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# Mechanism analysis on finishing of reaction-sintered silicon carbide by combination of water vapor plasma oxidation and ceria slurry polishing

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**Abstract.** Reaction-sintered silicon carbide (RS-SiC), which is considered as a promising mirror material for space telescope systems, requires a high surface property. An ultrasmooth surface with a Ra surface roughness of 0.480 nm was obtained after water vapor plasma oxidation for 90 min followed by ceria slurry polishing for 40 min. The oxidation process of RS-SiC by water vapor plasma was analyzed based on the Deal–Grove model, and the theoretical calculation results are consistent with the measured data obtained by scanning white light interferometer (SWLI), scanning electron microscopy/energy-dispersive x-ray, and atomic force microscope. The polishing process of oxidized RS-SiC by ceria slurry was investigated according to the Preston equation, which would theoretically forecast the evolutions of RS-SiC surfaces along with the increasing of polishing time, and it was experimentally verified by comparing the surface roughnesses obtained by SWLI and the surface morphologies obtained by SEM. The mechanism analysis on the finishing of RS-SiC would be effective for the optimization of water vapor plasma oxidation parameters and ceria slurry polishing parameters, which will promote the application of RS-SiC substrates by improving the surface property obtained by the oxidation-assisted polishing method. © The Authors. Published by SPIE under a Creative Commons Attribution 3.0 Unported License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI. [DOI: [10.1117/1.OE.54.5.055106](https://doi.org/10.1117/1.OE.54.5.055106)]

Keywords: optical machining; reaction-sintered silicon carbide; water vapor plasma oxidation; ceria slurry polishing; Deal-Grove model; Preston equation.

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## 1 Introduction

Reaction-sintered silicon carbide (RS-SiC) has been considered as an ideal mirror material for space telescope systems and a promising ceramic material for the molds of glass lens,<sup>1,2</sup> as it has the excellent mechanical and chemical properties of a low thermal expansion coefficient, high thermal conductivity, high radiation resistance, high specific stiffness, good size stability, strong bending strength, and a low manufacturing cost.<sup>3,4</sup> Therefore, the fabrication process of RS-SiC has been a focus of research in the optics and ceramics fields.

However, RS-SiC is difficult to be precisely processed with a high accuracy because of its high hardness and chemical inertness.<sup>5</sup> Since SiC is harder than most other materials except diamond, the available technique for mechanical material removal of RS-SiC is very limited. Meanwhile, due to its chemical inertness, RS-SiC is difficult to react with common chemicals, which means that it is inefficient to remove RS-SiC through traditional chemical methods.

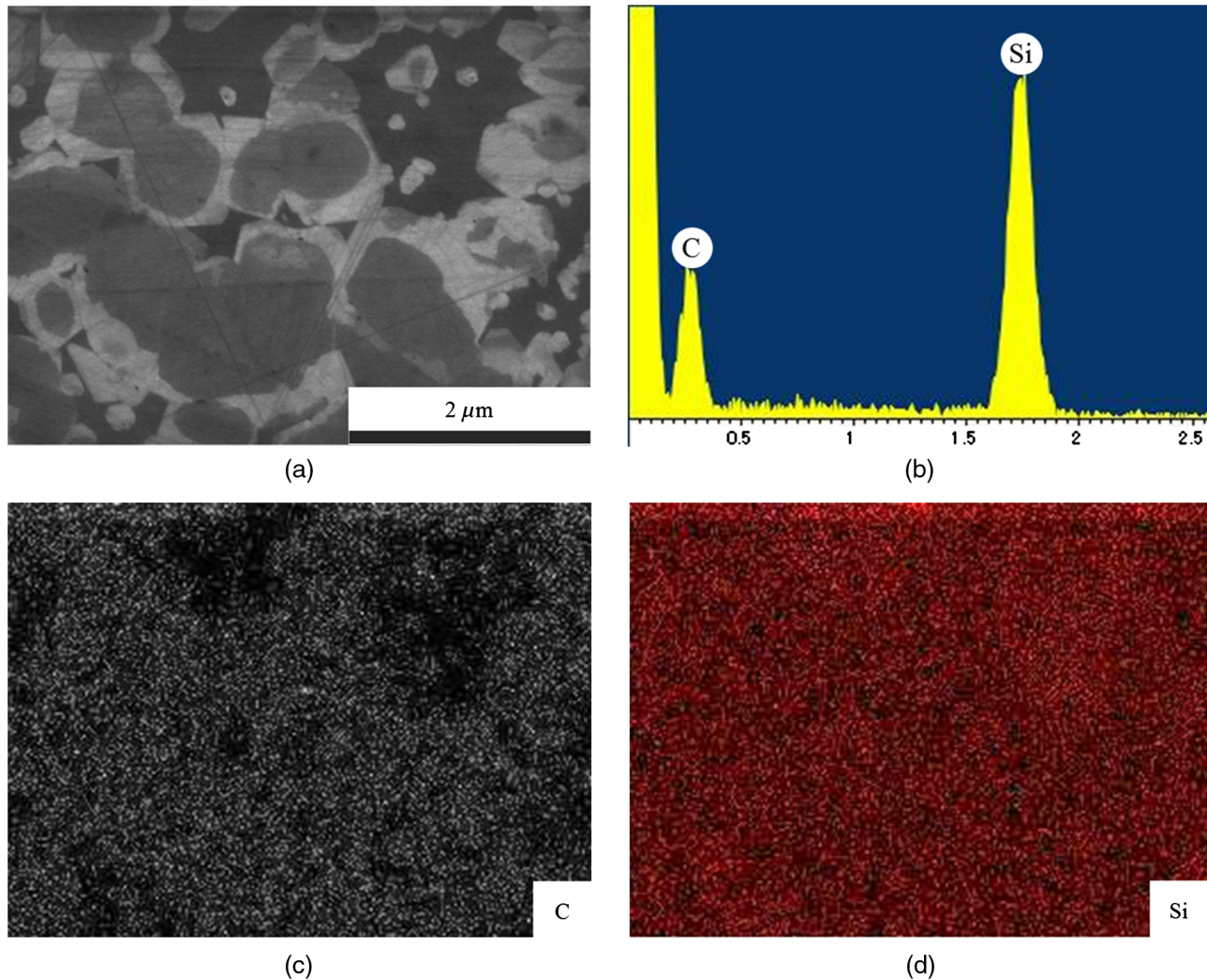
In addition, the fabrication process of RS-SiC generates a SiC domain and a Si domain as major components.<sup>6</sup> Scanning electron microscopy/energy-dispersive x-ray spectroscopy (SEM-EDX) was conducted to analyze the composition of the RS-SiC and the surface morphology with

