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X-ray Investigation of SiC Nanostructure on Cu Films

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This article presents the X-ray investigation results of the silicon carbide nanostructures which were synthesized by the method of Microwave Enhanced Chemical Vapor Deposition (MWCVD). The copper films deposited by the method of magnetron sputtering were used as catalysts. The experiments were performed at different temperatures from 600 to 900 °C (in 100 °C increments). SEM studies of the samples showed that the nanostructures have a diameter of 180-400 nm and a length from 670 nm to 1.5 microns. It is also seen from SEM studies that synthesized nanostructures have a rough surface. Analysis of the results of experiments on the synthesis of silicon carbide nanostructures using the MWCVD method shows that the growth of nanostructures starts from 700 °C. It should be emphasized that the intensity of the SiC peak increases with increasing synthesis temperature. Moreover, with increasing synthesis temperature it can be noticed that the next reflection peaks characteristic of silicon carbide appear. The results of X-ray analysis showed that samples grown at a temperature of 900 °C have the highest crystallinity. The results of research using the methods of SEM and X-ray demonstrated that, in the course of experiments, in addition to silicon carbide nanostructures, various carbon formations were obtained. Thus, at a temperature of 600 °C, amorphous carbon films are formed, while at 700 °C nanostructures of silicon carbide are formed.

Keywords: Silicon carbide, Chemical vapor deposition, Microwave plasma, Method of scanning Electron microscopy, X-ray analysis.

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1. INTRODUCTION

SiC with unique properties, such as wide band gap, excellent thermal conductivity, chemical inertness, high electron mobility and biocompatibility, are promising for applications in microelectronics and optoelectronics, as well as nanocomposites. Silicon carbide has many polytypes, among which the most studied are 3C-SiC, 4H-SiC and 6H-SiC, which have different physicochemical properties [1, 2].

Due to the high number of surface atoms in silicon carbide nanostructures, its properties are different from the bulk material, and it has unique physical, chemical and mechanical properties [3, 4].

For the synthesis of silicon carbide nanostructures, various methods were used, such as sol-gel [5], CVD [6], carbothermic reduction process [7], and others. Synthesis of cubic ß-SiC nanostructure by MWCVD was proposed in [8].

The aim of the study was to experimentally reveal the influence of technological parameters (synthesis temperature, operating pressure, plasma power, etc.) on formation of silicon carbide nanostructures.

2. EXPERIMENTAL

The joint studies on the synthesis of silicon carbide nanostructures by the MWCVD method were carried out at the Institute of Materials Science of the University of Siegen (Germany).

2.1 Substrate Preparation

The monocrystalline KDB-20 silicon wafers (Siegert Wafer GmbH, Germany) of 1×1 cm^2 with [100] and [111] orientations were used as substrates and basis for copper films. The substrates were chemically pre-cleaned. The treatment was carried out in a mixed solution of NH_4OH, H_2O_2 and distilled water at a volume ratio of 1:1:6.5 at a temperature of 20 °C for 10 minutes using sound waves with a frequency of 850 kHz and power of 250 Watt. Further, washout in distilled water and drying were performed. Copper films were deposited on substrates of polished silicon wafers by DC magnetron sputtering in VUP-5M. Sputtering was carried out in the flow of working gas Ar at a pressure of 10^{-2} Torr. The flow rate of Ar was 6 cm^3/min and it was controlled by the gas flow controller MCV-500SCCM. Experiments were conducted at a constant voltage on the anode target (740 V) and plasma current of 35 mA. The time of experiments was 5 minutes.

2.2 Synthesis of Silicon Carbide Nanostructures

Synthesis of silicon carbide nanostructures was carried out in the Institute of materials science, University of Siegen (Germany) at the installation of MWCVD of the ASTEX system (frequency 2.45 GHz). Before the experiments, the substrates were cleaned with ethanol and then washed with distilled water, drying was carried out at room temperature. The synthesis temperature varied from 600 °C to 900 °C with a step of 100 °C.

