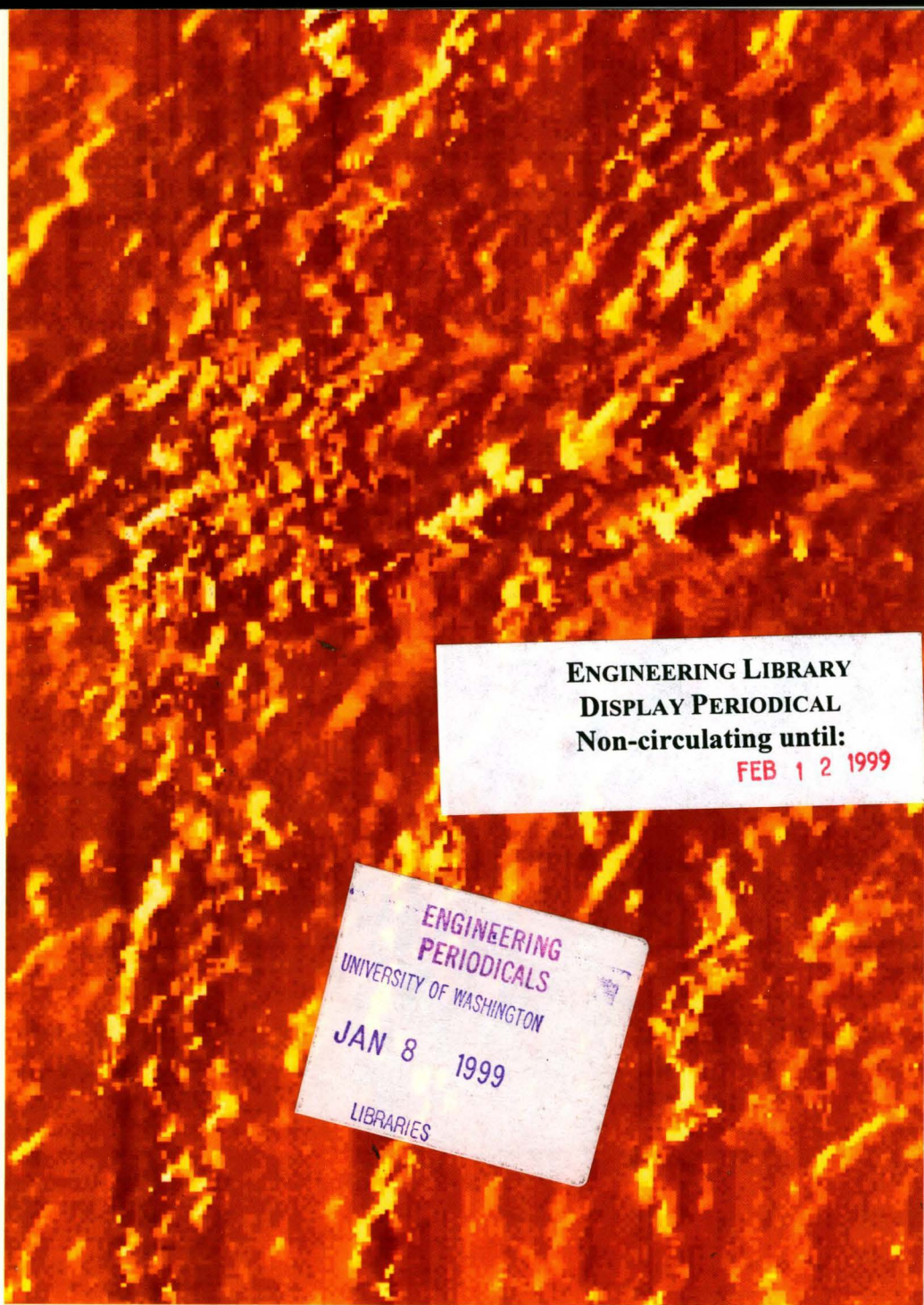


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# Journal

of the American Ceramic Society  
Incorporating Advanced Ceramic Materials and Communications  
Volume 81 Number 12 December 1998

GE-1024.001

# Journal

of the American Ceramic Society  
Incorporating Advanced Ceramic Materials and Communications

ISSN 0002-7820  
Volume 81  
Number 12  
December 1998

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**COVER PHOTOGRAPH:** Magnetic force microscopy image of superconducting current flow ( $J = 100 \text{ A/cm}^2$ ) in a thick film at 60–65 K. See feature article by Dawn Bonnell, "Local Structure and Properties of Oxide Surfaces: Scanning Probe Analyses of Ceramics," this issue, pages 3049–70.

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# Improvement in Mechanical Properties of Powder-Processed MoSi<sub>2</sub> by the Addition of Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub>

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**Additions of 1–20 mol% Sc<sub>2</sub>O<sub>3</sub> or Y<sub>2</sub>O<sub>3</sub> to MoSi<sub>2</sub> eliminate glassy SiO<sub>2</sub>, which improves mechanical properties at both ambient and high temperatures. In particular, only 1 mol% ScO<sub>3</sub> additions dramatically enhance three-point bending strength from 521 to 1081 MPa. Vickers hardness, Young's modulus, fracture toughness, and high-temperature strength are also improved by this low level of additive. The improvement of mechanical properties is attributed to the formation of crystalline silicates: Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, Y<sub>2</sub>SiO<sub>5</sub>, and Y<sub>4</sub>Si<sub>3</sub>O<sub>12</sub>, which are analyzed by XRD, SEM-EDS, and TEM-EDS methods.**

## I. Introduction

MOLYBDENUM DISILICIDE, MoSi<sub>2</sub>, is an attractive candidate material for high-temperature structural applications because of its high melting point (2030°C), moderate density (6.27 g/cm<sup>3</sup>), and excellent oxidation resistance.<sup>1,2</sup> Because of its intrinsically low ductility below the brittle-to-ductile transition temperature (BDTT: 900°–1400°C, depending on grain size and impurities),<sup>1,3</sup> powder processes are widely used for fabricating bulk MoSi<sub>2</sub> materials.<sup>4</sup> Because MoSi<sub>2</sub> powders that are prepared by fusing and crushing always contain a certain amount of oxygen, a monolithic MoSi<sub>2</sub> sintered body is actually composed of MoSi<sub>2</sub>, Mo<sub>5</sub>Si<sub>3</sub>, and glassy SiO<sub>2</sub> phases. Mo<sub>5</sub>Si<sub>3</sub>, a low-silicide phase, is formed by a deviation from stoichiometry. With carbon impurity, the Mo<sub>≤5</sub>Si<sub>3</sub>C<sub>≤1</sub> Nowotny phase can also appear.<sup>5–9</sup> Inclusions, such as low silicides (here we include the Mo<sub>≤5</sub>Si<sub>3</sub>C<sub>≤1</sub> phase as a type of low silicide) and glassy SiO<sub>2</sub>, affect various properties of MoSi<sub>2</sub>. Low silicides have less oxidation resistance than MoSi<sub>2</sub>,<sup>10</sup> but they can strengthen MoSi<sub>2</sub> as second-phase reinforcements.<sup>11</sup> While dispersion of low silicides in MoSi<sub>2</sub> compacts positively affects mechanical properties, glassy SiO<sub>2</sub> always affects them negatively.<sup>11,12</sup> The glassy SiO<sub>2</sub> phase forms discrete regions in MoSi<sub>2</sub> sintered bodies mainly at triple points,<sup>8–9,11–13</sup> as opposed to Si<sub>3</sub>N<sub>4</sub>, where glassy SiO<sub>2</sub> wets two-grain boundaries and triple points as layers. Nevertheless, glassy SiO<sub>2</sub> in MoSi<sub>2</sub> sintered bodies promotes creep by grain-boundary sliding, as in Si<sub>3</sub>N<sub>4</sub>, as it softens at high temperatures. Thus, glassy SiO<sub>2</sub> degrades mechanical properties, not only at room temperature, but also at high temperatures.<sup>12</sup>

The approaches to eliminate the siliceous grain-boundary phase can be classified into five categories: (1) reducing oxygen content in starting MoSi<sub>2</sub> powders, e.g., in-flight treatment

in an argon–hydrogen plasma<sup>4</sup> and HF treatment;<sup>14</sup> (2) deoxidation during sintering by, e.g., carbothermal reduction<sup>6–7,15</sup> and hydrogen reduction;<sup>16</sup> (3) metathesis reaction during sintering, e.g., replacing SiO<sub>2</sub> by Al<sub>2</sub>O<sub>3</sub> through aluminum metal addition;<sup>17–19</sup> (4) predominant oxidation of alloying element, e.g., aluminum, in starting powders;<sup>9,20</sup> and (5) double oxide formation during sintering.<sup>8</sup> For category (5), we have already reported that ZrO<sub>2</sub> (3Y) additions effectively decrease glassy SiO<sub>2</sub> in MoSi<sub>2</sub> sintered bodies by forming the more stable and crystalline double oxide ZrSiO<sub>4</sub>.<sup>8</sup> Various mechanical properties are considerably improved by ZrSiO<sub>4</sub> formation and perhaps by the transformation toughening of unreacted ZrO<sub>2</sub>.

