

OXIDATION BEHAVIOUR OF SILICON CARBIDE - A REVIEW

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Abstract. Silicon Carbide as an inorganic material possesses properties like high thermochemical stability, high hardness and fracture toughness, low thermal expansion coefficient etc. It is therefore, widely used in the making of refractory, semiconductor devices, combustion engines, etc. Being a nonoxide, it has a tendency to get oxidized at elevated temperature under oxidizing atmosphere. Oxidation of silicon carbide can be either active or passive. Active oxidation reduces the strength of the samples whereas passive oxidation leads to the formation of coherent silica layer over silicon carbide surface, thereby improving its performances in several applications. Being an interesting area of research, numerous works have been reported on the oxidation behaviour of silicon carbide. In this paper a comprehensive review has been made on different works related with the oxidation behaviour of silicon carbide.

1. INTRODUCTION

Silicon Carbide (SiC) made up of silicon and carbon, can be found in nature as extremely rare mineral called moissanite. It was first discovered by H. Moissan on a meteorite located in the Diablo Canyon of Arizona in 1893. Before that in 1891, E. G. Acheson synthesized silicon carbide in the laboratory and coined the name 'Carborundum'.

SiC has a three dimensional structure of Si and C atoms, each atom tetrahedrally surrounded by four of the other kind. It can exist in more than 250 polymorphs. Some of them present as amorphous phases in thin films and fibers where as others form a large family of similar crystalline structures called polytypes which are identical in two dimensions and differ in the third. Two major polytypes of silicon carbide are alpha silicon carbide (α -SiC) having hexagonal crystal structure (similar to Wurtzite) and the beta modification (β -SiC), with a zinc blende crystal structure (similar to diamond). Thermody-

namically α -form is marginally more stable than the β -form.

Pure SiC is pale yellow to colourless in appearance. But it is usually obtained in dark coloured form because of the presence of impurities like iron and iridescence due to formation of a very thin layer of SiO₂ by surface oxidation. SiC has some unique physical properties like high thermal and chemical stability, high strength, high thermal conductivity, controllable electrical conductivity, low temperature coefficient of linear expansion, etc. Based on the properties, it is widely used as refractory material [1]. SiC is a potential semiconductor material for high power, high frequency and high temperature application. SiC based materials are increasingly used in coating to protect materials from corrosive environments, combustion engines [2-6], etc.

SiC is a non-oxide and it has the tendency to get oxidized in presence of oxygen or oxidizing agents. Depending upon the nature of the substrate

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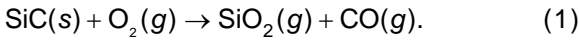
used, atmosphere, oxygen partial pressure, etc., oxidation of silicon carbide and its composites may lead to different results. During last few decades, numerous works were carried out on the oxidation behaviour of SiC and its composites by different researcher and in this paper a comprehensive review on these works has been presented.

2. VARIOUS ASPECTS OF SILICON CARBIDE OXIDATION PROCESS

2.1. Mode of oxidation

Depending upon the oxygen concentration, high temperature oxidation of silicon carbide may be either active or passive [7-27].

Active oxidation occurs at oxygen pressure less than one bar according to the following equation



SiO formed gets vaporised after its formation leading to loss of mass.

Passive oxidation proceeds at oxygen pressures close to one bar by the following reaction:



SiO₂ forms during passive oxidation get deposited over the surface of SiC leading to net increase in the mass. It was observed that SiO₂ formed at lower temperature develops layer over the surface of SiC and protects it from further catastrophic oxidation to SiO occurred at higher temperatures. Such protective action continues up to the melting point of SiO₂ (1996K) [28].

A number of studies were performed on the two modes of oxidation [29-35]. Most of these works were based on either Wagner's initial analysis for Si [36] or Turkdogan theory for enhanced metal vaporization [37].

Based on Wanger's theory, Hunze and Graham [22] derived the following expression to determine the partial pressure of oxygen for active-to-passive transition

$$P_{\text{O}_2}^{\text{active-to-passive}} = \left(\frac{D_{\text{SiO}}}{D_{\text{O}_2}} \right)^{\frac{1}{2}} \cdot P_{\text{SiO}}^{\text{eq}} \quad (3)$$

where, D_{SiO} and D_{O_2} are the gas-phase diffusivities of SiO(g) and O₂(g) respectively and $P_{\text{SiO}}^{\text{eq}}$ is the equilibrium pressure of SiO(g). To calculate gas-phase diffusivities, Chapman-Enskog correlation was used [38].

Heuer and Lou [39] modified the model of Turkdogan and proposed that when $P(\text{O}_2) =$

$1/2P(\text{SiO})$, active-to-passive transition will be possible. With the help of Turkdogan's theory, Harder et al. [40] derived the expression for the $P(\text{O}_2)$ for the passive-to-active transition,

$$P_{\text{O}_2}^{\text{passive-to-active}} = \frac{\alpha P(\text{SiO})}{2h} \cdot \sqrt{\frac{RT}{2\pi M}}, \quad (4)$$

where, α is the evaporation coefficient, M is molecular weight of SiO₂, R is the gas constant, T is the absolute temperature, h is the mass transfer coefficient. The evaporation coefficient value describes the reduced pressure generated due to kinetics factor. The values of α are 8.5×10^{-4} and 5.4×10^{-4} for SiC/SiO₂ and SiC-C-SiO₂ reactions, respectively [40].

After formation of SiO₂ layer during passive oxidation, further oxidation proceeds by the following steps [41]:

- Gaseous oxygen molecules is transported to the surface of oxide and comes in contact with SiC through the oxide film by diffusion;
- Reaction of diffused oxygen with SiC at the oxide/SiC interface;
- Product gases formed comes out through the oxide film again by diffusion.

At the beginning of the oxidation at lower temperature, the oxidation product is amorphous which tend to be crystalline at higher temperature with longer period of exposure. The mode of oxidation greatly affects the strength of SiC. During active oxidation, the strength of the material is affected by the introducing surface flaws or load-bearing cross-section. In active oxidation range, the reductions in strength of the samples were linearly proportional to the measured weight loss. At lower temperature formation of a thin SiO₂ layer by pre-oxidation was found to be ineffective to protect SiC from active oxidation [42]. Passive oxidation makes the SiC stronger as thin layer of SiO₂ blunts strength-limiting surface flaws and useful in different fields [6,43].

