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ORIGINAL RESEARCH

Observations on the infrared reflectance spectra of Neutron-Irradiated 3C-Sic

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Recent analyses of neutron-irradiated cubic silicon carbide (3C-SiC) samples using Fourier-transform infrared spectroscopy (FTIR spectroscopy) reported on observed changes of the reflectance as well as extracted dielectric parameters of the samples. The changes were thought to be associated with sample surface roughness. Since neutron irradiation is not expected to damage the surface of irradiated samples, these observations were reassessed. It is suggested that changes could more likely be ascribed to the grain sizes of the polycrystalline 3C-SiC samples that were irradiated.

Keywords: 3C-SiC, neutron irradiation, infrared spectroscopy

Waarnemings oor die infrarooi reflektansiespektra van Neutronbestraalde 3C-SiC: Onlangse ontledings van neutronbestraalde kubiese silikonkarbied monsters (3C-SiC-monsters) met behulp van Fourier-transform-infrarooispektroskopie (FTIR-infrarooispektroskopie) het gerapporteer oor waargenome veranderinge van die reflektansie sowel as onttrekte diëlektriese parameters van die monsters. Die veranderinge hou vermoedelik verband met die oppervlakgrofheid van die monsters. Aangesien daar nie verwag word dat neutronbestraling die oppervlak van bestraalde monsters sal beskadig nie, is hierdie waarnemings herevalueer. Daar word voorgestel dat veranderinge waarskynliker toegeskryf kan word aan die korrelgroottes van die polikristallyne 3C-SiC-monsters wat bestraal is.

Sleutelwoorde: 3C-SiC, neutronbestraling, infrarooispektroskopie

Introduction

The use of silicon carbide (SiC) as a containment layer in the triple-coated isotropic layers in the new generation nuclear reactors has already been proposed in 1989 by Wacholz (Wacholz, 1989). During the operation of these nuclear reactors, the SiC is expected to be subjected to high fluences of various nuclear fission particles, as well as to varying temperatures. Consequently several investigations into the effect of irradiation on SiC have already been reported, using various characterisation techniques to obtain information on any influence of the irradiation on material properties (Snead et al., 1998a, 1998b, 1999; Dkaki et al., 2001; Ryazanov et al., 2002; Katoh et al., 2006; Newsome et al., 2007; Kondo et al., 2008a, 2008b; Katoh et al., 2008, 2011; Brink et al., 2009; Talwar et al., 2012; Dong et al., 2012; Malherbe, 2013; O'Connell & Neethling, 2014; Lillo & Van Rooyen, 2016).

Previous investigations reported on 3C-SiC wafers irradiated at (i) varying irradiation temperatures but more or less constant neutron radiation fluences (Engelbrecht et al., 2014) and (ii) at constant irradiation temperature but varying neutron radiation fluences (Engelbrecht et al., 2015). Fourier-transform infrared (FTIR) spectroscopy and atomic force microscopy (ATM) were used to characterise the irradiated samples. Changes were observed in the infrared reflectance as well as in the extracted dielectric parameters of the samples. The changes were ascribed to sample surface roughness. However, the latter proposal required further investigation, as damage to the surface of samples subjected to neutron radiation was not expected (O'Connell and Neethling, 2014).

This article reports on the reassessment of the same previously analysed neutron-irradiated 3C-SiC samples.

Experimental

Two sets of SiC samples in the shape of discs were analysed at:

a. A constant neutron irradiation temperature of 800 °C, while the neutron fluences were varied between 5 x 10^{19} and 7,7 x 10^{21} n/cm^2; and
b. Irradiation temperatures of 200 °C, 300 °C and 400 °C, and at neutron fluences ranging from 5 x10^{19} to 4,8 x 10^{20} n/cm^2.

Irradiation of samples was done at the Oak Ridge National Laboratory (ORNL), USA. Information related to specimens analysed in this report is contained in Tables I and II.