In this study, we extended the *in situ* double oxide formation method to other oxide additives. C-rare-earth-structured oxides, Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub>, were selected as additives because they have high reactivity with SiO<sub>2</sub> to form very refractory double oxides, i.e., Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> (mp, 1860°C), Sc<sub>2</sub>SiO<sub>5</sub> (mp, 1950°C), Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> (peritectic point, 1775°C), and Y<sub>2</sub>SiO<sub>5</sub> (mp, 1980°C).<sup>21,22</sup> To clarify the effects of grain-boundary phases, we preferred to use a fine MoSi<sub>2</sub> powder (~1 μm) with rather high oxygen content (1.21 wt%). R<sub>2</sub>O<sub>3</sub> (R = Sc and Y) powders (1–20 mol%) were added to the MoSi<sub>2</sub> powder so the effects of double oxide formation and second phase dispersion of excess, unreacted R<sub>2</sub>O<sub>3</sub> in the MoSi<sub>2</sub> bodies could be examined.

Morgan *et al.*<sup>23,24</sup> previously studied the effects of Sc<sub>2</sub>O<sub>3</sub> addition on Si<sub>3</sub>N<sub>4</sub> and reported that oxidation resistance and creep resistance were significantly improved by the easy crystallization of the Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> phase (also known as the mineral thortveitite); these observations were confirmed in later works.<sup>25,26</sup> We conjectured similar positive effects for the Mo–Si–Sc–O system because both Si<sub>3</sub>N<sub>4</sub> and MoSi<sub>2</sub> have siliceous grain-boundary phases that greatly affected various mechanical properties. For Y<sub>2</sub>O<sub>3</sub>, Kulenko *et al.*<sup>27</sup> reported on the MoSi<sub>2</sub>–Y<sub>2</sub>O<sub>3</sub> system prepared by pressureless sintering. The main purpose of their study was to increase the electrical resistivity of MoSi<sub>2</sub> bodies without necessarily affecting the mechanical properties. They examined relatively high amounts of Y<sub>2</sub>O<sub>3</sub> (10%–50%, presumably weight fraction, as the authors did not clearly state), and mechanical properties were scarcely mentioned. Research on MoSi<sub>2</sub>–Y<sub>2</sub>O<sub>3</sub> composites was also conducted by Zalkind *et al.*,<sup>28</sup> who demonstrated that the MoSi<sub>2</sub>–Y<sub>2</sub>O<sub>3</sub> composites showed good corrosion resistance to melts of fuel slag, indicating the potential benefits of the MoSi<sub>2</sub>–Y<sub>2</sub>O<sub>3</sub> system.

This initial investigation shows that even minor amounts of Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> additions (1 mol%) are effective in greatly improving mechanical properties of MoSi<sub>2</sub>-based materials.

## II. Experimental Procedure

### (1) Sample Preparation

Starting materials were MoSi<sub>2</sub> (1.01 μm, Japan New Metals Co., Ltd.), Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> (~2 μm, Kojundo Chemical Laboratory Co., Ltd.). The R<sub>2</sub>O<sub>3</sub> additions in this investigation were restricted to 1–20 mol% because a larger amount of oxides

J. J. Petrovic—contributing editor

Manuscript No. 190747. Received August 28, 1997; approved February 23, 1998. Supported by the Japan Ministry of Education, Science, Sports, and Culture under Grant-in-Aid for Science Research No. 08044150 and 09450262, and for JSPS Research Fellowships.

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might degrade the high-temperature mechanical properties. The chemical analysis of the starting MoSi<sub>2</sub> powder is shown in Table I. Spectrochemical analysis revealed that the major impurities were 1.21 wt% oxygen, 0.20 wt% carbon, and 0.21 wt% iron. The high oxygen level was associated with the fine particle size (1.01 μm) of the MoSi<sub>2</sub> powder. Table II shows the composition of the MoSi<sub>2</sub> starting powder calculated from Table I, assuming that the phases in the powder were MoSi<sub>2</sub>, Mo<sub>5</sub>Si<sub>3</sub>, glassy SiO<sub>2</sub>, free carbon, and Fe<sub>2</sub>O<sub>3</sub>. This result assumed that 1.21 wt% oxygen in the MoSi<sub>2</sub> powder was equivalent to ~6 vol% of glassy SiO<sub>2</sub>. MoSi<sub>2</sub> and additive oxide powders were wet ball-milled with ZrO<sub>2</sub> balls for 24 h in *n*-hexane, which was used to avoid the mechanochemical oxidation by usual alcohol solvents, as reported for Si<sub>3</sub>N<sub>4</sub>.<sup>29</sup> Nominal compositions of the powder mixture are summarized in Table III. The mixture was packed into carbon dies, coated with BN, and hot-pressed at 1600°C under an applied pressure of 30 MPa for 1 h under vacuum. The sintered disks, 44 mm in diameter and 5.0 mm thick, were cut, ground, and polished into rectangular bars 4 mm × 3 mm × 36 mm in size for three-point bending test.

## (2) Evaluation Methods

Phase identification was conducted by X-ray diffraction (XRD) analysis. To minimize the anisotropic orientation effect, liquid paraffin was mixed with the pulverized specimen. The microstructure was characterized by scanning electron microscopy (SEM, Model S-5000, Hitachi, Tokyo, Japan) with energy dispersive X-ray spectrometry (EDS, Model EMAX-5770, Horiba, Tokyo, Japan) with the samples precoated with carbon. Transmission electron microscopy (TEM, Model H-8100, Hitachi) with EDS (Model PV9900, Phillips, Eindhoven, The Netherlands) was also used for the microstructural characterization. The bulk density was measured by the Archimedes method in toluene at room temperature; temperature corrections were always conducted for the density measurements. Young's modulus was determined by the resonance vibration method with the thickness correction. Five or six specimens in each case were subjected to three-point bending test (span: 30 mm, in air) to determine fracture strength up to 1300°C. The bending load was applied parallel to the hot-pressing axis with a crosshead speed of 0.5 mm/min. Hardness ( $H_V$ ) was evaluated by Vickers indentation under conditions of 98 N load with loading duration of 15 s in air. Fracture toughness ( $K_{IC}$ ) at room temperature was determined simultaneously by the indentation fracture (IF) method.<sup>30</sup>  $K_{IC}$  values were calculated using the following empirical equation for a median crack:

$$K_{IC} = 0.203(c/a)^{-3/2}H_V a^{1/2} \quad (1)$$

where  $c$  and  $a$  are the lengths of a median crack and half of a diagonal of an indentation, respectively.

## III. Results

### (1) Phase Analysis of Sc<sub>2</sub>O<sub>3</sub>-Containing Materials

Figure 1 shows the XRD patterns for (a) MoSi<sub>2</sub> with added Sc<sub>2</sub>O<sub>3</sub> and (b) MoSi<sub>2</sub> with added Y<sub>2</sub>O<sub>3</sub> hot-pressed bodies with increasing additive amounts. Because the XRD intensities of MoSi<sub>2</sub> are quite strong in comparison with those of other phases, the peak tops are truncated. XRD analysis of the phases in the sintered bodies is summarized in Table IV. XRD analysis indicates that the monolithic MoSi<sub>2</sub> body is composed of MoSi<sub>2</sub> (tetragonal, C11<sub>b</sub>), a small amount of Mo<sub>5</sub>Si<sub>3</sub>C<sub>5</sub>

**Table II. Calculated Composition of the MoSi<sub>2</sub> Starting Powder**

	MoSi <sub>2</sub>	Mo <sub>5</sub> Si <sub>3</sub> <sup>†</sup>	Glassy SiO <sub>2</sub>	Free carbon <sup>‡</sup>	Fe <sub>2</sub> O <sub>3</sub>
Content (mol%)	91.20	0.90	5.10	2.50	0.30
Content (wt%)	94.00	3.40	2.10	0.20	0.30
Content (vol%)	90.90	2.50	5.70	0.50	0.40
Density (g/cm <sup>3</sup> )	6.27	8.19	2.22	2.25	5.24

<sup>†</sup>Mo<sub>5</sub>Si<sub>3</sub> increases with oxidation and reacts with carbon to form Mo<sub>5</sub>Si<sub>3</sub>C<sub>5</sub>.  
<sup>‡</sup>Some carbon can dissolve in SiO<sub>2</sub> glass.