2.2. Anti-oxidation protective coating

In most of the cases, SiC oxidation is passive with wide variation in the reaction rates and the morphology of the reaction products [44]. During passive oxidation liberated SiO₂ forms a dense layer on the surface of SiC which acts as an anti-oxidation protective layer [45,46]. Lin et al. [47] describe a process to develop multi-functional anti-oxidation protective coating of silicon carbide or SiC/SiO₂ layers on the surface of the carbon/carbon composite by a single step electrically induced liquid infiltration (EILI) method. Studies showed that the layers formed on

the composite surfaces acts as physical protection barriers for oxygen penetration. Mukherjee et al. [48] described a modified chemical vapour deposition process of liquid polycarbosilane derived SiC coating on silicon wafer in presence of argon gas. Observations showed smooth coating of β -SiC which subsequently transferred into α -SiC. Such coating enhanced hardness and toughness of the samples. Yang et al. [49] described a simple and low cost method of oxidation protective ZrB_2 /SiC coating on C/SiC composites produced by precursor infiltration and pyrolysis (PIP). At first ZrB_2 was mixed with SiC and then fabricated by painting slurry on the surface of the composite. Finally SiC was deposited by using chemical vapour deposition (CVD) technique on the top. It was observed that such composite coating can protect C/SiC composites at very high temperature ($\sim 1973K$) and improved ablation resistance by forming zirconia and silicon dioxide which absorbed heat from the flame and reduced the erosive attack to carbon fibres and SiC matrix. Li et al. [50] observed that formation of a thin amorphous SiO_2 coating on the surface of multi-walled carbon nanotubes (MWCNTs) can improve its oxidation resistance.

Recent studies showed that at high temperature, nanowire toughened SiC coatings exhibited excellent oxidation and thermal shock resistance [51,52]. Yanhui et al. [53] studied the effect of addition of SiC nanowires in Si-Mo-Cr composite coating deposited on carbon/carbon composites by chemical vapour deposition method. Experimental results revealed that addition of SiC nanowires significantly increased the hardness, elastic modulus and fracture toughness. Again by incorporating SiC nanowires, the weight loss of the coated carbon/carbon samples was also decreased. Yang et al. [54] utilized chemical vapour deposition method to prepare double layer nanostructure SiC coating on carbon/carbon composites to protect it from oxidation. The crystalline structured coating having uniform thickness distribution and limited porosity, showed good compatibility and adherence with the substrate. Large SiC crystals formed nearly a dense structure along with microcrystals, nanocrystals and nanowires of SiC. Crystalline structured coating adhere well with the substrate and at high temperature, such coating exhibited excellent antioxidation behavior and thermal shock resistance.

2.3. Oxidation kinetics

It has been observed that high temperature oxidation kinetics of silicon carbide followed diffusion

controlled mechanism with parabolic rate law [55-69]. During SiC oxidation, silica rich surface layer developed above $600^\circ C$ and oxidation process was controlled by the diffusion of oxidant through the amorphous silica [41,70]. Experiments showed that at lower temperatures the oxidation process was controlled by the transport of molecular oxygen where as ionic oxygen transportation dominates the oxidation process at higher temperatures [11]. Costello et al. [71] determined the parabolic rate constants corresponding to the short-time data (up to 100 min) and activation energies of sintered and hot-pressed silicon carbide. The results indicated that oxidation resistance of sintered material is higher compared with hot-pressed material as rate constant of the latter is larger than that of former. Ebrahimpour et al. [72] studied the oxidation behavior of SiC powders by performing a thermogravimetric analysis (TGA). It was observed that oxidation rate was controlled by diffusion (bulk, intra and inter particle diffusion) and chemical reaction. Hou et al. [73] studied oxidation kinetics of SiC powders in isothermal modes by thermogravimetry (TG) in the temperature range from 1623-1673K and observed that the oxidation rate followed diffusion controlled mechanism.

2.4. Model to describe oxidation process

Deal and Grove [74] proposed a model to describe the oxidation process of SiC. The model was later modified by Song et al. [41] by considering the outward diffusion of gases formed during oxidation. They reported that anisotropic oxidation from the surface structure was influenced due to severe differences between the physico-chemical properties of four surfaces. Fahrenholtz [75] developed a thermodynamic model to explain the formation of a SiC-depleted layer due to active oxidation of ZrB_2 -SiC composites in air at $1500^\circ C$. The model predicted the formation of a structure consisting of thermodynamically stable SiO_2 -rich layer; Zn-rich oxidized layer and SiC-depleted zirconium diboride layer. It was observed that although oxidation of SiC was in the active region, the overall process was consistent with the passive oxidation by the formation of a protective surface layer. Hou et al. [76] developed a new model for the study of oxidation kinetics of SiC and express the weight gain during oxidation as a function of temperature, oxygen partial pressure and size of the materials used. They applied this model to chemical vapour deposited SiC pallet and ZrB_2 -SiC pallet and observed that this model

could be applied in both isothermal and non-isothermal condition. Shah et al. [77] developed a statistical model for SiC oxidation by determining the change in weight with time and assuming parabolic rate law based on diffusion-controlled mechanism for the growth of the oxide passivation layer. They considered that at the very early stage, oxidation was controlled by interfacial reaction mechanism. Statistical analysis gave much better result compared with mono sized particle analysis for the determination of the parabolic rate constant of oxidation.

2.5. Thickness of oxide layer

It has been observed that oxide layer thickness increases with the temperature and time up to certain range [78]. Weight gain during oxidation and thickness of oxide layer formed hold parabolic relationship with the oxidation time [9,24-25,55-56,73]. To determine the thickness of oxide layer, Rutherford backscattering spectrometry (RBS) was observed to a powerful tool which was also capable of characterizing the thickness of the oxide layer and the subsurface damage. The thickness of the oxide layer can be calculated using the following formula [11]:

$$t = \frac{\Delta E}{|\epsilon|_{\text{Si}} N} \quad (5)$$

Here ΔE = energy difference between two layers, N = no of Silicon atoms, $|\epsilon|_{\text{Si}}$ = stopping cross section factor for Silicon.