corresponding element distributions was shown in Figs. 1(a) to 1(d). Thus, the differences in hardness and chemical activity among the nonuniform domains make the homogeneous removal of RS-SiC almost impossible, which means that it is difficult to obtain a smooth surface through mechanical or chemical methods.

Up until now, various researches on the smoothing of RS-SiC have been widely conducted, but few of them can yield an ultrasmooth surface. Diamond lapping<sup>7</sup> by mechanical material removal and plasma chemical vaporization machining (PCVM)<sup>8</sup> by chemical material removal of RS-SiC were conducted as the representations to verify the traditional techniques. Through SEM observation and scanning white light interferometer (SWLI) measurement, obvious scratches were observed on the surface processed by diamond lapping and bumpy structures appeared on that processed by PCVM, as shown in Figs. 2(a) to 2(d), since the mechanical hardnesses and chemical reactivities of Si and SiC differ. This is also a problem for the ultraprecision machining of other products in the SiC family, such as reaction bonded silicon carbide (RB-SiC) and hot-pressing silicon carbide (HP-SiC), since all of them consist of SiC and Si domains.

The traditional method to improve the surface property by the machining of RS-SiC is combination of deposition of the chemical vaporization deposition silicon carbide (CVD-SiC) on the RS-SiC surface and polishing of the deposition layer, since the components of CVD-SiC are uniform. However, we suppose there are two disadvantages for this method.

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**Fig. 1** Composition analysis of reaction-sintered silicon carbide (RS-SiC) substrate by scanning electron microscopy/energy-dispersive x-ray spectroscopy (SEM-EDX): (a) surface morphology observed by SEM, (b) element analysis by EDX, (c) carbon element distribution, (d) silicon element distribution.

First, the deposition of SiC by chemical vaporization is a high-cost, inefficient, and complex process, which would take a large amount of material and financial resources. Second, the hardness of CVD-SiC is quite high, which makes the machining of the deposition layer difficult and the material removal rate is low. Therefore, it is urgent to develop new techniques.

An effective way to reduce the difficulty in the finishing of RS-SiC is a combination of surface oxidation of RS-SiC and polishing of the oxide, since the oxide silica ( $\text{SiO}_2$ ) is an easy-to-machine material and an ultrasmooth  $\text{SiO}_2$  surface can be easily obtained.<sup>9</sup> An ultrasmooth RS-SiC surface with an Ra surface roughness of 0.480 nm can be obtained by the combination of water vapor plasma oxidation of RS-SiC for 90 min and ceria slurry polishing of oxide for 40 min. In this study, the water vapor plasma oxidation process of RS-SiC was analyzed based on the Deal–Grove model,<sup>10</sup> and the ceria slurry polishing process of the oxide was investigated according to the Preston equation.<sup>11</sup> Afterward, the theoretically calculated results were verified by the measured data obtained by SEM and SWLI. The obtention of an ultrasmooth RS-SiC surface by the combination of water

vapor plasma oxidation and ceria slurry polishing is a significant attempt to develop the machining level of RB-SiC, HP-SiC, and other SiC products.