(hexagonal, D8<sub>8</sub>), and a trace of SiO<sub>2</sub> (cristobalite). The Mo<sub>5</sub>Si<sub>3</sub>C<sub>5</sub>, Nowotny phase,<sup>31</sup> is the only stable Mo-Si-C ternary phase. TEM selected area diffraction analysis reveals that most of the SiO<sub>2</sub> exists as glassy phase; however, the cristobalite observed by XRD must be attributed to partial crystallization of glassy SiO<sub>2</sub>. In the 1–2-mol%-Sc<sub>2</sub>O<sub>3</sub>-MoSi<sub>2</sub> sintered bodies, Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, thortveitite, (Sc<sub>2</sub>O<sub>3</sub>:SiO<sub>2</sub> = 1:2) is identified. Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> is a stable refractory compound (mp, 1860°C)<sup>21</sup> in the Sc<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> binary system. Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> is reported to form readily in the hot-pressed Si<sub>3</sub>N<sub>4</sub>-SiO<sub>2</sub>-Sc<sub>2</sub>O<sub>3</sub> system also.<sup>23–26</sup> The cristobalite peak around  $2\theta = 22^\circ$  decreases (as expected) with increasing Sc<sub>2</sub>O<sub>3</sub> content (0–5 mol%) as Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> forms with the inevitable reaction with glassy SiO<sub>2</sub>. For the MoSi<sub>2</sub> with 5 mol% Sc<sub>2</sub>O<sub>3</sub>, most of the SiO<sub>2</sub> in the sintered body is converted to the silicate, because the cristobalite peak disappears and small peaks from unreacted Sc<sub>2</sub>O<sub>3</sub> are observed. With higher Sc<sub>2</sub>O<sub>3</sub> additions (10–20 mol%), the constituent phases are the same as in the 5-mol%-Sc<sub>2</sub>O<sub>3</sub>-MoSi<sub>2</sub> case; the amount of unreacted Sc<sub>2</sub>O<sub>3</sub> simply increases. In the Sc<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> binary system,<sup>21</sup> there should be another silicate, Sc<sub>2</sub>SiO<sub>5</sub> (Sc<sub>2</sub>O<sub>3</sub>:SiO<sub>2</sub> = 1:1), when Sc<sub>2</sub>O<sub>3</sub> content increases. However, the Sc<sub>2</sub>SiO<sub>5</sub> phase is never observed in this experiment, as discussed later.

### (2) Phase Analysis of Y<sub>2</sub>O<sub>3</sub>-Containing Materials

For MoSi<sub>2</sub> with 1–2 mol% Y<sub>2</sub>O<sub>3</sub>, the formation of the 1:2 silicate, i.e., Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, is confirmed, similar to Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> formation in the MoSi<sub>2</sub>-SiO<sub>2</sub>-Sc<sub>2</sub>O<sub>3</sub> system. Three polymorphs of Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> phase are identified:  $\beta$ -phase (keiviite-(Y)),  $\gamma$ -phase, and  $\delta$ -phase.  $\beta$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> has the thortveitite-type crystal structure and is stable at high temperatures around the hot-pressing temperature (1600°C).<sup>22,32</sup> Dinger *et al.*<sup>33</sup> studied the crystallization behavior of the glasses in the Y<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-AlN system and reported that  $\gamma$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>,  $\delta$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, and  $\beta$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> phases were observed, with  $\gamma$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> and  $\delta$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> crystallizing at ~1000° and 1200°C, respectively, and  $\beta$ -Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> crystallizing at higher temperatures. Because the hot-pressing time in this study is not long, it is perhaps not surprising that metastable polymorphs of Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> remain in the sintered bodies. The intensity of the cristobalite peak at  $2\theta = 22^\circ$  also decreases with increasing Y<sub>2</sub>O<sub>3</sub> content (0–5 mol%), similar to the Sc<sub>2</sub>O<sub>3</sub> case. A trace of Y<sub>2</sub>O<sub>3</sub> is also identified in the 1–2-mol%-Y<sub>2</sub>O<sub>3</sub>-MoSi<sub>2</sub>, despite the presence of free SiO<sub>2</sub> presumably because of diffusion limitation and separation by the majority MoSi<sub>2</sub> matrix.

For MoSi<sub>2</sub> with 5 mol% Y<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, unreacted SiO<sub>2</sub> and Y<sub>2</sub>O<sub>3</sub> are observed in addition to MoSi<sub>2</sub> and Mo<sub>5</sub>Si<sub>3</sub>C<sub>5</sub>. Furthermore, a notable peak at  $2\theta = 32^\circ$  is observed, which was never seen in the other specimens, and is identified as the 2:3 silicate Y<sub>4</sub>Si<sub>3</sub>O<sub>12</sub>. According to the published phase diagram,<sup>22</sup> this phase is not stable below 1650°C; nevertheless, it appears here. Although it may cast some doubt on the veracity of this early diagram, it is not surprising that the formation of Y<sub>4</sub>Si<sub>3</sub>O<sub>12</sub> takes place, because the eutectoid temperature is only 50°C higher than the hot-pressing temperature (1600°C); thus, some impurity, pressure, and local temperature gradient (e.g., by a reaction heat) presumably affects the reactivity. For compacts with higher Y<sub>2</sub>O<sub>3</sub> additions (10–20 mol%), SiO<sub>2</sub> and Y<sub>4</sub>Si<sub>3</sub>O<sub>12</sub> disappear, and Y<sub>2</sub>SiO<sub>5</sub> (Y<sub>2</sub>O<sub>3</sub>:SiO<sub>2</sub> = 1:1) forms. The equivalent 1:1 silicate is not observed in the MoSi<sub>2</sub>-SiO<sub>2</sub>-Sc<sub>2</sub>O<sub>3</sub> system.

**Table I. Chemical Analysis of the MoSi<sub>2</sub> Starting Powder**

	Mo <sup>†</sup>	Si	O	C	Fe
Content (wt%)	62.18	36.20	1.21	0.20	0.21

<sup>†</sup>Molybdenum content was calculated so that total content was 100%.

Table III. Nominal Compositions of the Powder Mixture<sup>†</sup>

Sample No.	Composition (mol%)			Composition (wt%) <sup>‡</sup>			Composition (vol%) <sup>§</sup>		
	MoSi <sub>2</sub>	Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>	MoSi <sub>2</sub>	Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>	MoSi <sub>2</sub>	Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>
1	100			100.0			100.0		
2	99	1		99.1	0.9		98.5	1.5	
3	98	2		98.2	1.8		97.1	2.9	
4	95	5		95.4	4.6		92.8	7.2	
5	90	10		90.8	9.2		85.9	14.1	
6	85	15		86.2	13.8		79.3	20.7	
7	80	20		81.5	18.5		73.0	27.0	
8	99		1	98.5		1.5	98.2		1.8
9	98		2	97.1		2.9	96.4		3.6
10	95		5	92.8		7.2	91.1		8.9
11	90		10	85.8		14.2	83.0		17.0
12	85		15	79.2		20.8	75.4		24.6
13	80		20	72.9		27.1	68.4		31.6

<sup>†</sup>Assuming that the starting powders are pure MoSi<sub>2</sub>, Sc<sub>2</sub>O<sub>3</sub>, and Y<sub>2</sub>O<sub>3</sub>. <sup>‡</sup>Formula weight: MoSi<sub>2</sub> = 152.111, Sc<sub>2</sub>O<sub>3</sub> = 137.91, and Y<sub>2</sub>O<sub>3</sub> = 225.81. <sup>§</sup>X-ray density:  $D_{\text{MoSi}_2} = 6.27 \text{ g/cm}^3$ ,  $D_{\text{Sc}_2\text{O}_3} = 3.840 \text{ g/cm}^3$ , and  $D_{\text{Y}_2\text{O}_3} = 5.032 \text{ g/cm}^3$ .