It has been observed that even at the smallest exposures to O_2 , oxidation of Si-surface could be possible but no such discernible change was noted on the C-terminated surface [15]. The thickness of coating formed on SiC surface was found to be the maximum on carbon face whereas minimum thickness was measured for silicon faces. The activation energy of coating formation on these faces followed completely reverse trend. The variation of activation energy was related with the large driving force for C atoms on the C face which causes easy breakage of Si-C bond to form very stable C-O bond [79,80]. Hornetz et al. [81] reported that the thicker interface layer on the Si face hinders the oxidation, so that the resulting oxide thickness on the Si face is smaller. As the thickness of SiO_2 films increased either due to exposure at longer times or at higher temperatures, some new defects like bubbles and pits are generated in the film but the strength remained the same or it decreases slightly [82]. Atomic force microscopic (AFM) studies on the Si

surface after the removal of thermal oxide layers showed uniform pits with a typical depth of 10-15 nm and a diameter of 200 nm. The formation of these pits is possibly related to the preferential oxidation at the site of dislocations [83,84]. Again formation of porous oxide was also observed after the oxidation process and the porosity of the oxide was found to be propagated onto the SiC surface [85]. Formation of this layer inhibits further oxidation of SiC compound. During oxidation etching of the surface and surface segregation of carbon was also observed [57].

3. FACTORS INFLUENCING THE OXIDATION PROCESS

Oxidation process is greatly influenced by the factors like nature of substrate, moisture in the environment, particle size, sintering additives used, etc.

3.1. Nature of the substrate

Quality of the substrate greatly influenced the oxidation kinetics of SiC as rate of oxidation depends on SiC terminal faces. The degree of hexagonality determined the oxidation kinetics on Si-face where as substrate conductivity is the key factor for oxidation on C-face [86,87]. Presence of impurities in the substrate also plays an important role as the diffusion of CO or CO_2 from SiC-oxide interface to the surface as well as diffusion of oxygen molecules or oxygen ions from the surface to the interface through the oxide layer is responsible for oxidation process [9,41].

3.2. Moisture content

The oxidation behavior of SiC in dry atmosphere [70,88-90] and wet atmosphere [91-93] were studied extensively. Wet oxidation carried out at comparatively lower temperature by supplying a mixture of saturated water vapour and O_2 to the oxidation furnace whereas oxidation at higher temperature (1000-1300 °C) in dry environment leads to dry oxidation. In wet oxidation, water vapor in the atmosphere greatly accelerated the oxidation process compared with that in dry atmosphere. But the degradation in strength by the oxidation in dry and wet atmosphere was not so prominent. During wet oxidation, growth rate is much faster than that in dry atmosphere because leaving water molecule has much higher solubility in SiO_2 than O_2 ; therefore it provides greater supply of oxidizing species to the SiC surface. As a result thicker oxide layer is formed in the case of wet oxidation. But the oxide quality

and dielectric breakdown strength are very high in the case of dry oxidation.

3.3. Particle size

Size of SiC particles plays an important role during oxidation process. In case of nano-sized SiC powders, the oxidation occurs at comparatively lower temperature to that in case of micro meter-sized SiC powders [94,61]. Hou et al. [95] showed that the particle size has a great effect on the oxidation kinetics. As the particle size became finer the activation energy of the oxidation process was also tending to reduce. During oxidation kinetics study of micro-sized and nano-sized SiC powders by using thermogravimetry Quanli et al. [96] observed that oxidation process of SiC powders was strongly influenced by particle size. With decreasing the particle size, the weight gain and rate constant increased. Again the weight gain process of both micro-sized and nano-sized SiC powders followed parabolic relationship with oxidation time over 1100-1200 °C. The activation energy values were about 110.74 and 82.64 kJ mol⁻¹ in case of micro-sized and nano-sized SiC powders respectively.

3.4. Presence of additive

It has been observed that oxidation rates are adversely affected in the presence of additives [88]. Because of covalent bonding, densification of SiC is a difficult process without additive. The crystallization of the oxide film is also enhanced in presence of additive. Again during oxidation, the additive gets redistributed into the oxide film and lowers the viscosity which can increase the transport of the oxidant through the film. This process is temperature dependent and affects the activation energy of the process [97-101]. Biswas et al. [102] showed that high temperature oxidation behaviour of liquid-phase-sintered (LPS) SiC in presence of Lu₂O₃-AlN additive in air between 1200 and 1500 °C followed parabolic law. Below 1400 °C, oxidation resistance of the material was high but above this temperature significant degradation in oxidation kinetics was observed due to the formation of a low-melting eutectic as a result of a reaction between the growing oxide layer (mainly SiO₂) and the second phase (Lu₂O₃). Noviyanto et al. [103] carried out different experiments to get an idea about the effectiveness of various sintering additives for β-SiC at high temperatures. It was observed that Al₂O₃, MgO, Y₂O₃ and their mixed systems were effective sintering additives as these systems were unreactive

with β-SiC at high temperatures. Kumar et al. [104] studied the high temperature (1300-1500 °C) oxidation behaviour of hot-pressed SiC and its composites containing 10-30 vol.% molybdenum disilicide processed with aluminium nitride and yttria additives. It was observed that oxidation resistance of SiC was improved by the addition of molybdenum disilicide where as sintering additives influenced the overall oxidation kinetics. Zawrah et al. [105] observed that in the presence of aluminium metal the oxidation resistance of SiC was enhanced and the rate of densification process also increased. Shen et al. [106] observed that addition of boron nitride into SiC improved both oxidation resistances during 800-1100 °C and compression strength of SiC.

3.5. Oxidation of silicon carbide in composites

SiC is useful for synthesizing composites materials with other substances to enhance different properties of such substances. Oxidation of such composites is also an interesting topic for research. There are a number of methods available to synthesis such composites materials [107-115]. It has been observed that the presence of SiC in such materials enhances different properties [116-124]. Molybdenum (Mo) addition to SiC showed high density, good mechanical properties and oxidation resistance of these composites. Refractoriness of such composites also verified during high temperature long term oxidation [125]. Patel et al. [126] studied the effect of SiC concentration (10-30 vol.%) on the strength of hot pressed ZrB₂-SiC composites during exposure to high temperatures (1000-1700 °C) for 5 h in air. After exposure, multilayer oxide scale structures were developed whose composition and thickness were dependent on exposure temperature and SiC concentration in composites. The residual strength of the composites having higher SiC concentration (20 and 30 vol.% SiC) was improved at lower temperature (~1000 °C) but drastically reduced at higher temperature (~1700 °C). To predict the oxidation behaviour of C-SiC composite, Roy [127] developed mathematical theory and numerical scheme derived from the mechanics of the flow of ideal gases through porous solid. During oxidation of ZrB₂-SiC composites, it was observed that SiO₂ formed during oxidation is thermochemically stable below 1800 °C and will lose its protective properties at temperatures above 2300 °C [128]. Han et al. [129] studied the oxidation resistance capacity of hot-pressed ZrB₂-20 vol.% SiC using an oxy-acetylene torch. No macro-cracks or spallation were