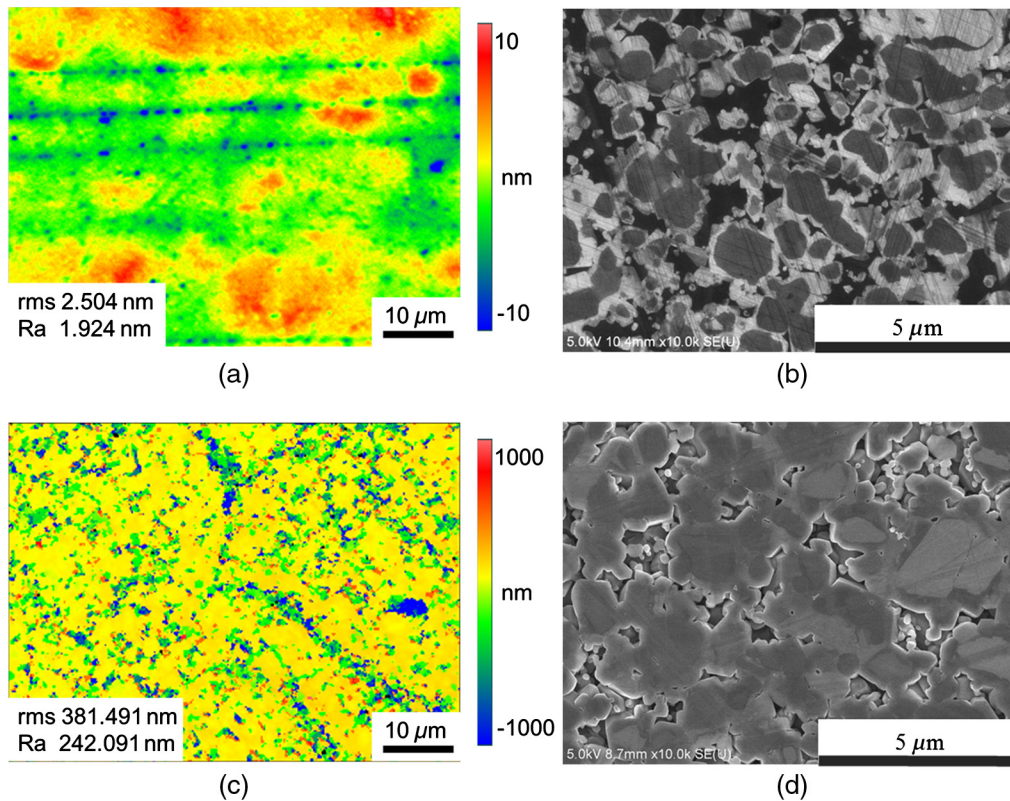
## 2 Experiment Setup

The initial RS-SiC specimen was prepared by diamond lapping, and the surface morphology is shown in Fig. 2(a). The atmospheric water vapor plasma was generated via applying an RF ( $f = 13.56$  MHz) electric power, and helium-based water vapor with a flow rate of 1.52 L/min was supplied as a process gas. Schematic diagram of the plasma oxidation system is shown in Fig. 3(a) and the oxidation parameters are listed in Table 1. After oxidation, the oxidized sample was polished with ceria slurry. A schematic diagram of the ceria slurry polishing system is shown in Fig. 3(b) and the polishing parameters are listed in Table 2.

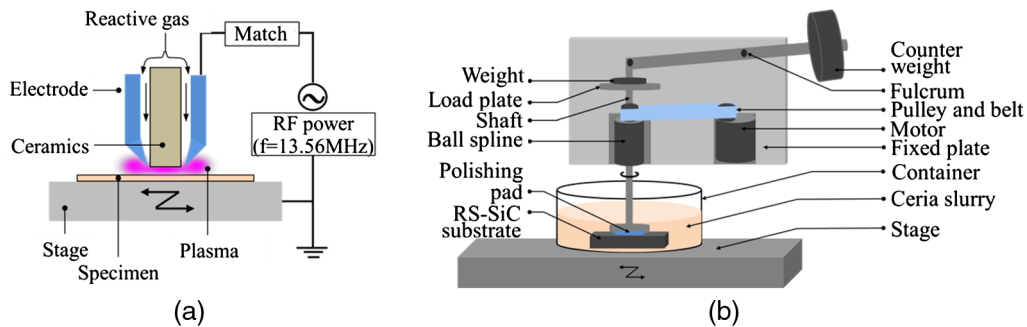
## 3 Results and Discussion

### 3.1 Theoretical Analysis

The combined process includes the water vapor plasma oxidation of RS-SiC and ceria slurry polishing of the oxide



**Fig. 2** Processed RS-SiC substrate analyzed by scanning white light interferometer (SWLI) and SEM: (a) processed by diamond lapping and measured by SWLI, (b) processed by diamond lapping and observed by SEM, (c) processed by plasma chemical vaporization machining (PCVM) and measured by SWLI, (d) processed by PCVM and observed by SEM.