### (3) Microstructure

Figure 2 shows typical SEM photographs on the polished surfaces of monolithic MoSi<sub>2</sub> and MoSi<sub>2</sub> with R<sub>2</sub>O<sub>3</sub> additives: (a) monolithic MoSi<sub>2</sub>; (b) 1, (c) 5, (d) 10, and (e) 20 mol% Sc<sub>2</sub>O<sub>3</sub>; (f) 1, (g) 5, (h) 10, and (i) 20 mol% Y<sub>2</sub>O<sub>3</sub>. In these figures, the dark regions are composed of the mixed oxides detailed in the phase analysis (not pores). The regions with light-gray contrast are the Mo<sub>5</sub>Si<sub>3</sub>C<sub>≦1</sub> phase. In the monolithic MoSi<sub>2</sub> sintered body, glassy SiO<sub>2</sub> particles with spherical or irregular shape are isolated, unlike in Si<sub>3</sub>N<sub>4</sub> sintered bodies, where glassy SiO<sub>2</sub> is distributed as a grain-boundary film. The Mo<sub>5</sub>Si<sub>3</sub>C<sub>≦1</sub> phase is usually found next to glassy SiO<sub>2</sub> regions. This microstructure may be explained as follows: The starting MoSi<sub>2</sub> powder has oxidized glassy SiO<sub>2</sub> on the surfaces and a thin Mo<sub>5</sub>Si<sub>3</sub> region below the SiO<sub>2</sub> layer.<sup>34</sup> During hot-pressing, the glassy SiO<sub>2</sub> becomes concentrated mainly at triple points, and the Mo<sub>5</sub>Si<sub>3</sub>C<sub>≦1</sub> is formed by reactions between Mo<sub>5</sub>Si<sub>3</sub> and impurity carbon.<sup>6,7</sup> The carbon impurity arises from the powder used, the ball-milling solvent, and the CO gas from a die and a furnace element. SEM-EDS study reveals that scandium and yttrium elements appear predominantly in the SiO<sub>2</sub> pockets. Thus, it is inferred that the additives and SiO<sub>2</sub> react together at grain-boundary pockets. With higher additive content, the oxide phases become continuous, as seen in Figs. 2(d), (e), (h), and (i).

### (4) Density

Figure 3 shows the variation of density of hot-pressed bodies with Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> addition. The density of the monolithic MoSi<sub>2</sub> compact is 5.92 g/cm<sup>3</sup>. This value is smaller than the theoretical density of the MoSi<sub>2</sub> phase determined by XRD (6.27 g/cm<sup>3</sup>). Because almost no pores are observed by SEM for the MoSi<sub>2</sub> sintered body, the lower density is attributed to the formation of glassy SiO<sub>2</sub> with its rather low density (2.20 g/cm<sup>3</sup>). For the Y<sub>2</sub>O<sub>3</sub>-added system, density increases then decreases with Y<sub>2</sub>O<sub>3</sub> content. This suggests the formation of a higher-density phase compared to glassy SiO<sub>2</sub>. With Sc<sub>2</sub>O<sub>3</sub> addition, the density curve has a maximum at 5 mol% Sc<sub>2</sub>O<sub>3</sub>, also suggesting the formation of a higher-density phase. The density dependence is discussed later in detail.

### (5) Mechanical Properties

Young's modulus dependence on Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> content is shown in Fig. 4. The Young's modulus of the monolithic MoSi<sub>2</sub> compact is 372 GPa. This value is smaller than the theoretical Young's modulus of polycrystalline MoSi<sub>2</sub> calculated from the single crystal (440 GPa).<sup>35</sup> In general, Young's modulus is strongly affected by the presence of porosity.<sup>36</sup> However, because almost no pores are observed in the sintered body as mentioned above, the decrease in Young's modulus is explained by the formation of glassy SiO<sub>2</sub> phase. For both

Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> additions, Young's modulus indicates the maximum values at 5 mol% addition, the minimum additive composition to remove glassy SiO<sub>2</sub> in the sintered bodies. Because Young's moduli of Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> are smaller than that of MoSi<sub>2</sub>, 5 mol% addition results in the maximum values. The Young's modulus dependence is discussed further in the next section.

Figure 5 shows the variation of fracture strength with additive content. The strength value of monolithic MoSi<sub>2</sub> is 521 MPa. Fracture strength increases then decreases with increasing additive content. For both Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> addition, maximum fracture strength is obtained for only 1 mol% addition. The strength values of MoSi<sub>2</sub> with 1 mol% Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> are 1081 and 1028 MPa, respectively. These values are extremely high for MoSi<sub>2</sub>-based materials.<sup>2</sup> At higher oxide phase contents, fracture strength decreases, perhaps because the oxides become connected in a larger potential flaw fracture origin. For 1 mol% addition, the total amount of oxide phase is still small, and oxide regions are discrete; the glassy SiO<sub>2</sub> has sufficiently reacted with the additive to form 1:2 silicate, apparently causing strengthening. Because only 1 mol% R<sub>2</sub>O<sub>3</sub> additive is sufficient to greatly improve fracture strength, the use of expensive Sc<sub>2</sub>O<sub>3</sub> may become more viable from a cost standpoint.

Figure 6 shows the variation of Vickers hardness with additive content. The change of Vickers hardness with additive content resembles that of Young's modulus. Hardness shows its maximum value at 5 mol% for both Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> addition. This is attributed to the approach to total crystallization of the glassy phase. The decrease in hardness with higher additive content is due to the softness of unreacted Y<sub>2</sub>O<sub>3</sub> or Sc<sub>2</sub>O<sub>3</sub> additive.

Fracture toughness dependence on additive content is shown in Fig. 7. The variation of fracture toughness with additive content is similar to that of Young's modulus and Vickers hardness for the same reasons. Too much additive also decreases fracture toughness, also for the same reasons.

Figure 8 shows high-temperature strength of MoSi<sub>2</sub> with 1 mol% Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub>. At >1100°C, both materials display plastic deformation without fracture. The values of two different monolithic MoSi<sub>2</sub> samples are also shown in the figure for reference: fine MoSi<sub>2</sub> is fabricated by hot-pressing a fine MoSi<sub>2</sub> powder (1.0 μm, almost identical to this experiment) at 1500°C, whose grain size is ~5 μm,<sup>37</sup> and coarse MoSi<sub>2</sub> is made by hot-pressing a coarse powder (several tens of micrometers, typical values for conventional MoSi<sub>2</sub>) at 1700°C.<sup>38</sup> Because coarse-grained MoSi<sub>2</sub> contains less glassy phase, it maintains almost the same, albeit low, strength (~150 MPa) up to 1200°C. On the other hand, fine-grained MoSi<sub>2</sub> shows improved strength for the entire temperature range compared to coarse-grained MoSi<sub>2</sub>, although strength degradation is observed above 800°C. The improvement of strength up to 800°C