detected even at 2200 °C which indicated super oxidation resistance capacity of the composite. The oxidation behaviour of HfB_2 -SiC monoliths in the 20–1700 °C temperature range in flowing O_2/He mixtures exhibited good oxidation resistance almost up to 1700 °C [130]. Monteverde et al. [131] investigated the oxidation resistance of HfB_2 -SiC composite through non-isothermal and isothermal treatments at temperatures up to 1600 °C in air. It was observed that that initial heating (up to 10 h), the oxidation kinetics followed para-linear law and after that the weight gaining process followed linear plot. Oxidation resistance was improved markedly in presence of SiC due to the formation of a protective borosilicate glassy coating on the outer surfaces. Kovalcikova et al. [132] studied the oxidation behaviour of liquid-phase-sintered SiC-Si₃N₄. The oxidation followed parabolic rate with diffusion controlled mechanisms and resistance property was enhanced with increasing temperature from 1650 to 1850 °C. Maity et al. [133] observed the oxidation process of Si/SiC ceramic composite synthesized from processed cellulosic bio-precursor in dry air over the temperature range 1200–1350 °C. The process followed a parabolic equation with activation energy equal to 141.4 kJ/mol. Oxidations of both SiC and Si phases were controlled by the transport of molecular oxygen through the growing oxide layer. Yue et al. [134] reported that with increasing temperature oxidation of ZrB_2 -ZrC-SiC ceramics composites changes from reaction-controlled process to diffusion-controlled. Again oxidation resistance of such composites was found to decay at elevated temperatures. Guo et al. [135] studied the oxidation behavior and the effect of the amount of SiC added on oxidation resistance of different compositions of ZrB_2 -MoSi₂-SiC composites in dry air at 1500 °C for up to 10 h. Oxidation process of both the composites followed parabolic rate law. In all the specimens oxidation resistance was also improved in presence of SiC and improvement was enhanced with amount of SiC added due to formation of a thicker dense outermost scale layer which inhibited inward diffusion of oxygen through it.

4. APPLICATION OF SiC AND IT'S COMPOSITES

The most common use of SiC and its composites is as refractory material not only because of its high melting point (~2730 °C), but also of high thermal stability and thermal strength (~800-1000 MPa), fracture toughness (6-10 MPa.m^{1/2}). Again due to formation of protective coating on its surface at high

temperature, it can be used in aggressive environments [136].

SiC refractories are available as both non-oxide and composite refractories. The base of most of the SiC refractories is the α -SiC crystals. Depending on bonding, two forms of SiC refractories are available: one type where grains of SiC are inter-connected (self bonded) and another type where SiC grains are connected with one or more than one of the following components: graphite, silicon, silica, silicon nitride, periclase, mullite, sillimanite, cristoballite, corundum and other phases that contain iron, calcium, titanium etc. as impurities [137]. The major chemical components and major phases exist in different SiC refractories are listed in the following table (Table -1).

Various studies were performed on SiC-based refractory material [138-148]. Nourbakhsh et al. [149] investigated the development of microstructure of nitride-bonded SiC refractories in the presence of Al_2O_3 and MgO. It was observed that MgO addition decreased the strength of the refractories whereas in presence of alumina, the strength decreases with increasing concentration of Al_2O_3 mainly due to liquid phase formation and grain morphology.

By varying the amount of SiC and graphite, materials of the system Al_2O_3 -SiC-C can be extensively used as unmolded refractories [150]. Most important concrete of the system Al_2O_3 -SiC-C is one which contains an antioxidant, a small amount of organic fiber (1%) and fillers Al_2O_3 /SiC in almost 1:1 ratio. Antioxidant is added in order to protect graphite from oxidation. Organic fiber is used to develop the desired matrix structure of finely ground binder component of concrete during heat treatment. With the presence of considerable amount of SiC in the mixture and depending on charge composition, a combined binder consisting of high-alumina cement and refractory clay in an amount from 1 to 10% can be used [151]. Sivov [152] developed a method of production of clay-SiC refractories on the basis of broken SiC plates by slip casting method considering multiple heating and chilling. It was observed that such materials can be used as refractory stacks and sagers in the production of fine ceramics.

The thermal conductivities of SiC-based refractories are superior to that of alumina or chrome based refractories. Again monolithic SiC refractories have good corrosion resistance against coal ash. So such refractories can be used as protective coating on ceramic heat exchangers in advanced coal combustion system [153,154].

Silicate bonded SiC refractories are widely used in metallurgical industries for manufacturing of

Table 1. Classification of SiC refractories [137].

Type of refractories	Major Chemical Components	Major phases
SiC	SiC	SiC
SiC-C	SiC and C	α -SiC and Graphite
SiC-Si ₃ N ₄	SiC and Si ₃ N ₄	α -SiC and ($\alpha + \beta$) Si ₃ N ₄
Al ₂ O ₃ -SiO ₂ -C-SiC	SiO ₂ , Al ₂ O ₃ , SiC	Mullite/andalusite, cristoballite, SiC
	SiO ₂ , Al ₂ O ₃ , SiC, C	Mullite/andalusite, cristoballite, SiC, graphite

bricks, muffles, slabs, tiles, crucibles, kiln furniture etc. as such refractories have good strength and good thermal, corrosion and abrasion resistance. Reddy [155] discussed a method of preparation of silicate bonded SiC refractories with molybdenum disilicate (4 wt.%) infiltration. For bonding of SiC, mullite and sillimanite powders were used. It was observed that modulus of rupture (MOR) increases with increase of amount of sillimanite or mullite. Again, MoSi₂ infiltration improved the strength of SiC refractories and lowered the thermal expansion coefficient.

Foamed SiC refractories have the potential to use as heat-insulating and heat protective materials. Guzman et al. [156] discussed a method for production of foamed nitride bonded SiC refractories by firing porous blanks, obtained from a mixture of SiC and Si (3:2), in nitrogen atmosphere. The product obtained with silicon oxynitride at the intermediate layer and silicon nitride at the centre. Such nitride bond enhanced the quality of the products.