**Fig. 3** Schematic diagrams of experimental setup: (a) plasma oxidation system and (b) ceria slurry polishing system.

**Table 1** Plasma oxidation parameters.

RF power	32 W
Process gas	He + H <sub>2</sub> O, 1.52 SLM
Duty ratio	10%
Oxidation time	90 min

layer. Therefore, in order to investigate the plasma oxidation process and the polishing process, we had analyzed the plasma oxidation of RS-SiC in accordance with the Deal-Grove model<sup>10</sup> and the removal of the oxide by ceria slurry polishing using the Preston equation,<sup>11</sup> respectively.

First, we used an atomic force microscope (AFM) to investigate the difference of oxidation rates between SiC and Si grains, and the measurement results when the oxidation time is 60 min and 120 min are shown in Figs. 4(a) and 4(b), respectively. From the two transversals, we found

**Table 2** Ceria slurry polishing parameters.

Abrasive		Ceria
Particle size	$\phi$ 190 nm	
Concentration	0.1 wt. %	
Load	10 g	
Rotation speed	1000 rpm	
Polishing pad	K0017 (FILWEL Co. Ltd.)	
Scan speed	200 mm/min	
Polishing time	10, 20, 40, 50, 60, 80, 100, 120, 140, 160 min	

that the oxidation rates of Si grains are slightly higher than that of SiC grains, which can be judged from the difference of the oxide height between SiC and Si grains. However, the height difference is not more than 10 nm after plasma oxidation of RS-SiC for 120 min. Taking into account that the rate of volume expansion in the oxidation of Si to SiO<sub>2</sub> is slightly higher than that in oxidation of SiC to SiO<sub>2</sub>, the

interface between the RS-SiC substrate and oxide layer is smooth. Therefore, an ultrasmooth RS-SiC surface is expected to be obtained after polishing of the oxide layer generated by water vapor plasma.

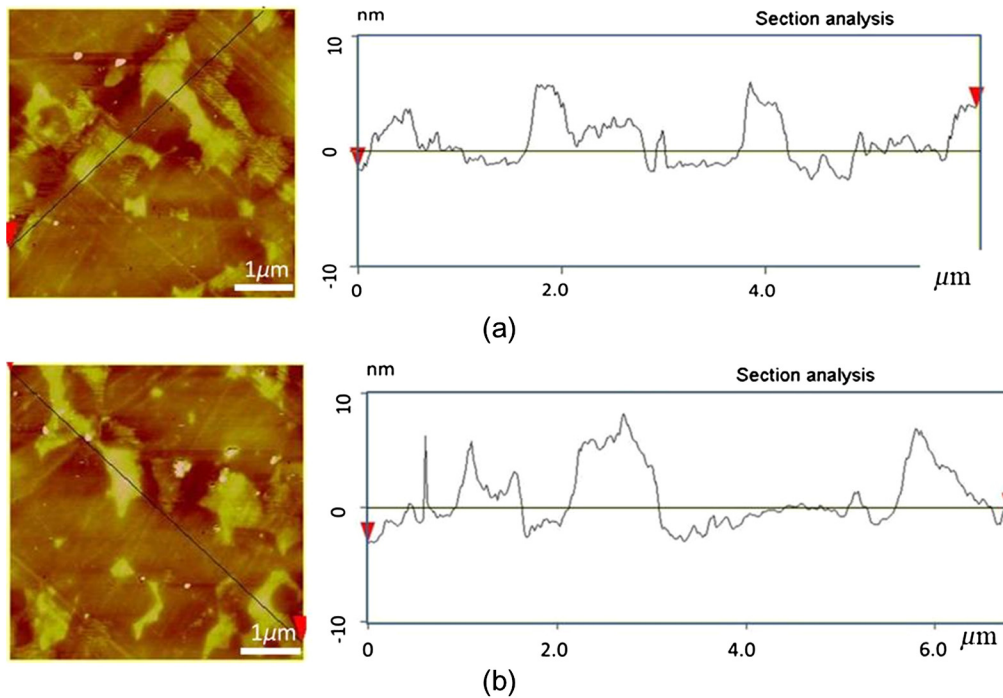
Schematic diagram of the plasma oxidation of RS-SiC and the polishing of the oxide is shown in Fig. 5. The relationship between oxidation depth  $d$  (nm) and oxidation time  $t_1$  (min) can be calculated based on the classic Deal-Grove model, as shown in Eq. (1), where  $A$  and  $B$  are the parameters in the Deal-Grove model:

$$d^2 + Ad = Bt_1. \tag{1}$$

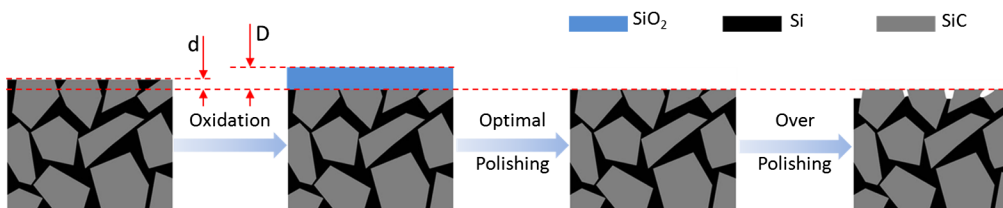
The thickness of the oxide layer  $D$  (nm) is the product of the oxidation depth  $d$  and the rate of volume expansion  $r$ , which is shown in Eq. (2):

$$D = rd. \tag{2}$$

In order to obtain an ultrasmooth RS-SiC surface without any remaining oxide, the polishing depth should be equal to the thickness of the oxide layer. Therefore, the relationship between  $D$  and the polishing time  $t_2$  (min) can be expressed by Eq. (3) according to the Preston equation,



**Fig. 4** Analysis of oxidized surface by atomic force microscope (AFM): (a) oxidation 60 min and (b) oxidation 120 min.