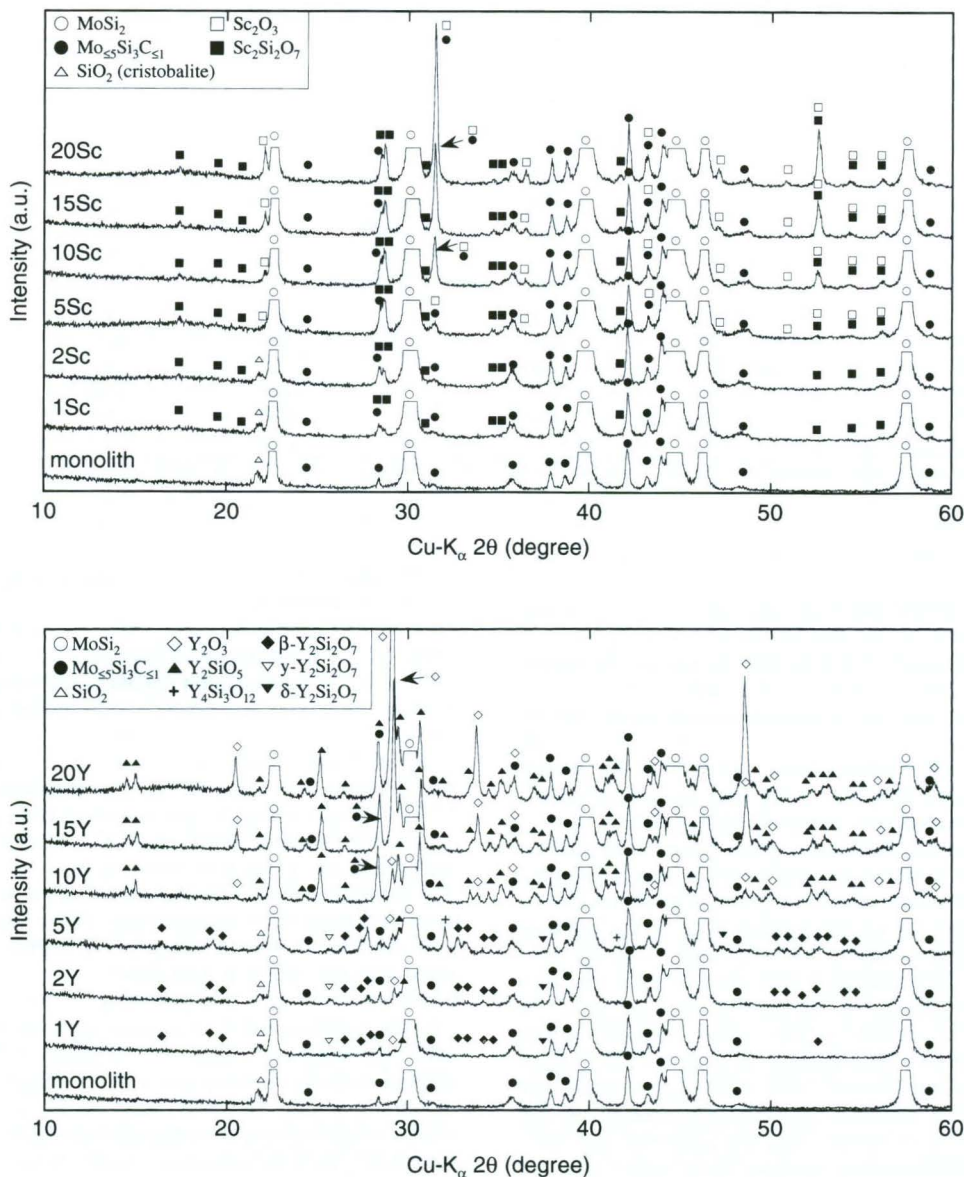


Fig. 1. XRD patterns for (a)  $\text{MoSi}_2\text{-SiO}_2\text{-Sc}_2\text{O}_3$  and (b)  $\text{MoSi}_2\text{-SiO}_2\text{-Y}_2\text{O}_3$  systems.

Table IV. Phase Contents of the Hot-Pressed Products in the  $\text{MoSi}_2\text{-R}_2\text{O}_3$  ( $\text{R} = \text{Sc}$  and  $\text{Y}$ ) Systems

Sample No.	Nominal starting composition (mol%)	Phases in product (identified by XRD)
1	$\text{MoSi}_2$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{SiO}_2^\dagger$
2, 3	$\text{MoSi}_2 + 1.2\% \text{Sc}_2\text{O}_3$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{SiO}_2^\dagger$ , $\text{Sc}_2\text{Si}_2\text{O}_7$
4	$\text{MoSi}_2 + 5\% \text{Sc}_2\text{O}_3$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{Sc}_2\text{Si}_2\text{O}_7$ , (trace $\text{Sc}_2\text{O}_3$ )
5, 6, 7	$\text{MoSi}_2 + 10, 15, 20\% \text{Sc}_2\text{O}_3$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{Sc}_2\text{Si}_2\text{O}_7$ , $\text{Sc}_2\text{O}_3$
8, 9	$\text{MoSi}_2 + 1.2\% \text{Y}_2\text{O}_3$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{SiO}_2^\dagger$ , $\text{Y}_2\text{Si}_2\text{O}_7$ , (trace $\text{Y}_2\text{O}_3$ )
10	$\text{MoSi}_2 + 5\% \text{Y}_2\text{O}_3$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{SiO}_2^\dagger$ , $\text{Y}_2\text{Si}_2\text{O}_7$ , $\text{Y}_4\text{Si}_2\text{O}_7$ , (trace $\text{Y}_2\text{O}_3$ )
11, 12, 13	$\text{MoSi}_2 + 10, 15, 20\% \text{Y}_2\text{O}_3$	$\text{MoSi}_2$ , $\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}$ , $\text{Y}_2\text{Si}_2\text{O}_7$ , $\text{Y}_2\text{O}_3$

<sup>†</sup>Cristobalite, partially crystallized from glassy  $\text{SiO}_2$ .

is due to the healing effect of the glassy phase (i.e., recovering of surface flaws), while the drop in strength near  $1000^\circ\text{C}$  is mainly due to the glassy phase allowing grain-boundary sliding.  $\text{MoSi}_2$  with 1 mol% oxide additives shows much higher strength values ( $>750$  MPa) up to  $1100^\circ\text{C}$  than conventional  $\text{MoSi}_2$ -based materials. However, because these materials still contain some unreacted glassy  $\text{SiO}_2$ , their strength declines around  $1100^\circ\text{C}$ , similar to the fine-grained  $\text{MoSi}_2$ . Further crystallization of glassy  $\text{SiO}_2$  may somewhat improve strength above  $1200^\circ\text{C}$ .

#### IV. Discussion

##### (1) Difference of Phases Appearing between Two Systems

Interesting differences between the formed silicates with  $\text{Sc}_2\text{O}_3$  and  $\text{Y}_2\text{O}_3$  additions were mentioned in Section III (Fig. 1 and Table IV). Because there are limited thermodynamic data in the  $\text{Sc}_2\text{O}_3\text{-SiO}_2$  and  $\text{Y}_2\text{O}_3\text{-SiO}_2$  binary systems, we are basically limited to phase diagrams. Figures 9(a) and (b) show these for  $\text{Sc}_2\text{O}_3\text{-SiO}_2$ <sup>21</sup> and  $\text{Y}_2\text{O}_3\text{-SiO}_2$  systems.<sup>22</sup>

For 1–5 mol%  $\text{Y}_2\text{O}_3$  addition, some  $\text{SiO}_2$  remains in the

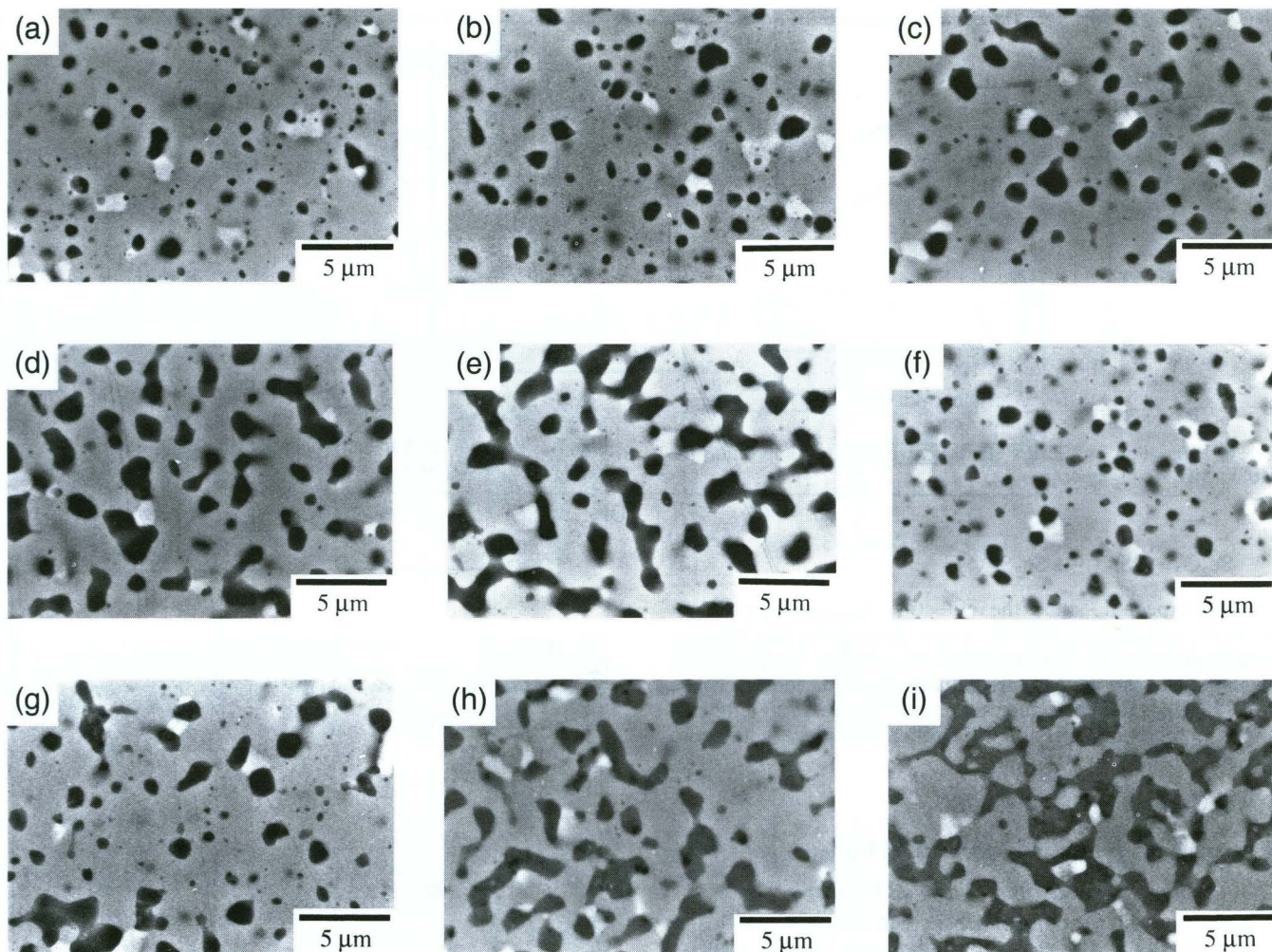


Fig. 2. SEM photographs of polished surfaces: (a) monolithic MoSi<sub>2</sub>; (b) 1, (c) 5, (d) 10, and (e) 20 mol% Sc<sub>2</sub>O<sub>3</sub>; (f) 1, (g) 5, (h) 10, and (i) 20 mol% Y<sub>2</sub>O<sub>3</sub>.