SiC refractories can also be used for lining blast furnaces as it has good alkali resistance property. Pitak et al. [157] studied the resistance of SiC to the action of alkalis and slag. It was observed that up to 1200 °C, the quantity of SiC in the mixture containing alkalis decreases as compared to that in the original mixture mainly due to oxidation of SiC to SiO₂. But after 1200 °C, with increasing time, a thin SiO₂ film form over the surface of SiC grains protecting it from further oxidation. Such protective action was continued up to 1500 °C.

Besides use as refractory, silicon carbide based compounds have other uses also. As its atomic number is low, it can be used as wall material for fusion reactor [158,159]. Due to its large band gap (3 eV) leading to low leakage of currents and higher electronic breakdown voltage, it is used in electronic device making [160,161]. Pure SiC is an intrinsic semiconductor, but in presence of controlled amounts of impurities SiC can be used as potential extrinsic semiconductor for high power, high fre-

quency and high temperature application etc [162,163]. Again it is the only compound semiconductor that can be thermally oxidized to form SiO₂ [164]. Oxidation of SiC results into an anisotropic coating with SiO₂ on its surface which has several remarkable applications like fabrication of metal oxide semiconductor (MOS) structures and metal oxide semiconductor field effect transistors (MOSFET), manufacturing of reverse voltage blocking diodes of capacity > 1 kV, for masking operations in doping process as passive film for p-n junctions or as a gate dielectric [165-168] etc. The film formed can also heal deep surface cracks [169].

5. CONCLUSION

A review work was carried out on the oxidation behavior of SiC based compounds. SiC has some unique properties like high hardness, toughness, chemical and thermal stability etc. Based on this property SiC based compounds are widely used in refractory, electronic device making, space applications etc. Presently it is used in coating industry for environmental protection of high-temperature materials and combustion engines by utilizing its antioxidant property. A scan of contemporary literatures reveals that depending upon reaction conditions like oxygen concentration, environmental ambience etc., its oxidation may be either active or passive. During active oxidation, the strength of the samples was reduced linearly with respect to the measured weight loss. But in passive oxidation range, SiO₂ layer formed makes a coating over SiC substrates which protect it from further oxidation. From different studies it has been observed that passive oxidation makes the SiC surface stronger and makes its applicable in most of the fields.

REFERENCES

- [1] N. N. Greenwood and A. Earnshaw, *Chemistry of the Elements-Second Ed.* (Butterworth Heinemann, Oxford, 1998).

- [2] Z. Zheng, R. E. Tressler and K.E. Spear // *Corros. Sci.* **33** (1992) 545.
- [3] J. J. Burke, A. E. Gorum and R. N. Katz, *Ceramics for High Performance Applications I.* (Brook Hill Publishing Co. Chestnut Hill, MA, 1974).
- [4] J. J. Burke, A. E. Gorum and R. N. Katz, *Ceramics for High Performance Applications I.* (Brook Hill Publishing Co. Chestnut Hill, MA, 1977).
- [5] P. Krawiec and S. Kaskel // *J. Solid State Chem.* **179** (2006) 2281.
- [6] S. R. Levine, E. J. Opila, M. C. Halbig, J. D. Kiser, M. Singh and J. A. Salem // *J. Eu. Ceram. Soc.* **22** (2002) 2757.
- [7] E. J. Opila // *J. Am. Ceram. Soc.* **78** (1995) 1107.
- [8] E. J. Opila and R. E. Hann Jr. // *J. Am. Ceram. Soc.* **80** (1997) 197.
- [9] E. J. Opila // *J. Am. Ceram. Soc.* **77** (1994) 730.
- [10] L. U. J. T. Ogbuji and E. J. Opila // *J. Electrochem. Soc.* **14** (1995) 925.
- [11] Z. Zheng, R. E. Tressler and K.E. Spear // *J. Electrochem. Soc.* **137** (1990) 850.
- [12] K. L. Luthra // *J. Am. Ceram. Soc.* **74** (1991) 1095.
- [13] W. F. Du, H. N. Xiao and H.Q. Du // *J. Chin. Ceram. Soc.* **20** (1992) 417.
- [14] T. Y. Tian, Y. J. Zhang, F. Sun, and H. K. Wei // *Chin. Ceram.* **5** (2006) 19.
- [15] C. Virojanadara and L. I. Johansson // *Surf. Sci.* **519** (2002) 73.
- [16] M. Kildemo, U. Grossner, M. Juel, B. G. Svensson and S. Raaen // *Surf. Sci.* **600** (2006) 1300.
- [17] K. Knel, D. Sciti, E. Landi and A. Bellosi // *Appl. Surf. Sci.* **210** (2003) 274.
- [18] C. E. Ramberg and W. L. Worrell // *J. Am. Ceram. Soc.* **84** (2001) 2607.
- [19] E. A. Gulbransen, K. F. Andrew and F. A. Brassart // *J. Electrochem. Soc.* **113** (1966) 1311.
- [20] G. Ervin // *J. Am. Ceram. Soc.* **41** (1958) 347.
- [21] T. Narushima, T. Goto and T. Hirai // *J. Am. Ceram. Soc.* **72** (1989) 1386.
- [22] W. Hinze and H. C. Graham // *J. Electrochem. Soc.* **123** (1976) 1066.
- [23] E. A. Gulbransen and S. A. Jansson // *Oxid. Met.* **4** (1972) 181.
- [24] C. E. Ramberg, G. Cruciani, K. E. Spear, R.E. Tressler and C. F. Ramberg Jr. // *J. Am. Ceram. Soc.* **79** (1996) 730.
- [25] L. U. J. T. Ogbuji // *J. Am. Ceram. Soc.* **80** (1997) 1544.
- [26] H. Cappelen, K. Johansen and K. Motzfeldt // *Acta Chem. Scand.* **A35** (1981) 247.
- [27] D. E. Rosner and H. D. Allendorf // *J. Phys. Chem.* **74** (1970) 1829.
- [28] C. Varadachari, R. Bhowmick and K. Ghosh // *ISRN Thermodyn.* **2012** (2012) 1.
- [29] W. L. Vaughn and H. G. Maahs // *J. Am. Ceram. Soc.* **73** (1990) 1540.
- [30] K. G. Nickel // *J. Eu. Ceram. Soc.* **9** (1992) 3.
- [31] M. J. H. Balat // *J. Eu. Ceram. Soc.* **16** (1996) 55.
- [32] B. Schneider // *J. Mater. Sci.* **33** (1998) 535.
- [33] J. Wang // *J. Am. Ceram. Soc.* **91** (2008) 1665.
- [34] N. Jacobson and D. Myers // *Oxid. Met.* **75** (2011) 1.
- [35] N. Jacobson, B. Harder and D. Myers // *J. Am. Ceram. Soc.* **96** (2013) 838-844.
- [36] C. Wagner // *J. Appl. Phys.* **29** (1958) 1295.
- [37] T. Turkdogan, P. Grieveson and L. S. Darken // *J. Phys. Chem.* **67** (1963) 1647.
- [38] G. H. Geiger and D. R. Poirier, *Transport Phenomena in Metallurgy* (Addison-Wesley Publishing Company, Reading, MA, 1973).
- [39] A. H. Heuer and V. L. K. Lou // *J. Am. Ceram. Soc.* **73** (1990) 2789.
- [40] B. Harder, N. Jacobson and D. Myers // *J. Am. Ceram. Soc.* **96** (2013) 606.
- [41] Y. Song, S. Dhar, L. C. Feldman, G. Chung and J. R. Williams // *J. Appl. Phys.* **95** (2004) 4953.
- [42] H-E Kim and A. J. Moorhead // *J. Am. Ceram. Soc.* **73** (1990) 1868.
- [43] Y. Lin and L. Chen // *Ceram. Int.* **26** (2000) 593.
- [44] H. Du, R. E. Tressler and K.E. Spear // *J. Electrochem. Soc.* **136** (1989) 3210.
- [45] P. I. Jorgensen, M. E. Wadsworth and I. B. Cutler // *J. Am. Ceram. Soc.* **42** (1959) 613.
- [46] M. Auweter-Kurtz, G. Hilfer, H. Habiger, K. Yamawaki, T. Yoshinaka and H. D. Speckmann // *Acta Astronau.* **45** (1999) 93.
- [47] Y. C. Lin, E. M. Ruiz, R. G. Rateick Jr, P. J. McGinn and A. S. Mukasyan // *Carbon* **50** (2012) 557.
- [48] J. Mukherjee, A. Ranjan, A. K. Saxena, P. K. Das and R. Banerjee // *Appl. Surf. Sci.* **270** (2013) 219.
- [49] X. Yang, L. Wei, W. Song, Z. Bi-Feng and C. Zaho-Hui // *Composites, Part B* **45** (2013) 1391.