A Bruker 80V FTIR/Raman spectrometer, fitted with a Pike 10 Spec specular reflection unit enabling near-normal incidence, was employed to obtain infrared reflectance spectra from samples taking 50 scans at a resolution of 8 cm^{-1}. Surface roughness of the samples was measured using a CSM Instruments Nano-indenter, fitted with an atomic force microscope.

Sample areas of size 25 x 25 μm² were analysed by atomic force microscopy (AFM). Samples were subsequently polished with diamond suspension, followed by successively less coarse diamond paste and finally with 50 nm colloidal silica. The IR reflectance of all samples were then re-measured. Polished samples were then inspected using a JEOL JSM 7001F scanning electron microscope (SEM), while the size of grains in the samples were determined by electron backscattered diffraction (ESBD) using AZtec software. The average grain sizes were determined from three SEM micrographs obtained for each sample.

Results and discussion

Constant temperature, varying neutron fluence

Figure 1 depicts the change in infrared reflectance of two samples before and after polishing. In one case (No. 15) a decrease in reflectance is observed, while in another case (No. 16) the reflectance increased after polishing. These observations, in particular that for sample 15, is already an indication that the previous interpretation of the reason for changes in reflectance (Engelbrecht et al., 2014; Engelbrecht et al., 2015) might not be correct. Kroon showed that surface roughness has a significant effect on reflectance, and the reflectance can also increase with surface roughness, depending on the associated change in the dielectric constant ε of the semiconducting material (Kroon, 2007). This is clearly displayed in Figure 1 for sample 15.

The sample surface root mean square (RMS) roughness, as measured by AFM, is shown in Figure 2 for all samples irradiated at 800 °C. Clearly, the surface roughness is approximately equal for all samples after polishing, since the RMS roughness decreased from the μm to nm range. Values are contained in Table I.

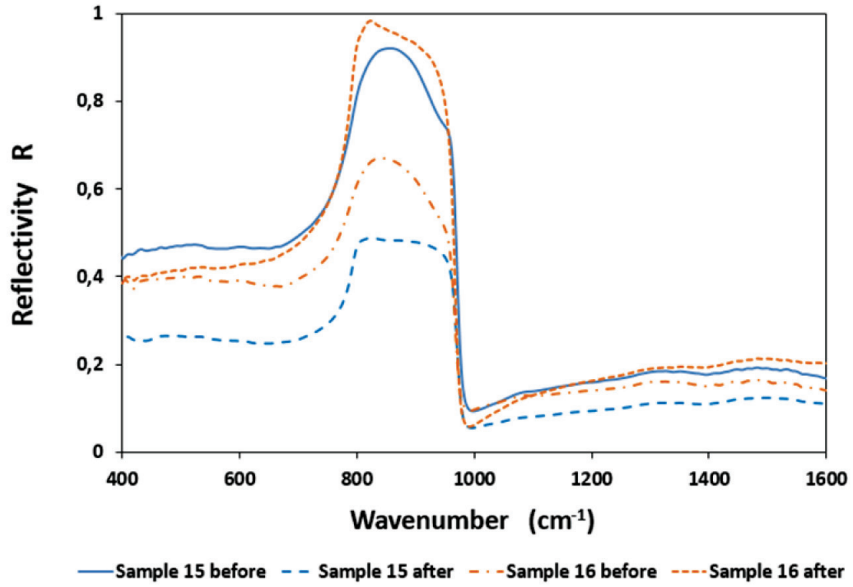

Figure 1: Infrared reflectance spectra of two samples irradiated at 800 °C, before and after polishing