**Fig. 5** Schematic diagram of the water vapor plasma oxidation of RS-SiC and removal of the oxide layer by ceria slurry polishing.

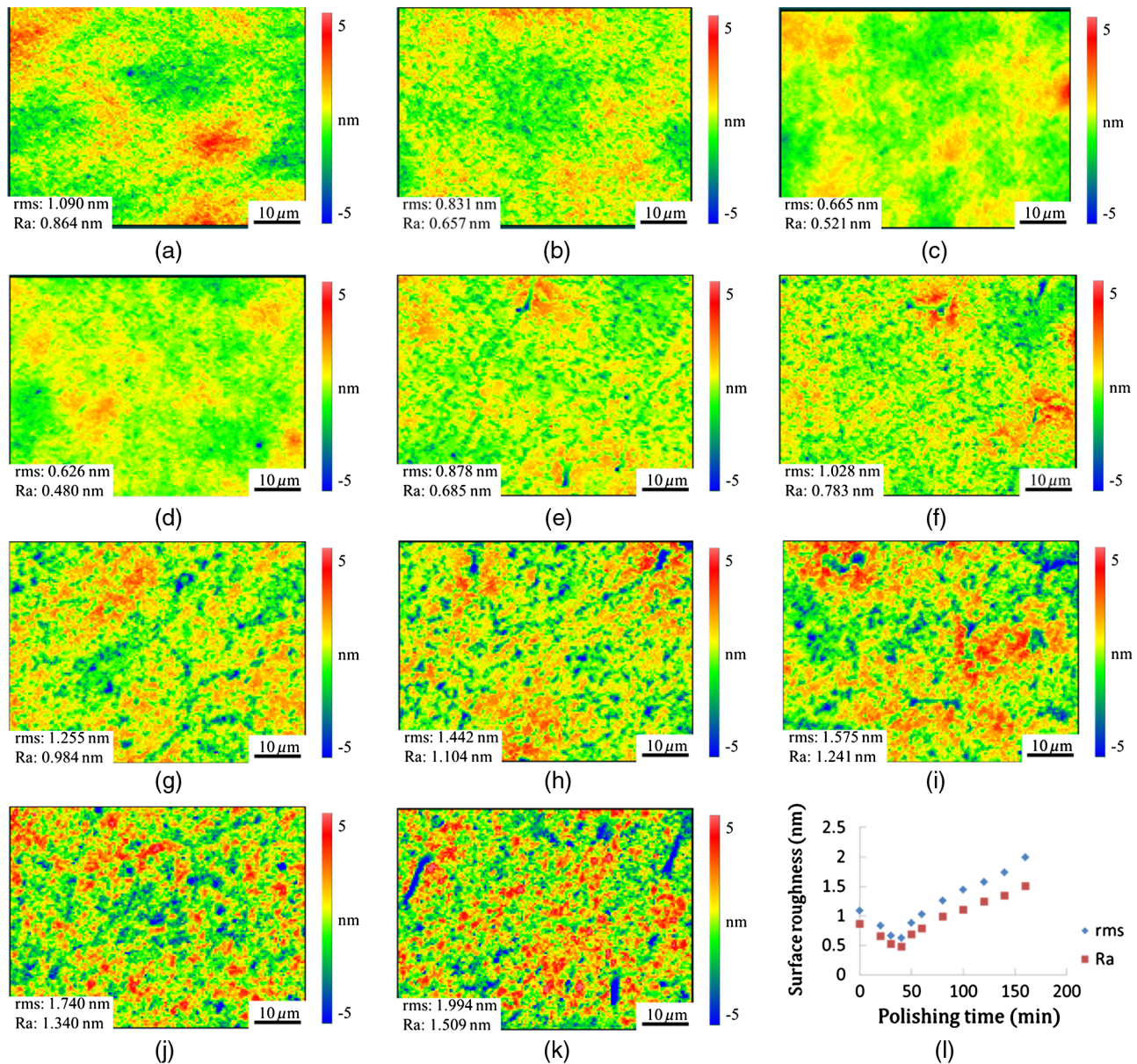
where  $k$ ,  $p$ , and  $v$  are the Preston's coefficient, the downward pressure, and the relative velocity between the polishing pad and the sample, respectively.

$$D = kpv t_2. \quad (3)$$

For better control of the plasma oxidation time and corresponding ceria polishing time, in future research we will confirm the exact Deal–Grove model in water vapor plasma oxidation of RS-SiC by measuring the oxidation depth corresponding to a certain oxidation time. Meanwhile, through experiments on the ceria abrasive polishing of silicon oxide, we can verify the polishing rate with the aim of keeping the polishing depth equal to the thickness of the oxide layer.