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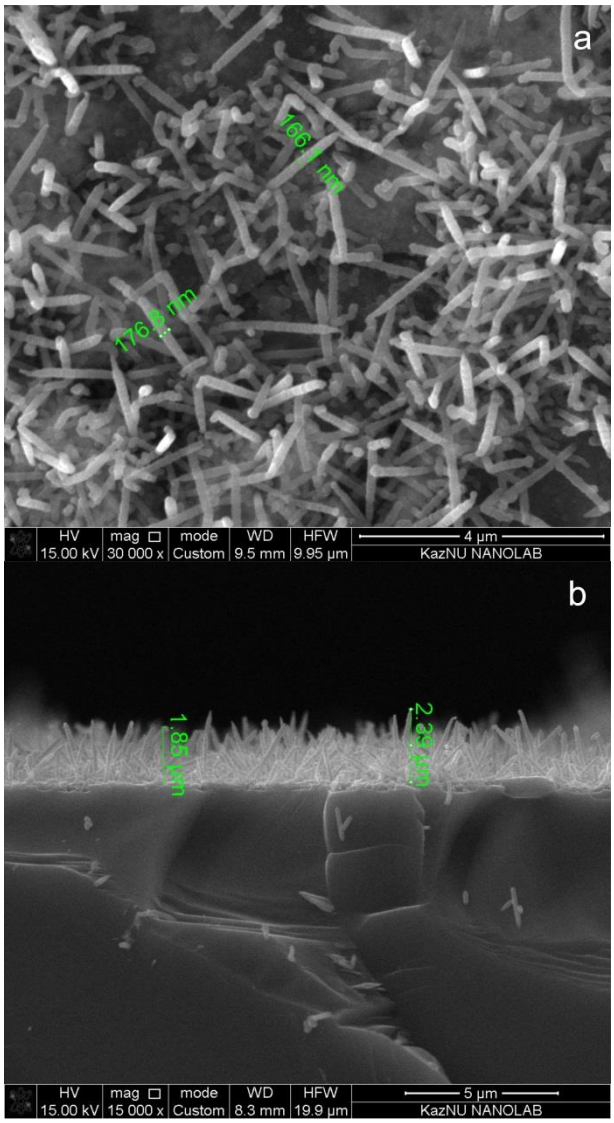


Fig. 1 – SEM image of silicon carbide nanostructure: a) top view, b) cross-section view of the sample

The temperature was measured by Chino IR-AP M0011 infrared pyrometer (Japan). The experiments were carried out at three plasma powers of 1800, 2000 and 2200 W. The pressure in the chamber was changed depending on the plasma power. As a working gas, a mixture of trimethylsilane ($(CH_3)_4Si$) and hydrogen was used, the flow rate of which was 10 and 400 cm^3/min, respectively. The duration of the experiments is 120 min.

2.3 Methods and Apparatus for the Study of the Morphology and Structure of Silicon Carbide Nanostructures

The obtained samples were investigated by scanning electron microscopy (SEM) in the National nanotechnological laboratory of open type using a microscope Quanta 3D 200i. The study on the structure of silicon carbide nanostructures was carried out by the method of X-ray analysis using diffractometers Rigaku Mini Flex 600 XRD (X-ray analysis laboratory of Al-Farabi, KazNU) Radiographs of samples were obtained using copper radiation ($\lambda = 1.5406$ Å) in digital form. The voltage on the X-ray tube was 40 kV, the tube current was 15 mA, the goniometer motion step was equal to 0.02° and the shooting speed was 10° per second. PDXL2 software package with the base of diffractometric data PDF-2 was used for the phase analysis. Processing of X-ray spectra to determine the angular position and intensity of the reflection was performed in program OriginPro 8.1.

3. RESULTS AND DISCUSSION

Fig. 1 shows the top and side views of obtained SiC. It can be seen from this figure that nanostructure preferentially oriented grows to the substrate. The diameter of the nanostructure is about 180-400 nm, the length varies from 670 nm to 1.5 microns.

Fig. 2-Fig. 5 show the X-ray spectra of SiC nanostructures obtained at different temperatures. X-ray qualitative analysis of the phase composition of the sample indicates that Si, SiC and Cu are contained in the composition of the sample. The diffraction patterns of all samples present the reflection from the plane (111) ($2\theta \approx 43.1°$, Fm-3m {225}, PDF # 04-0836) which is the characteristic of copper. Also, the characteristic reflection from the substrate of monocrystalline silicon Si (111) appears at an angle of 28.4°. For monocrystalline silicon with orientation (100), the angle of the characteristic reflection from the plane (400) is 69.1° [Fd-3m {227}, PDF # 05-0565].