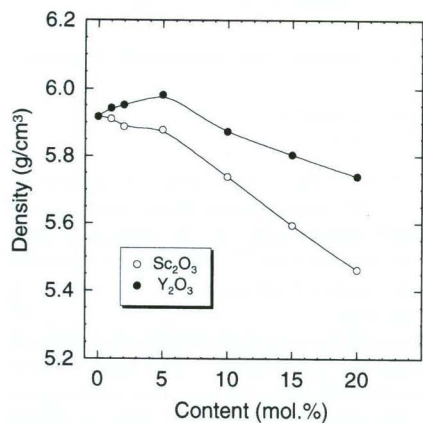


Fig. 3. Variation of density with R<sub>2</sub>O<sub>3</sub> (R = Sc and Y) composition for MoSi<sub>2</sub>-SiO<sub>2</sub>-R<sub>2</sub>O<sub>3</sub> system.

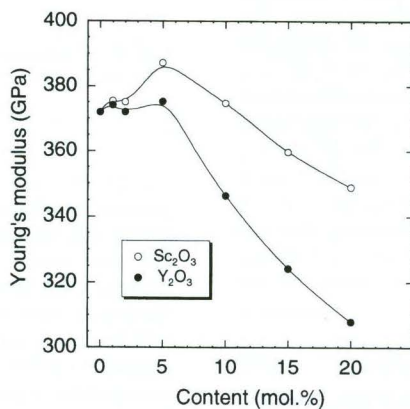


Fig. 4. Variation of Young's modulus with R<sub>2</sub>O<sub>3</sub> composition for MoSi<sub>2</sub>-SiO<sub>2</sub>-R<sub>2</sub>O<sub>3</sub> system.

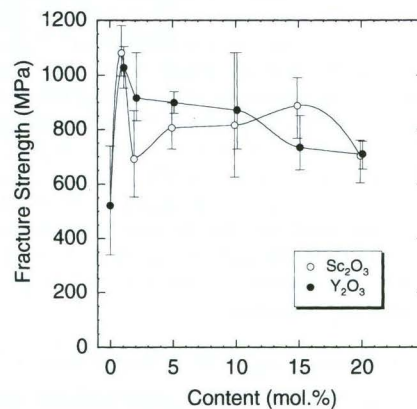


Fig. 5. Variation of fracture strength with R<sub>2</sub>O<sub>3</sub> composition for MoSi<sub>2</sub>-SiO<sub>2</sub>-R<sub>2</sub>O<sub>3</sub> system.

sintered bodies and Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> appears as a crystalline phase. With greater Y<sub>2</sub>O<sub>3</sub> addition (10–20 mol%), SiO<sub>2</sub> disappears and Y<sub>2</sub>SiO<sub>5</sub> phase is formed, replacing Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> phase, as anticipated from Fig. 9(b), with some excess Y<sub>2</sub>O<sub>3</sub> remaining.

For Sc<sub>2</sub>O<sub>3</sub> addition, the lower additive content (1–5 mol%)

has almost the same results as Y<sub>2</sub>O<sub>3</sub> addition (i.e., formation of 1:2 silicate, Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>), but, on the other hand, higher additive content (10–20 mol%) does not produce 1:1 silicate, Sc<sub>2</sub>SiO<sub>5</sub>. The hot-pressing temperature used in this study is 1600°C, and this temperature is evidently high enough for the following reaction to occur:

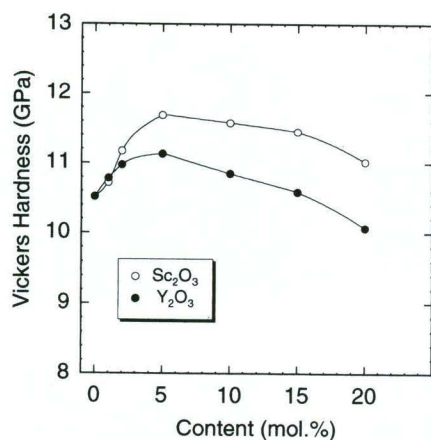


Fig. 6. Variation of Vickers Hardness with  $R_2O_3$  composition for  $MoSi_2-SiO_2-R_2O_3$  system.



This is expected, since the eutectic temperature between  $Sc_2Si_2O_7$  and  $SiO_2$  at  $1660^\circ C$  is near enough for this reaction to take place easily. For greater  $Sc_2O_3$  content (10–20 mol%) when the eutectic temperature between  $Sc_2SiO_5$  and  $Sc_2Si_2O_7$ , at  $1770^\circ C$ , is much higher than the hot-pressing temperature, the following reaction evidently takes place with more difficulty:



suggesting a larger kinetic or nucleation barrier.

## (2) Change of Density and Young's Modulus

As is shown in Section III, density and Young's modulus of sintered bodies increases then decreases with additive content. To discuss such changes, densities and Young's moduli of constituent phases are listed in Table V.<sup>35,39–49</sup> The density of monolithic  $MoSi_2$  sintered body in this study is  $5.92 \text{ g/cm}^3$ , which is lower than the theoretical density of  $6.27 \text{ g/cm}^3$  calculated from XRD;<sup>39</sup> thus, the decrement of density must be attributed to the formation of low density ( $2.20 \text{ g/cm}^3$ ) glassy  $SiO_2$ . As discussed in the phase analysis part, some cristobalite exists that spontaneously crystallizes from glassy  $SiO_2$ . The density of cristobalite is  $2.33 \text{ g/cm}^3$ ;<sup>41</sup> therefore, the difference in density between glassy  $SiO_2$  and cristobalite is small. Although the density of  $Mo_{\leq 5}Si_3C_{\leq 1}$  phase depends on its chemical composition, a typical value for  $Mo_{4.8}Si_3C_{0.6}$  is reported as  $7.891 \text{ g/cm}^3$ ,<sup>40</sup> which is slightly higher than that of  $MoSi_2$ . Because the content of  $Mo_{\leq 5}Si_3C_{\leq 1}$  phase is small ( $\sim 3 \text{ vol}\%$  detected through image analysis of SEM photographs), it can be neglected for the density calculation. Supposing that the  $MoSi_2$  sintered body is composed of only  $MoSi_2$  and glassy  $SiO_2$ , the density of the sintered body is calculated by the following equation:

$$D_{\text{sintered body}} = (1 - V_{SiO_2})D_{MoSi_2} + V_{SiO_2}D_{SiO_2} \quad (4)$$

where  $V$  and  $D$  represent volume fraction and density.  $D_{\text{sintered body}}$ ,  $D_{MoSi_2}$ , and  $D_{SiO_2}$  are  $5.92$ ,  $6.27$ , and  $2.22 \text{ g/cm}^3$ , respectively.  $V_{SiO_2}$  is calculated as  $8.6\%$  by Eq. (4), in good agreement with the volume fraction from image analysis ( $\sim 8.5\%$ ). This value is somewhat larger than the total oxide content in the starting  $MoSi_2$  powder ( $\sim 6 \text{ vol}\%$ ) as listed in Table II. It may be attributed to more oxidation of fresh surface by ball milling.

The volume changes ( $\Delta V$ ) associated with the reactions

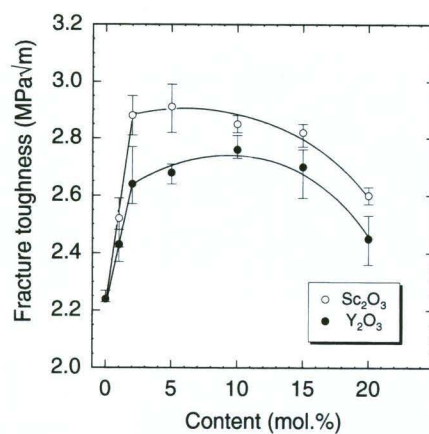
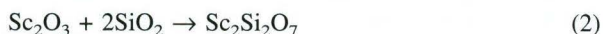


Fig. 7. Variation of fracture toughness with  $R_2O_3$  composition for  $MoSi_2-SiO_2-R_2O_3$  system.