### 3.2 Evaluation of Surface Morphology

The original surface, shown in Fig. 2(a), was obtained by diamond lapping. After water vapor plasma oxidation of RS-SiC for 90 min, the root-means-square (rms) and Ra surface roughnesses measured by SWLI are 1.090 and 0.864 nm, respectively, as shown in Fig. 6(a). SWLI measurements were conducted to evaluate the surface roughness after polishing for a certain period, as shown in Figs. 6(b) to 6(k). The evolutions of surface roughnesses corresponding to polishing time were summarized in Fig. 6(l). From the SWLI measurements, it could be concluded that the surface roughness reached its optimum after polishing for 40 min, where the corresponding rms and Ra roughnesses are 0.626 and 0.480 nm, respectively.

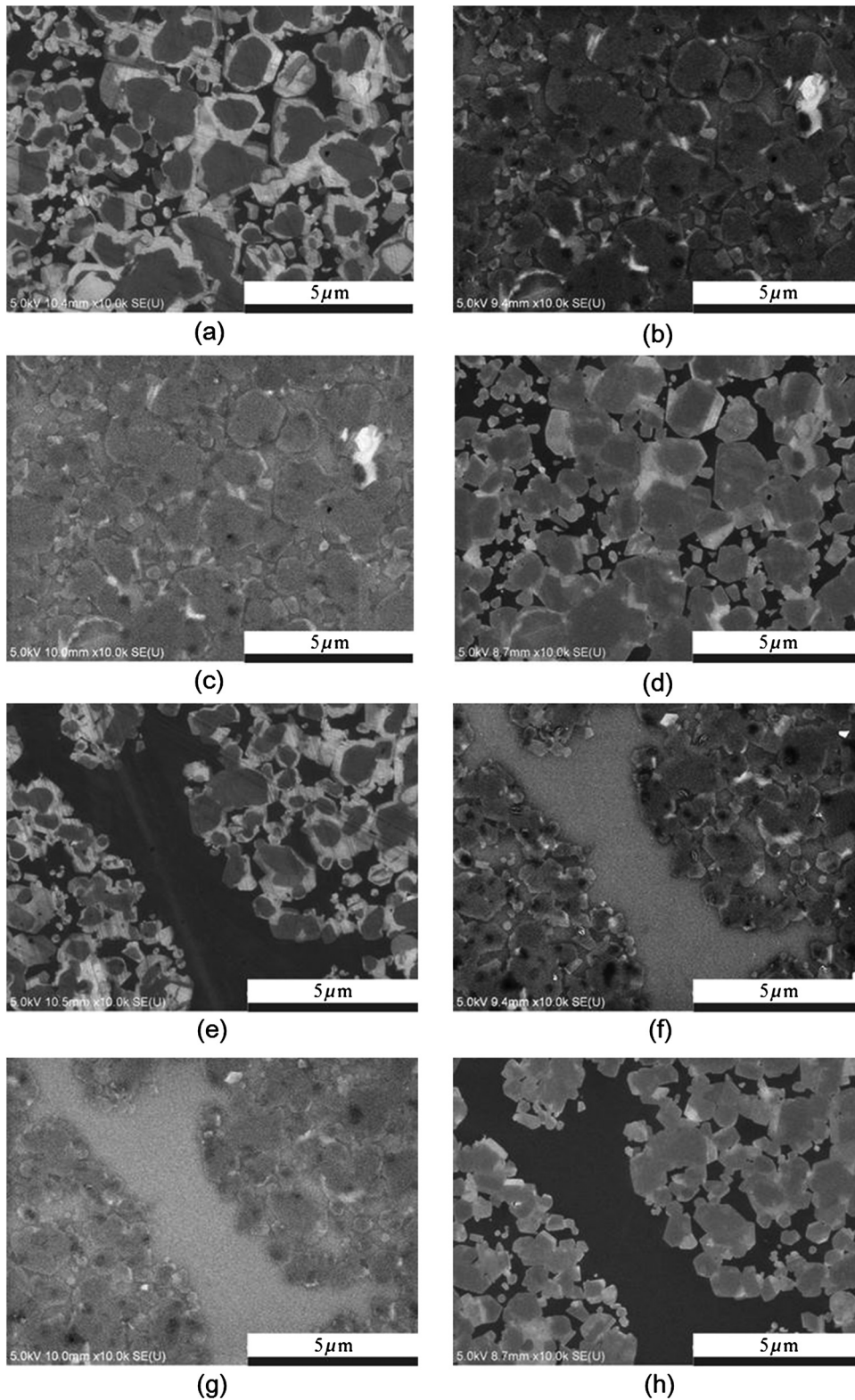


**Fig. 6** Surface roughness measured and evaluated by SWLI: (a) after plasma oxidation 90 min, (b) after polishing 10 min, (c) after polishing 20 min, (e) after polishing 40 min, (e) after polishing 50 min, (f) after polishing 60 min, (g) after polishing 80 min, (h) after polishing 100 min, (i) after polishing 120 min, (j) after polishing 140 min, (k) after polishing 160 min, (l) evolution of surface roughnesses rms and Ra corresponding to polishing time.