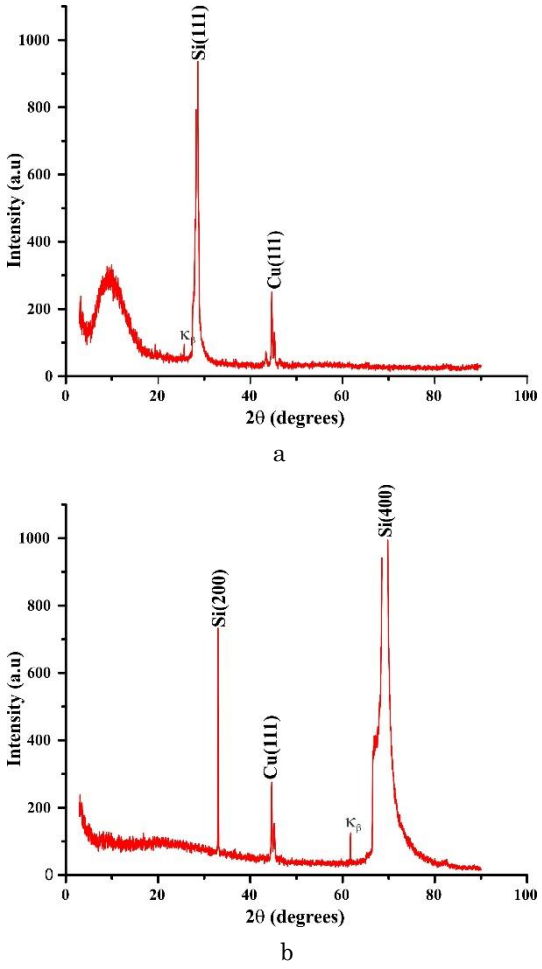
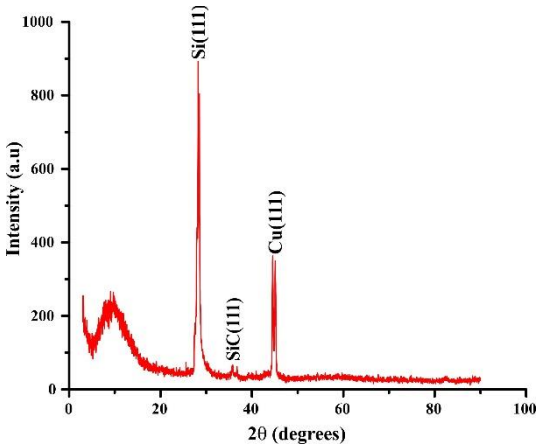
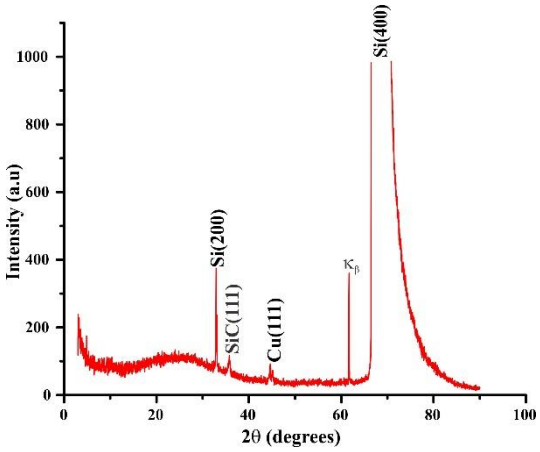


Fig. 2 – Diffraction pattern of the samples synthesized at a substrate temperature of 600 °C: a) Cu/Si (111), b) Cu/Si (100)

In Fig. 2, there is no reflection from silicon carbide, which indicates that the temperature of 600 °C is not sufficient for the synthesis of SiC.



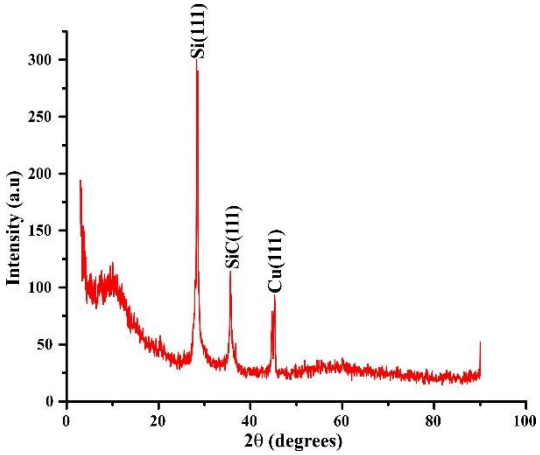
a



b

Fig. 3 – Diffraction pattern of the samples synthesized at a substrate temperature of 700 °C: a) Cu/Si (111), b) Cu/Si (100)

Fig. 3 shows the diffraction pattern of SiC synthesized at 700 °C. Reflections from β-SiC with low intensity appear at an angle of 35.7°, which indicates that the growth of silicon carbide begins with an increase in temperature, and the atomic mobility is low.