- [50] Y. Li, M. Luo, S. Jin, S. Sang and L. Zhao // *Ceram. Int.* **38** (2012) 4105.
- [51] Y. H. Chu, H. J. Li, Q. G. Fu, H. P. Wang, X. H. Hou, X. Zou and G. N. Shang // *Carbon* **50** (2012) 1280.
- [52] X. F. Qiang, H. J. Li, Y. L. Zhang, D. J. Yao, L. J. Guo and J. E. Wei // *Corros. Sci.* **59** (2012) 343.
- [53] C. Yanhui, H. Li, F. Qiangang, S. Xiaohong, Q. Lehua and W. Bingbo // *Corros. Sci.* **58** (2012) 315.
- [54] X. Yang, Q. Huang, Z. Su, L. Chai, X. Wang and L. Zhou // *Ceram. Int.* **39** (2013) 5053.
- [55] D.M Liu // *Ceram. Int.* **23** (1997) 425.
- [56] K. N. Lee and R. A. Miller // *Chem. Inform. Abstract.* **27** (1996) 620.
- [57] Y. Song and F. W. Smith // *J. Am. Ceram. Soc.* **88** (2005) 1864.
- [58] M. Yoshimura, T. Ohji and K. Nihara // *J. Am. Ceram. Soc.* **80** (1997) 797.
- [59] H. Y. Liu, K. L. Weisskopf, M. J. Hoffmann and G. Petzow // *J. Eu. Ceram. Soc.* **5** (1989) 123.
- [60] R. Vaben and D. Stover // *J. Mater. Sci.* **29** (1994) 3791.
- [61] Y. Oyama and K. Kamigaito // *Jpn. J. Appl. Phys.* **10** (1971) 1637.
- [62] X. B. Li, Q. S. Zhou, Y. X. Liu, X. Xiang and H. Jie // *Chin. J. of Nonferrous Met.* **10** (2000) 560.
- [63] J. W. Fergus and W. L. Worrell // *J. Am. Ceram. Soc.* **78** (1995) 1961.
- [64] D. Das, J. Farjas and P. Roura // *J. Am. Ceram. Soc.* **87** (2004) 1301.
- [65] R. F. Adamsky // *J. Phys. Chem.* **63** (1959) 305.
- [66] F. E. Wagstaff // *J. Am. Ceram. Soc.* **49** (1966) 118.
- [67] K. Motzfeldt // *Acta. Chem. Scand.* **18** (1964) 1596.
- [68] J. Schlichting and K. Kriegsmann // *Ber. Dtsch. Kerum. Ges.* **56** (1979) 72.
- [69] S. C. Singhal // *J. Mater. Sci.* **11** (1976) 1246.
- [70] A. H. Heuer, L. U. Ogbuji and T. E. Mitchell // *J. Am. Ceram. Soc.* **63** (1980) 354.
- [71] J. A. Costello and R. E. Tressler // *J. Am. Ceram. Soc.* **64** (1981) 327.
- [72] O. Ebrahimpour, J. Chaouki and C. Dubois // *J. Mater. Sci.* **48** (2013) 4396.
- [73] X. Hou, G. Zhang and K. Chou // *J. Alloys Compd.* **477** (2009) 166.
- [74] B. E. Deal and A. S. Grove // *J. Appl. Phys.* **36** (1965) 3770.
- [75] W. G. Fahrenholtz // *J. Am. Ceram. Soc.* **90** (2007) 143.
- [76] X. Hou, K. Chou and F. Li // *Ceram. Int.* **35** (2009) 603.
- [77] S. R. Shah, A. Saha and R. Raj // *J. Am. Ceram. Soc.* **86** (2003) 351.
- [78] P. J. Macfarlane and M. E. Zvanut // *Micorelectron. Eng.* **48** (1999) 269.
- [79] K. Christiansen and R. Helbig // *J. Appl. Phys.* **79** (1996) 3276.
- [80] M. Kanaya, H. Yashiro, N. Ohtani, M. Katauno, J. Takahashi and S. Shinoyama // *Mater. Sci. Forum* **264-268** (1998) 359.
- [81] B. Hornetz, H-J. Michel and J. Halbritter // *J. Mater. Res.* **9** (1994) 3088.
- [82] T. E. Easler, R. C. Bradt and R. E. Tressler // *J. Am. Ceram. Soc.* **64** (1981) 731.
- [83] S. E. Sadow, T.E., Schattner, J. Brown, L. Grazulis, K. Mahalingam, G. Landis, R. Bertke and W. C. Mitchel // *J. Electron. Mater.* **30** (2001) 228.
- [84] C. Li, R. Wang, J. Seiler and I. Bhat // *Mater. Sci. Forum.* **457-460** (2004) 801.
- [85] W. V. Muench and I. Pfaffeneder // *J. Electrochem. Soc.* **122** (1975) 642.
- [86] J. W. Hinze, W. C. Tripp and H. C. Graham, In: *The High-Temperature Oxidation of Hot-pressed Silicon Carbide. Mass Transport Phenomena in Ceramics*, ed. by A. R. Cooper and A. H. Heuer (Plenum Press, New York, 1975), p. 409.
- [87] S. C. Singhal and F. F. Lange // *J. Am. Ceram. Soc.* **58** (1975) 1433.
- [88] D. M. Mieskowski, T. E. Mitchell and A. H. Heuer // *J. Am. Ceram. Soc.* **67** (1984) C-17.
- [89] J. A. Costello and R. E. Tressler // *J. Am. Ceram. Soc.* **69** (1986) 674.
- [90] P. J. Jorgensen, M. E. Wadsworth and I. B. Cutler // *J. Am. Ceram. Soc.* **44** (1961) 258.
- [91] S. C. Singhal // *J. Am. Ceram. Soc.* **59** (1976) 81.
- [92] M. Maeda, K. Nakamura and T. Ohkubo // *J. Mater. Sci.* **23** (1988) 3933.
- [93] M. Mitomo, Y. W. Kim and H. Hirotsuru // *J. Mater. Res.* **11** (1996) 1601.
- [94] S. Wu and N. Claussen // *J. Am. Ceram. Soc.* **77** (1994) 2898.
- [95] X. Hou and K. Chou // *Corros. Sci.* **50** (2008) 2367.
- [96] J. Quanli, Z. Haijun, L. Suping and J. Xiaolin // *Ceram. Int.* **33** (2007) 309.