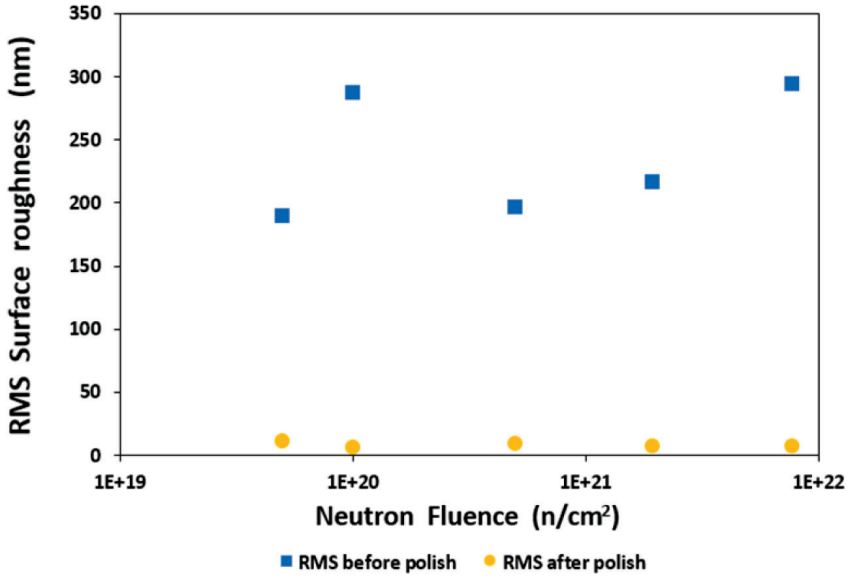

Figure 2: RMS surface roughness of all samples irradiated at 800 °C, before and after polishing

Table I: Sample properties before and after polishing for samples irradiated at constant temperature but varying neutron irradiation fluence

Sample number	Irradiation temperature (°C)	Fluence (n/cm²)	Mean RMS surface roughness before polish (nm)	Mean RMS surface roughness after polish (nm)	Average grain size after polish (µm)	Rmax
15	800	$5{,}0 \times 10^{19}$	189	11,1	8,6 ± 0,1	0,97
16	800	$1{,}0 \times 10^{20}$	287	5,8	3,8 ± 0,1	0,77
18	800	$5{,}0 \times 10^{20}$	196	9,2	3,7 ± 0,1	0,50
20	800	$1{,}9 \times 10^{21}$	216	6,7	8,8 ± 0,1	0,75
19	800	$7{,}7 \times 10^{21}$	294	6,6	3,7 ± 0,2	0,67

The mean RMS surface roughness was measured using AFM, and results for samples 15 and 16 (from Figure 1) are shown in Figure 3. The average grain sizes observed for samples 15 and 16 are presented in Figure 4. Values obtained for average grain size and mean surface roughness are contained in Table II.