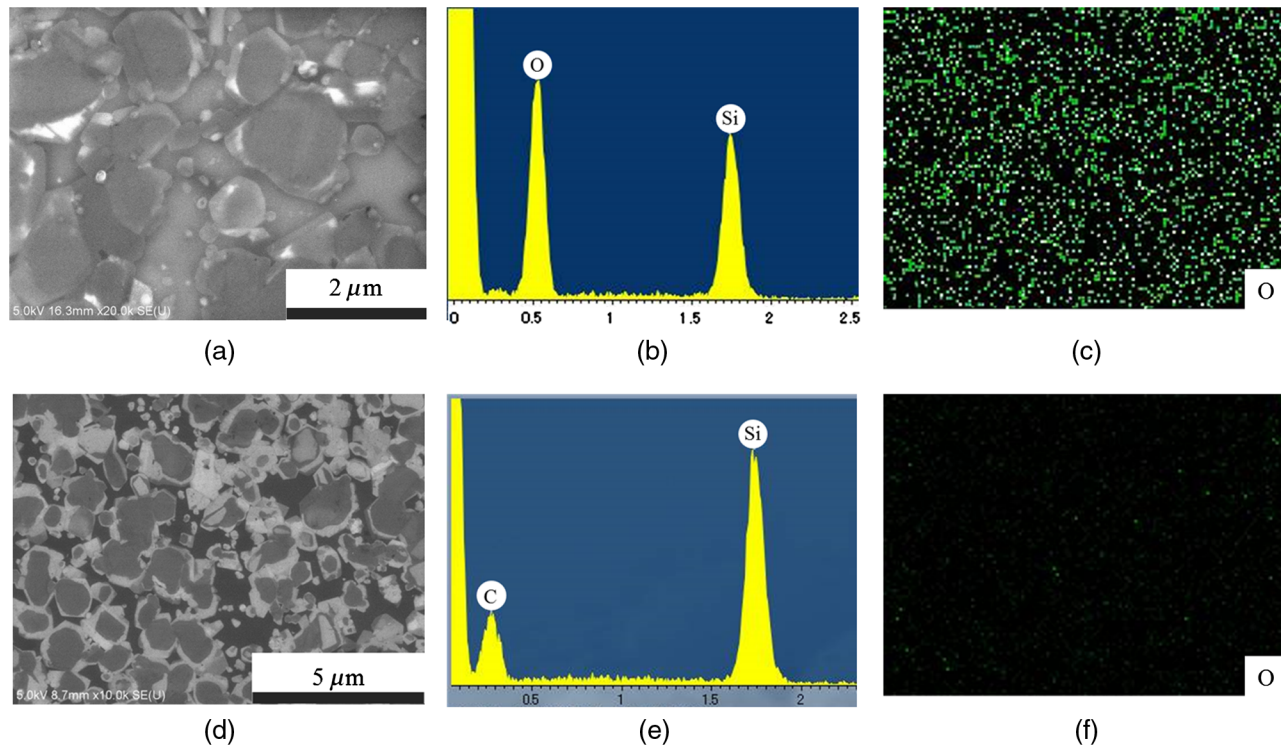
SEM observation was conducted to evaluate the surface morphologies before oxidation, after oxidation, after optimal polishing, and after over polishing at the same position, as shown in Figs. 7(a) to 7(h), which was aimed to investigate

the plasma oxidation behavior of RS-SiC and the polishing property of the oxide layer.

The elements and their distributions of the oxidized surface and the optimal polished surface were investigated by



**Fig. 7** Morphologies observed by SEM: (a), (b), (c), and (d) are morphologies before oxidation, after oxidation, after optimal polishing, and after over polishing at the same position. (e), (f), (g), and (h) are those at another position.



**Fig. 8** Elements and their distributions investigated by SEM-EDX: (a) oxidized RS-SiC surface, (b) element analysis corresponding to (a), (c) oxygen element distribution corresponding to (a), (d) optimal polished surface of plasma oxidized RS-SiC substrate, (e) element analysis corresponding to (d), (f) oxygen element distribution corresponding to (d).

SEM-EDX, as shown in Figs. 8(a) to 8(f). In contrast to the original RS-SiC surface in Fig. 1, after water vapor plasma oxidation, the surface was completely oxidized since there were few carbon elements on the oxidized surface and oxygen elements were distributed uniformly.