- [97] J. Schlichting // *Ber. Dtsch. Keram. Ges.* **56** (1979) 196.
- [98] J. Schlichting // *Ber. Dtsch. Keram. Ges.* **56** (1979) 256.
- [99] S. C. Singhal, In: *Properties of High-temperature Alloys*, ed. by Z. A. Foroulis and F. S. Pettit (The Electrochem. Soc. Princeton, NJ, 1976), p. 697.
- [100] J. Schlichting and K. Schwetl // *High Temp. High Press.* **14** (1982) 219.
- [101] J. A. Costello, R. E. Tressler and S. T. Tsong // *J. Am. Ceram. Soc.* **64** (1981) 332.
- [102] K. Biswas, G. Rixecker and F. Aldinger // *Mater. Sci. Eng. A.* **374** (2004) 56.
- [103] A. Noviyanto and D. H. Yoon // *Curr. Appl. Phys.* **13** (2013) 287.
- [104] R. S. Kumat, D. Sivakumar and A. S. Gandhi // *Scr. Mater.* **66** (2012) 451.
- [105] M. F. Zawrah, A. A. El-Kheshen and A. A. El-Maghraby // *Ceram. Int.* **38** (2012) 3857.
- [106] Z. X. Shen, M. Ge, M. W. Chen and W. G. Zhang // *Ceram. Int.* **38** (2012) 6053.
- [107] Y. Lu, Y. Wang, Z. Pan, H. Shen and L. Wu // *Ceram. Int.* **39** (2013) 4421.
- [108] J. Hu, S. Dong, X. Zhang, H. Zhou, B. Wu, Z. Wang, P. He and L. Gao // *Composites Part A: Appl. Sci. Manuf.* **48** (2013) 73.
- [109] S. Y. Kim, I. S. Han, S. K. Woo, K. S. Lee and D. K. Kim // *Mater. Des.* **44** (2013) 107.
- [110] A. Thuault, S. Marinel, E. Savary, R. Heuguet, S. Saunier, D. Goeriot and D. Agrawal // *Ceram. Int.* **39** (2013) 1215.
- [111] D. Jiang, J. Zhang and L. Zhihui // *J. Eur. Ceram. Soc.* **32** (2012) 1419.
- [112] D. Galusek, R. Klement, J. Sedlacek, M. Balog, C. Fasel, J. Zhang, M. A. Crimp and R. Riedel // *J. Eur. Ceram. Soc.* **31** (2011) 111.
- [113] J. Cabrero, F. Audubert and R. Pailler // *J. Eur. Ceram. Soc.* **31** (2011) 313.
- [114] S. Kumar, A. Kumar, K. Sampath, V. V. B. Prasad, J. C. Chaudhary, A. K. Gupta and G. R. Devi // *J. Eur. Ceram. Soc.* **31** (2011) 2425.
- [115] J. Mentz, M. Müller, M. Kuntz, G. Grathwohl, H. P. Buchkremera and D. Stöver // *J. Eur. Ceram. Soc.* **26** (2006) 1715.
- [116] M. Patel, V. V. B. Prasad and V. Jayaram // *J. Eur. Ceram. Soc.* **33** (2013) 1615.
- [117] A. Udayakumar, A. Sri Ganesh, S. Raja and M. Balasubramanian // *J. Eur. Ceram. Soc.* **31** (2011) 1145.
- [118] W. Jeremy, G. Hilmas, W. G. Fahrenholtz, D. Brown and B. Clausen // *J. Eur. Ceram. Soc.* **31** (2011) 1811.
- [119] G. Magnani, L. Beaulardi, A. Brentari, T. Toyoda and K. Takahashi // *J. Eur. Ceram. Soc.* **30** (2010) 769.
- [120] L. H. He, F. H. Li, X. P. Lu, Y. W. Bao and Y. C. Zhou // *J. Eur. Ceram. Soc.* **30** (2010) 2147.
- [121] J. Watts, G. Hilmas, W. G. Fahrenholtz, D. Brown and B. Clausen // *J. Eur. Ceram. Soc.* **30** (2010) 2165.
- [122] F. Monteverde, R. Savino, M. S. Fumo and A. Maso // *J. Eur. Ceram. Soc.* **30** (2010) 2313.
- [123] J. Wang, M. Lin, Z. Xu, Y. Zhang, Z. Shi, J. Qian, G. Qiao and Z. Jin // *J. Eur. Ceram. Soc.* **29** (2009) [14] 3091–3097.
- [124] D. Alfanoa, L. Scatteia, S. Cantonia and M. Balat-Pichelin // *J. Eur. Ceram. Soc.* **29** (2009) 2045.
- [125] G. Magnani, A. Brentari, E. Burresti and A. Coglitore // *Ceram. Int.* **39** (2013) 3345.
- [126] M. Patel, J. J. Reddy, V. V.B. Prasad and V. Jayaram // *J. Eur. Ceram. Soc.* **32** (2012) 4455.
- [127] M. S. Roy // *Carbon* **43** (2005) 275.
- [128] P. Hu, W. Guolin and Z. Wang // *Corros. Sci.* **51** (2009) 2724.
- [129] J. Han, P. Hu, X. Zhang, S. Meng and W. Han // *Compos. Sci. Technol.* **68** (2008) 799.
- [130] P. Lespade, N. Richet and P. Goursat // *Acta Astronaut.* **60** (2007) 858–864.
- [131] F. Monteverde and A. Bellosi // *J. Eur. Ceram. Soc.* **25** (2005) 1025.
- [132] A. Kovalčíková, J. Dusza, P. Šajgalík // *Ceram. Int.* **39** (2013) 7951.
- [133] A. Maity, D. Kalita, N. Kayal, T. Goswami, O. Chakrabarty and P. G. Rao // *Ceram. Int.* **38** (2012) 4701.
- [134] Z. Yue, G. Dong, X. Chunlai, S. Yang and S. Xiaobin // *Int. J. Appl. Ceram. Technol.* DOI: 10.1111/ijac.12000.
- [135] S. Guo, T. Mizuguchi, M. Ikegami and Y. Kagawa // *Ceram. Int.* **37** (2011) 585.
- [136] O. N. Grigor'ev // *Refract. Ceram. Mat.* **51** (2012) 697.
- [137] M. F. Gazulla, M. P. Gomez, M. Orduna and A. Barba // *J. Eur. Ceram. Soc.* **26** (2006) 3451.
- [138] N. I. Voronin, N. I. Krasotkina and V. A. Smirnova // *Refract.* **1** (1960) 265.