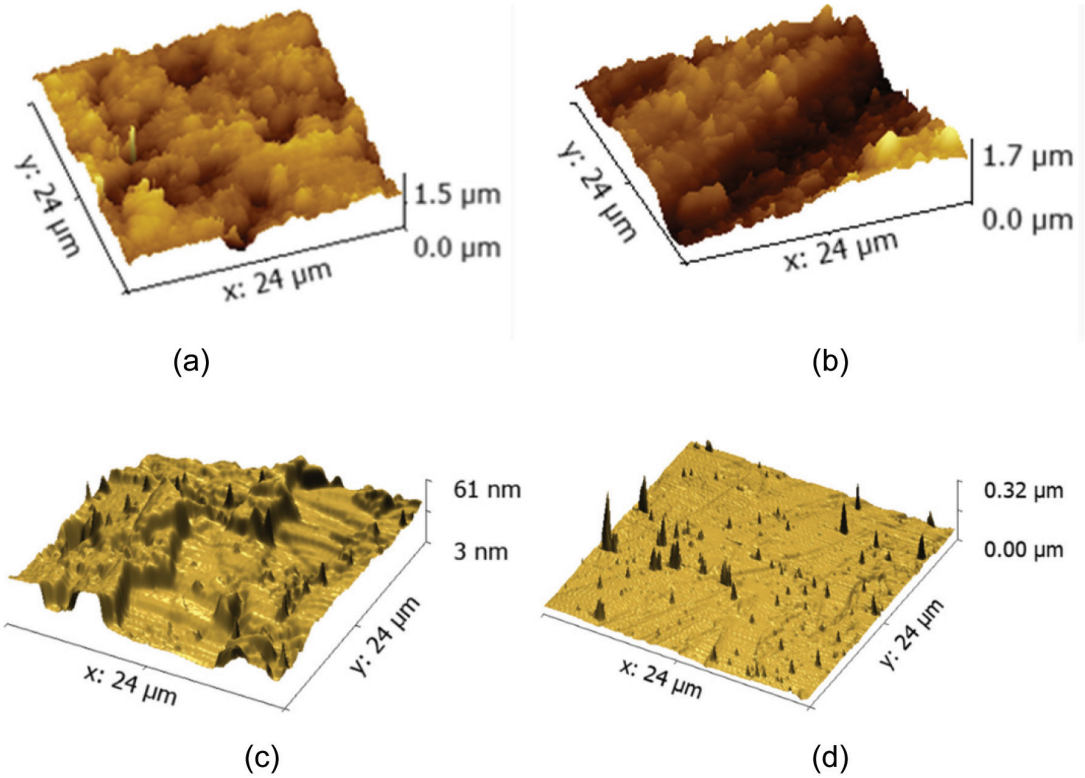

Figure 3: AFM measurements of the mean RMS surface roughness of sample 15 (a) and sample 16 (b) before polishing, and sample 15 (c) and sample 16 (d) after polishing

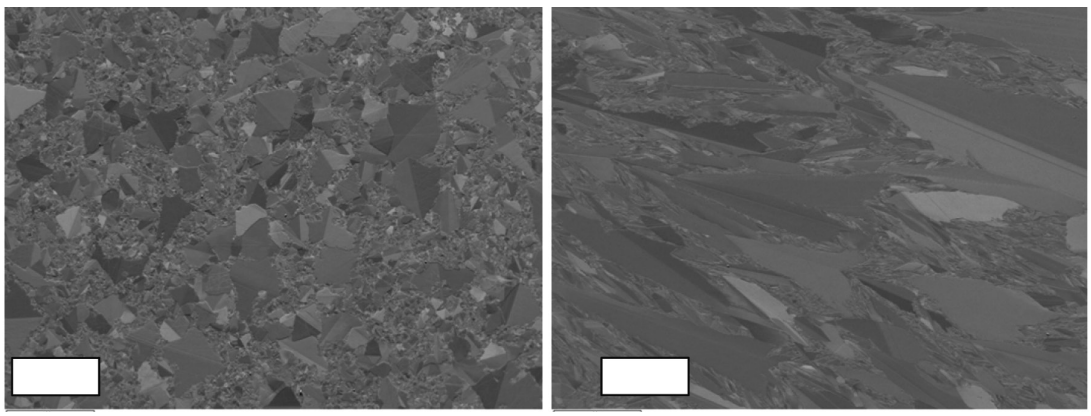

Figure 4: SEM micrographs of the grains in (a) sample 15 and (b) sample 16 (after polishing) (the white bar represents 100 micron)

Comparing the maximum infrared reflectance R, mean RMS surface roughness and the average grain size for all the samples in this set (Figure 4), it is clear that (i) there is no relation between the reflectance and the surface roughness after polishing the samples, and (ii) there is a definite relation between the reflectance and the average grain size. (Errors in reflectance are ± 10%).

The above observation was subsequently verified by repeating the investigation for samples irradiated at roughly the same neutron fluences, but at varying temperatures.

Same neutron fluence, different irradiation temperatures

Repeating the analyses as in 3.1, the results for the mean surface roughness before and after polishing, and average grain size after polishing were obtained as presented in Table II.

The maximum infrared reflectance for each of the samples, together with the mean surface roughness and average grain size, are shown in Figure 5 (low fluence range) and Figure 6 (high fluence range) – refer to Table I for neutron fluences.