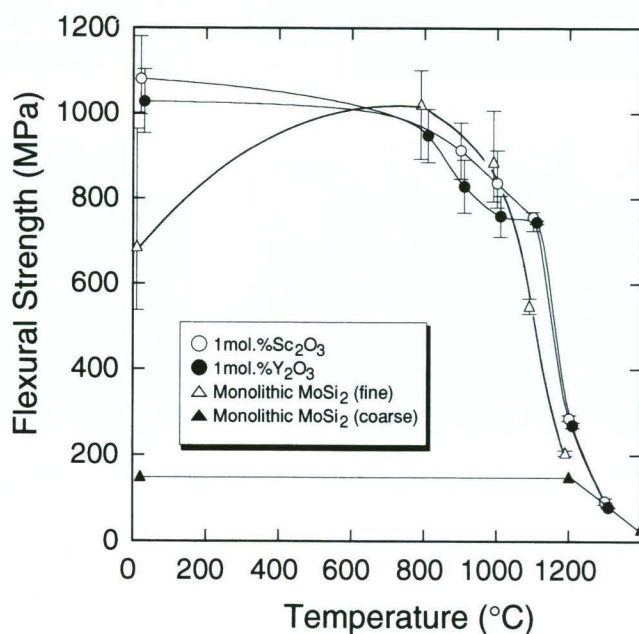


Fig. 8. Variation of flexural strength with temperature for  $MoSi_2-SiO_2-R_2O_3$  system.

are  $\Delta V = -18\%$  and  $-15\%$ , respectively. Because almost no porosity is observed by SEM, the progress of the reactions (2) or (5), i.e., the removal of  $SiO_2$ , results in an increase in density. For higher additive content (10–20 mol%), the densities decrease, because the densities of unreacted  $Sc_2O_3$  and  $Y_2O_3$  are lower than that of  $MoSi_2$ .

The Young's modulus of polycrystalline  $MoSi_2$  ( $440 \text{ GPa}$ ) is calculated from the single crystal.<sup>35</sup> On the other hand, that of glassy  $SiO_2$  is much smaller ( $73 \text{ GPa}$ ).<sup>41</sup> To a first approximation, the monolithic  $MoSi_2$  sintered body is composed of  $8.5 \text{ vol}\%$  glassy  $SiO_2$  phase and  $91.5 \text{ vol}\%$   $MoSi_2$  phase, and the Young's modulus of the sintered body without pores is calculated as  $386 \text{ GPa}$  using the following equation:<sup>50</sup>

$$\ln E_{\text{sintered body}} = V_{SiO_2} \ln E_{SiO_2} + V_{MoSi_2} \ln E_{MoSi_2} \quad (6)$$

The value of  $386 \text{ GPa}$ , somewhat larger than the observed value of  $372 \text{ GPa}$ , is within a reasonable comparison. Then, on the second approximation, the effect of dispersion of the  $Mo_{\leq 5}Si_3C_{\leq 1}$  phase is accounted for in the calculation. Recently, we investigated the properties of polycrystalline

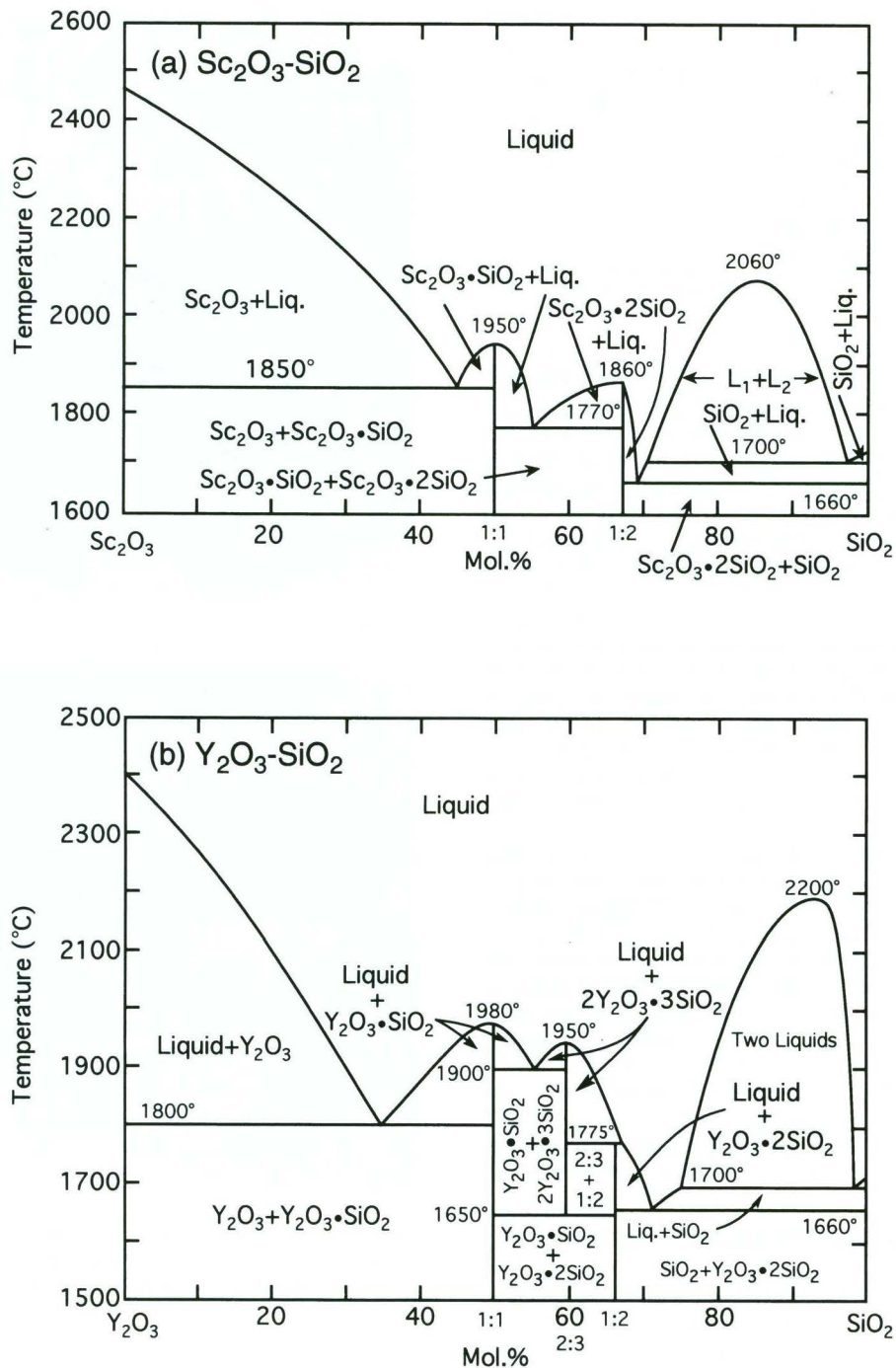


Fig. 9. Phase diagrams of (a) Sc<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> and (b) Y<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> binary systems.<sup>21,22</sup>

Mo<sub>≤5</sub>Si<sub>3</sub>C<sub>≤1</sub> phase and its composites.<sup>47</sup> The measured Young's modulus of polycrystalline Mo<sub>≤5</sub>Si<sub>3</sub>C<sub>≤1</sub> was 285 GPa. In this experiment, ~3 vol% of Mo<sub>≤5</sub>Si<sub>3</sub>C<sub>≤1</sub> phase is dispersed in the monolithic MoSi<sub>2</sub> sintered body. The Young's modulus of the MoSi<sub>2</sub> sintered body without pores is calculated using a modified equation:

$$\ln E_{\text{sintered body}} = V_{\text{SiO}_2} \ln E_{\text{SiO}_2} + V_{\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}} \ln E_{\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}} + V_{\text{MoSi}_2} \ln E_{\text{MoSi}_2} \quad (7)$$

where  $V_{\text{SiO}_2} = 8.5\%$ ,  $V_{\text{Mo}_{\leq 5}\text{Si}_3\text{C}_{\leq 1}} = 3.0\%$ , and  $V_{\text{MoSi}_2} = 88.5\%$ . The calculated Young's modulus is 373 GPa and is in unusually close agreement with the observed value of 372 GPa.