- [139] N. I. Voronin, N. I. Krasotkina, A. P. Stavorko and R. S. Milshenko // *Refract.* **2** (1961) 124.
- [140] N. I. Voronin and N. I. Krasotkina // *Refract.* **2** (1961) 372.
- [141] N. I. Voronin and N. I. Krasotkina // *Refract.* **3** (1962) 352.
- [142] I. Y. Guzman, M. S. Fraifeld and A. K. Karklit // *Refract.* **16** (1975) 429.
- [143] M. Berdichevskii, N. I. Teleshman and O. M. Kresin // *Glass. Ceram.* **41** (1984) 27.
- [144] A. V. Dunikov, G. D. Semchenko and Y. G. Gogotsi // *Refract.* **25** (1984) 71.
- [145] R. J. Julietti and B. C. Reeve // *Br. Ceram. Trans. J.* **90** (1991) 85.
- [146] T. A. Boiko, A. B. Ivanov and Z. V. Tret'yakova // *Refract. Indus. Ceram.* **38** (1997) 21.
- [147] X. Zhong and H. Zhao // *Am. Ceram. Soc. Bull.* **78** (1999) 98.
- [148] D. Shaoming, Y. Katoh and A. Kohyama // *J. Am. Ceram. Soc.* **86** (2003) 26.
- [149] A. A. Nourbakhsh, F. Golestani-Fard, H. R. Rezaie // *J. Eu. Ceram. Soc.* **26** (2006) 1737.
- [150] D. M. Borzov // *Ogneupory Tekhn. Keram.* **4** (2002) 30 – 34.
- [151] G. D. Semchenko, N. Y. Kobets, S. P. Savina, K. A. Lysak and V. V. Davyskiba // *Refract. Indus. Ceram.* **53** (2012) 46.
- [152] V. P. Sivov // *Glass. Ceram.* **58** (2001) 322.
- [153] M. K. Ferber and V. J. Tennery // *Ceram. Bull.* **62** (1983) 236.
- [154] T. M. Strobel and J. P. Hurley // *Fuel Process Technol.* **44** (1995) 201.
- [155] N. K. Reddy // *Mat. Lett.* **47** (2001) 305.
- [156] I. Y. Guzman, V. S. Pankratova and O. V. Aleksandrovskaia // *Refract.* **11** (1970) 449.
- [157] N. V. Pitak, R. M. Fedoruk, T. P. Khmelenko and V. S. Baksheeva // *Refract.* **29** (1988) 69.
- [158] J. Chin and T. Ohkawa // *Nucl. Technol.* **32** (1977) 115.
- [159] Y. Hirohata, M. Kobayasi, S. Maeda, K. Nakamura, M. Mohri, D. Watanabe and T. Yamashina // *Thin Solid Films* **63** (1979) 237.
- [160] W. V. Munch and P. Hoeh // *Solid-State Electron.* **21** (1978) 479.
- [161] S. Nishino, A. Ibaraki, H. Matsunami and T. Tanaka // *Jpn. J. Appl. Phys.* **19** (1980) L353.
- [162] D. J. Godfrey // *Metals Mater.* **2** (1968) 305.
- [163] A. Benfdila and K. Zekentes // *Afr. Phys. Rev.* **4** (2010) 25.
- [164] C. Raynaud // *J. Non-Cryst. Solids* **280** (2001) 1.
- [165] X. Chen, L. Ning, Y. Wang, J. Li, X. Xu, and X. Hu // *J. Mater. Sci. Technol.* **25** (2009) 115.
- [166] C. D. Fung and J. J. Kopanski // *Appl. Phys. Lett.* **45** (1984) 757.
- [167] K. Shihahara, S. Nishino and H. Matsunami // *Jpn. J. Appl. Phys.* **23** (1984) L862.
- [168] A. Suzuki, H. Ashida, N. Furui, K. Mameno and H. Matsunami // *Jpn. J. Appl. Phys.* **21** (1982) 579.
- [169] F. F. Lange // *J. Am. Ceram. Soc.* **53** (1970) 290.