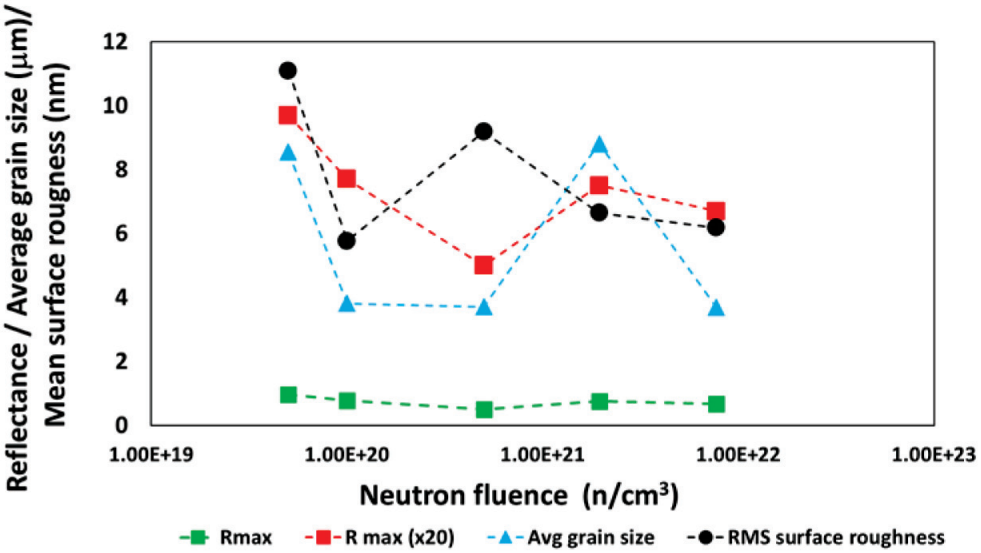

Figure 5: Maximum reflectance, RMS surface roughness and average grain size for all samples that were irradiated at 800 °C (note that the dotted lines are only a guide for the eye)