### 3.3 Discussion of Experimental Data

From the evolutions of surface roughnesses in Figs. 6(a) to 6(k), it can be observed that the rms and Ra are improved by ceria slurry polishing during the former 40 min. That is because the polishing depth is in the oxide layer and it is easy for the ceria slurry polishing of SiO<sub>2</sub> to obtain an ultra-smooth surface. Along with the increase of polishing time, the surface roughnesses are deteriorated since the oxide layer has already been removed as can be judged from the oxygen element distribution of the optimal polished surface (polishing for 40 min) in Fig. 8(f). The Vickers hardnesses (GPa) of SiC, ceria, SiO<sub>2</sub>, and Si are 24–28,<sup>12</sup> 5–7.5,<sup>13</sup> 7.6,<sup>14</sup> and 7–9,<sup>15</sup> respectively. As the hardness of a ceria particle is much less than that of SiC and is slightly smaller than those of silica and Si, the ceria slurry polishing of RS-SiC whose major components are SiC and Si would result in worse surface roughness in further polishing. Thus, the polishing depth should be kept within the oxide layer in order to obtain a smooth surface.

It is interesting to note that there are few scratches on the oxidized RS-SiC surfaces by water vapor plasma, as shown in Figs. 7(b), 7(f), and 8(a), although the initial surfaces, which are prepared by diamond lapping, have many scratches, as shown in Figs. 2(a), 2(b), 7(a), and 7(e). We hypothesize that there are two reasons for the disappearance

of scratches on the oxidized RS-SiC surface. First, the area with scratches has a larger surface area than the other areas, and the oxidation of RS-SiC increases its volume; thus, the scratches are closed by the expanded oxide. Second, the zone surrounding that with scratches has a higher oxidation rate because of lattice strain; thus, the rapidly expanding oxide will fill the scratches. Through the closing and filling of scratches by bulgy oxide, scratches are eliminated on the oxidized RS-SiC surface.<sup>16</sup> Thus, after plasma oxidation the surface roughness is reduced, which can be validated by comparing the original surface in Fig. 2(a) and the oxidized surface in Fig. 6(a).

As shown in Figs. 8(a) and 8(b), the entire RS-SiC surface is completely oxidized by plasma and the distribution of the oxygen element is uniform. It is difficult to estimate the difference of the oxidation rate between SiC and Si grains in RS-SiC, which are different from the oxidation behaviors of RS-SiC in anodic oxidation and thermal oxidation. In the anodic oxidation of RS-SiC, the oxidation rates of the SiC grains are higher than that of Si grains; in contrast, in thermal oxidation of RS-SiC, the oxidation rates of the SiC grains are lower than that of the Si grains.<sup>16</sup> The achievement of an ultrasmooth surface by ceria slurry polishing requires that the removal depth should be not more than the thickness of the oxide layer; thus, the uniform oxide layer in the plasma oxidation of RS-SiC is desirable in order to obtain a better surface.

Figures 8(d) to 8(f) reveal that there is little remaining oxide on the optimal polished surface, which means that we can obtain ultrasmooth RS-SiC surface and simultaneously keep the outstanding properties of the RS-SiC substrate. Otherwise, the remaining oxide will reduce the excellent



mechanical and chemical properties of RS-SiC. In the thermal oxidation of RS-SiC followed by ceria slurry polishing, although an ultrasmooth surface with an Ra surface roughness of 0.274 nm in  $5\ \mu\text{m} \times 5\ \mu\text{m}$  region measured by AFM can also be obtained by polishing for an appropriate time,<sup>16</sup> there is remaining oxide on the surface, since the oxidation rates of SiC and Si grains differ in thermal oxidation. Furthermore, plasma oxidation can be applied in the process of a large-scale RS-SiC substrate by scanning the oxidation spot through the entire surface. Therefore, plasma oxidation has an obvious superiority as a postprocessing method in finishing of RS-SiC.

#### 4 Conclusions

The plasma oxidation rate is almost uniform between the SiC and Si grains in RS-SiC, making the water vapor plasma oxidation suitable for the fine finishing of an RS-SiC substrate, and an ultrasmooth surface with an Ra roughness of 0.480 nm followed by ceria slurry polishing can be easily obtained. After polishing for an appropriate period, the excellent properties of RS-SiC are preserved, since there is little remaining oxide on the newly exposed surface. The water vapor plasma oxidation process of RS-SiC was analyzed based on the Deal-Grove model, and the ceria slurry polishing process of the oxide was investigated according to the Preston equation.

Relative to the combination of the deposition of CVD-SiC and polishing of the deposition layer, the method with a combination of water vapor plasma oxidation of RS-SiC and ceria slurry polishing of the oxide layer is a low-cost, efficient, and simple process, and the oxide layer is easy to machine, which can be an attractive technique for the machining of RS-SiC, RB-SiC, HP-SiC, and other SiC products by further development.

Therefore, the combination of plasma oxidation and ceria slurry polishing can be considered as a promising and post-processing technique for the fine finishing of RS-SiC, which will promote the application of RS-SiC products in the optics and ceramics fields.

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Biographies of the other authors are not available.