The 5 mol% addition results in the maximum Young's modulus for both Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> addition (Fig. 4). As is

discussed above, free SiO<sub>2</sub> is minimized around this composition. Although there are no available reported values of the Young's modulus for Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, and other silicate phases, it is considered that the Young's modulus of such crystalline phases is surely higher than that of glassy SiO<sub>2</sub>. Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> should have higher Young's modulus than Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, because Sc<sup>3+</sup> ion is smaller than Y<sup>3+</sup> ion, with inevitably stronger Sc-O-Si bonds than Y-O-Si (as also confirmed by the effectively higher melting point). The Young's moduli of Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> are 227.6 GPa<sup>48</sup> and 171.5 GPa,<sup>49</sup> respectively, suggesting again the stronger Sc-O versus Y-O bond (tracking mp at 2460°C versus 2400°C). Thus, the Young's modulus of Sc<sub>2</sub>O<sub>3</sub>-added system is always higher than that of Y<sub>2</sub>O<sub>3</sub>-added system. However, when the oxides are in excess at >5 mol%, Young's modulus of both MoSi<sub>2</sub>-SiO<sub>2</sub>-Sc<sub>2</sub>O<sub>3</sub> and MoSi<sub>2</sub>-SiO<sub>2</sub>-Y<sub>2</sub>O<sub>3</sub> systems inevitably drop.

**Table V. Density and Young's Modulus of Phases Present<sup>†</sup>**

Phase	Density (g/cm <sup>3</sup> ) <sup>‡</sup>	Young's modulus (GPa) <sup>‡</sup>
MoSi <sub>2</sub>	6.270 [39]	440 [35]
Mo <sub>0.5</sub> Si <sub>3</sub> C <sub>≤1</sub>	7.891 [40]	285 [47]
SiO <sub>2</sub> (glass)	2.20 [41]	73 [41]
Sc <sub>2</sub> O <sub>3</sub>	3.840 [42]	227.6 [48]
Y <sub>2</sub> O <sub>3</sub>	5.032 [43]	171.5 [49]
Sc <sub>2</sub> Si <sub>2</sub> O <sub>7</sub>	3.394 [44]	
β-Y <sub>2</sub> Si <sub>2</sub> O <sub>7</sub>	4.030 [45]	
Y <sub>2</sub> SiO <sub>5</sub>	4.441 [46]	

<sup>†</sup>Y<sub>4</sub>Si<sub>3</sub>O<sub>12</sub> data was not available. <sup>‡</sup>Numbers in brackets are Reference numbers.

### (3) Microstructural Dependence of Mechanical Properties

As detailed in Section III, only a small amount of rare-earth oxide addition strongly improves various mechanical properties, e.g., 1 mol% additive significantly improves fracture strength. To illuminate the effect of crystallization on mechanical properties, a TEM observation on 1-mol%-Sc<sub>2</sub>O<sub>3</sub>-added system with the highest bending strength is shown in Figs. 10(a) and (b). Figure 10(a) shows a siliceous pocket in the sintered body, in which crystalline Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> has formed. Another siliceous pocket is indicated in Fig. 10(b); dislocation networks are observed in the MoSi<sub>2</sub> around the oxide pockets. The crystalline silicates appear to be less deformable than glassy SiO<sub>2</sub>. Thus, thermal stress between matrix and oxides is not fully relaxed by the plastic deformation of glassy SiO<sub>2</sub>, leading to numerous dislocations in the MoSi<sub>2</sub>. When the pockets contain the crystalline silicates, more dislocations can be seen than in the case of noncrystalline or only slightly crystalline pockets.

For the 1–5 mol% addition, the amount of total oxide phase is not so large. Formed R<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> crystals effectively toughen the glassy phase and improve mechanical properties, perhaps by reducing the source flaw size in the glass. With greater addition, however, the oxide phases become continuous and may act as larger fracture origins. Mechanical properties, especially strength, decline. In Fig. 5, the variation of five to six specimen tests becomes large at 10 mol% addition, equivalent to 14 and 17 vol% addition for Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> addition, respectively (Table III). Thus, 10 mol% addition is at the threshold value of ~16 vol% for percolative contact of oxide phases and may well explain why the error bars increase at this level. These reasonings should be considered preliminary and tentative at this stage.

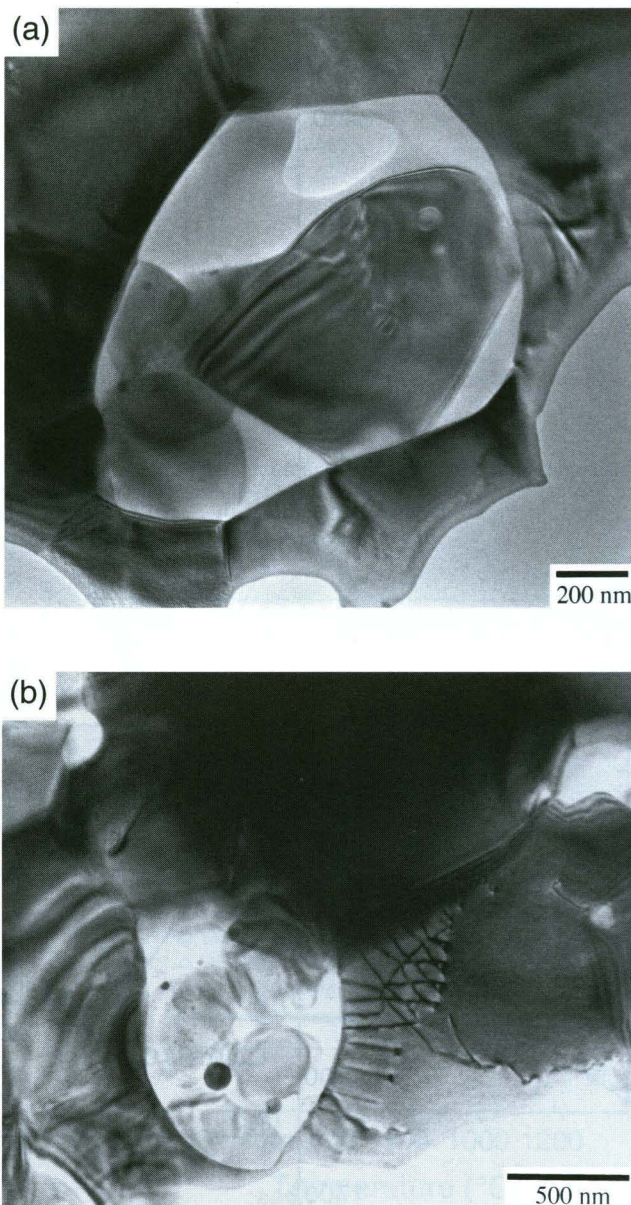
### (4) Comparison with Other Techniques to Remove Glassy SiO<sub>2</sub>

In the Introduction, SiO<sub>2</sub>-removing techniques were classified into five categories. The present study on MoSi<sub>2</sub>-SiO<sub>2</sub>-Sc<sub>2</sub>O<sub>3</sub> and MoSi<sub>2</sub>-SiO<sub>2</sub>-Y<sub>2</sub>O<sub>3</sub> is in the same vein as our previous study on MoSi<sub>2</sub>-SiO<sub>2</sub>-ZrO<sub>2</sub>.<sup>8</sup> This method is basically analogous to that used with Si<sub>3</sub>N<sub>4</sub>-based ceramics.<sup>23–26</sup> One difference, however, is that glassy SiO<sub>2</sub> and additive oxides in MoSi<sub>2</sub> do not form films at grain boundaries, concentrating at triple points, so that the reaction seems to progress easily under relatively mild conditions (1600°C for 1 h).

## V. Conclusions

In this investigation, we examined the modification of the grain-boundary phase in MoSi<sub>2</sub> hot-pressed bodies by additions of Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub>. In summary,

- (1) The intergranular glassy SiO<sub>2</sub> phase was converted to the crystalline silicates.
- (2) Mechanical properties at both room temperature and high temperatures were enhanced by Sc<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> addition, even at an amount as low as 1 mol%.
- (3) These results indicate that *in-situ* crystallization of the glassy SiO<sub>2</sub> phase by the addition of refractory oxides holds promise for fabricating MoSi<sub>2</sub>-based composites with im-



**Fig. 10.** TEM photographs of 1-mol%-Sc<sub>2</sub>O<sub>3</sub>-added MoSi<sub>2</sub>: (a) Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> formation inside the glassy SiO<sub>2</sub> pocket (by EDS), and (b) dislocation networks around siliceous pockets.

proved mechanical properties, and further optimization can be expected.

**Acknowledgment:** The authors gratefully acknowledge Professor S. W. Lee at Sun Moon University for helpful discussions. We gratefully acknowledge Japan New Metals Co., Ltd. for kindly providing the MoSi<sub>2</sub> powder.

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