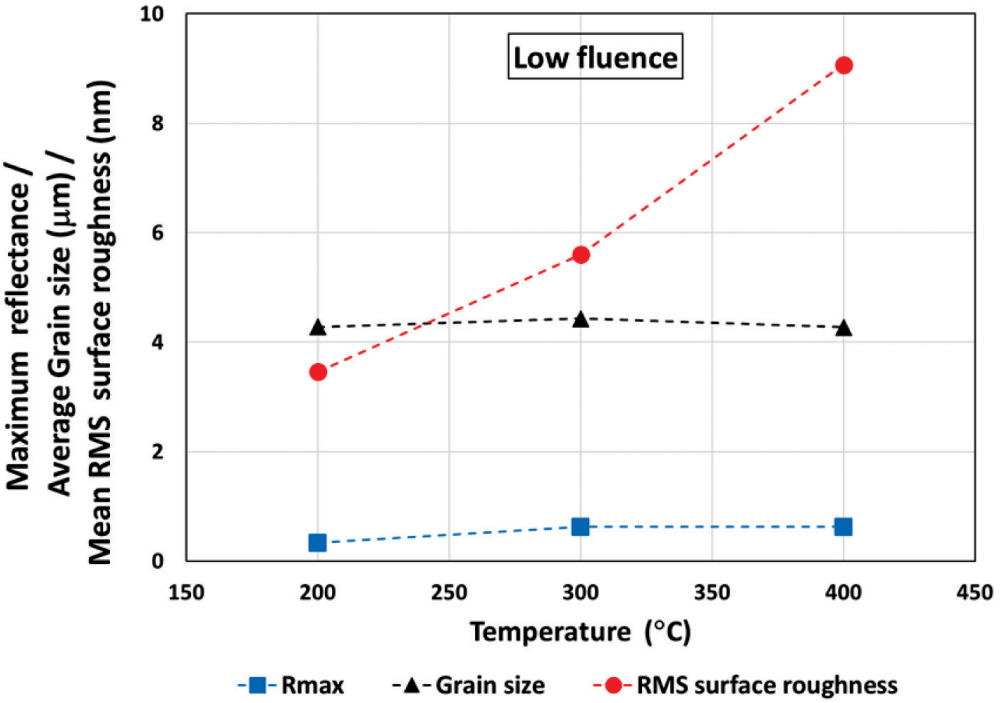

Figure 6: Reflectance, mean RMS surface roughness and average grain size for low neutron fluence samples irradiated at temperatures as indicated on the x-axis (dotted lines are only a guide for the eye)

Table II: Properties of neutron irradiated samples at varying temperatures as indicated

Sample number	Irradiation temperature (°C)	Fluence (n/cm²)	Mean RMS surface roughness before polish (nm)	Mean RMS surface roughness after polish (nm)	Average grain size after polish (μm)	Rmax Before polish	Rmax After polish
2	200	$5{,}6 \times 10^{19}$	110	3,5	4,3 ± 0,03	0,97	0,34
3	200	$9{,}7 \times 10^{19}$	260	4,7	3,2 ± 0,03	0,41	0,37
4	300	$6{,}0 \times 10^{20}$	100	5,6	4,4 ± 0,03	0,75	0,63
5	300	$1{,}4 \times 10^{21}$	190	4,2	2,2 ± 0,03	0,55	0,48
9	400	$7{,}7 \times 10^{21}$	160	9,1	4,3 ± 0,03	0,72	0,63
10	400	$4{,}8 \times 10^{20}$	169	5,3	5,7 ± 0,08	0,63	0,33

It is clear from the trends observed for the variation in reflectance as function of mean surface roughness and average grain size that these variations are due to the grain size and not the surface roughness, as reported earlier (Engelbrecht et al., 2014; Engelbrecht et al., 2015). It is likely that the trends reported earlier were in part due to the surface damage caused when the 3C-SiC rods were cut before irradiation. However, there could also have been a contribution from the grain sizes during IR measurements. The penetration depth of infrared radiation in silicon carbide is given as follows (Amirtharaj, P.M. & Seiler, D.M., 1995):

$$d = \lambda/(2\pi k) \quad (1)$$

where λ is the wavelength and k is the extinction coefficient of 3C-SiC, with $k = k(\lambda)$ (Adachi, S., 2012).

It is clear from Figure 8, as calculated and plotted from equation (1), that the penetration depth for some wavelengths in the Reststrahlen region where the maximum reflectance R was measured is > 30 μm. These values are greater than the surface roughness measured, and so it is likely that there would have been a contribution from the grains in the 3C-SiC samples when measuring the reflectance.

In conclusion, it should be noted that both surface roughness and grain size could play a role in the reflectance of the samples. However, since it has been reported that neutron irradiation does not affect the surface roughness of irradiated 3C-SiC samples, it is most likely that the changes in reflectance observed in the present study are due to the grain size of the respective samples. This finding thus contradicts the conclusion of the earlier publications regarding the surface roughness of the samples.

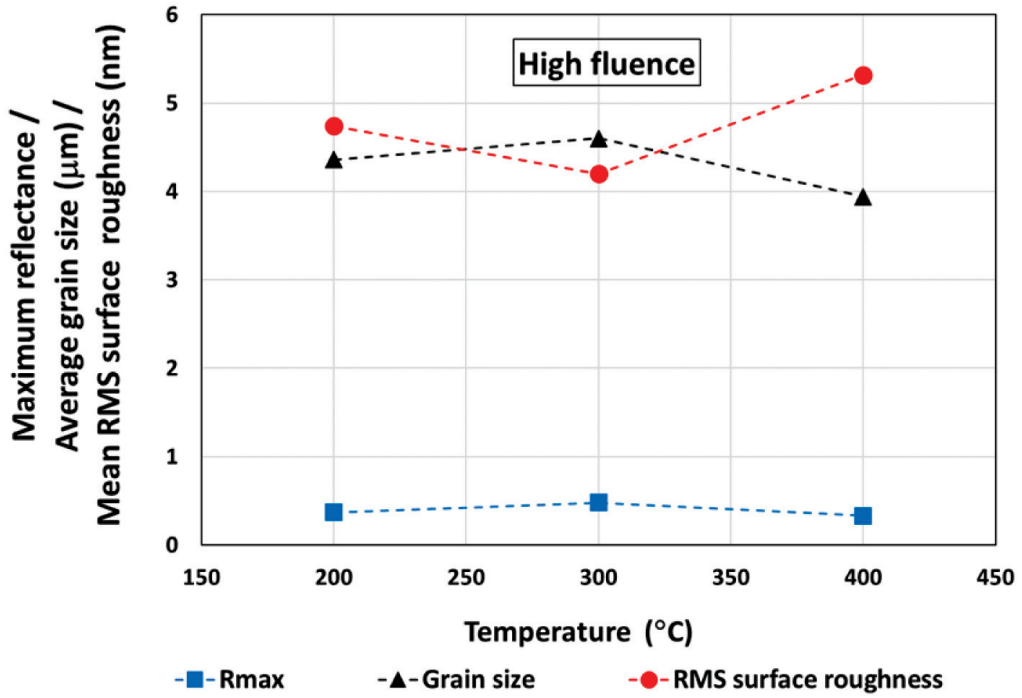

Figure 7: Reflectance, mean RMS surface roughness and average grain size for high neutron fluence samples irradiated at temperatures as indicated on the x-axis (dotted lines are only a guide for the eye)

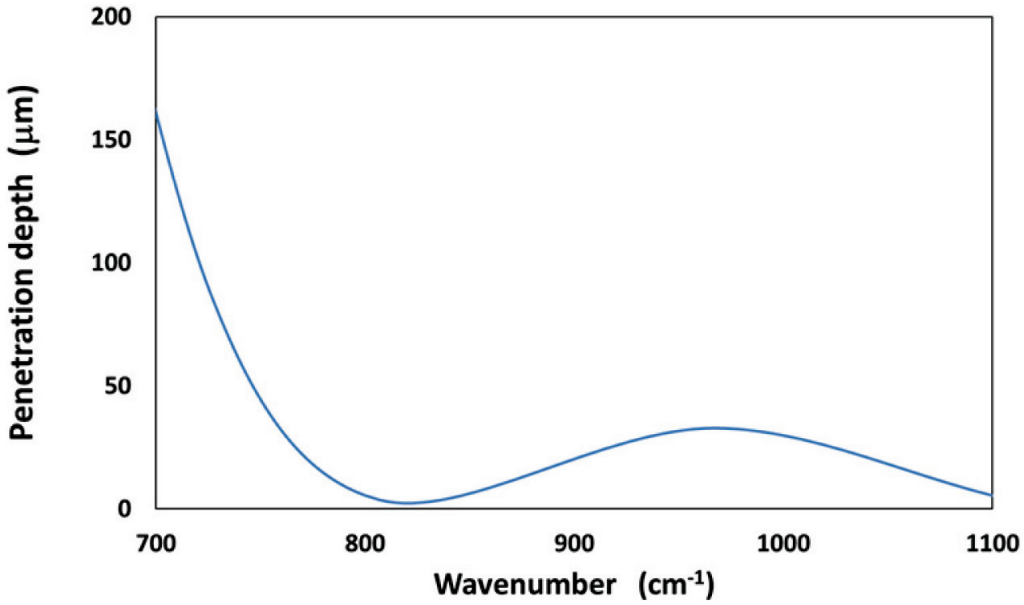

Figure 8: Penetration depth of infrared radiation in the Reststrahlen region of 3C-SiC

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