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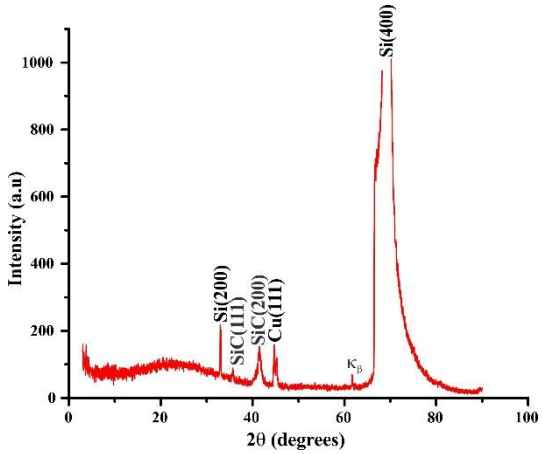
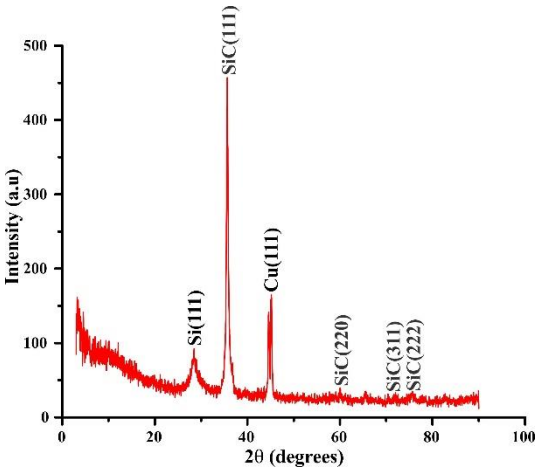


b

Fig. 4 – Diffraction pattern of samples synthesized at a substrate temperature of 800 °C: a) Cu/Si (111), b) Cu/Si (100)

The diffraction pattern in Fig. 4 shows that silicon carbide grows more intensively at a temperature of 800 °C. This is indicated by the intensity of the peak reflected from SiC (4a) and the number of reflexes (4b).



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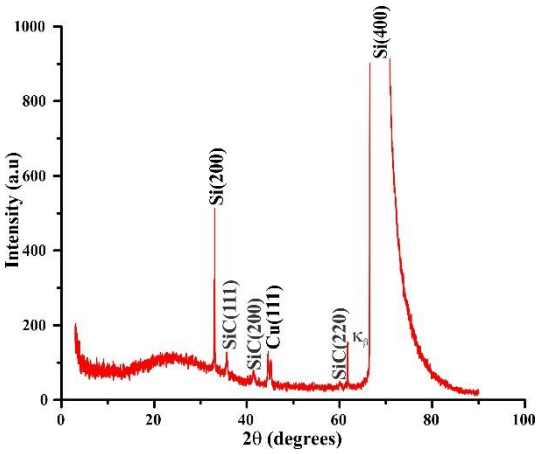

b

Fig. 5 – Diffraction pattern of samples synthesized at a substrate temperature of 900 °C: a) Cu/Si (111), b) Cu/Si (100)

On the X-ray pattern of the samples synthesized at a temperature of 900 °C there are reflections at angles of $2\theta \approx 35.7°$, 41.4°, 59.9°, 71.6° and 75.4° corresponding to β-SiC [F-43m {216}, PDF # 29-1129] from the (111) (200), (220), (311) and (222) planes. An additional peak at ~ 32.9° was observed on almost all Si (100) substrates. The interplanar spacing suggests that this peak appears due to the reflection from Si (200) plane. A peak (200) appears when there is excess defect formation. The peak at 61.7° corresponds to CuK_b radiation diffracted from Si (400) plane [9].

4. CONCLUSIONS

Analysis of the results of experiments on the synthesis of silicon carbide nanostructures using the MWCVD method shows that the nanostructure growth starts from 700 °C. Increase in temperature to 800 °C leads to increase in intensity and number of reflected peaks, hence, to increase in SiC yield. The highest intensity and number of peaks are present in the sample synthesized at 900 °C, which indicates the effective growth of nanostructures. This is confirmed by SEM images, which also show that the diameter of the nanostructure is ~ 180-400 nm and the length is ~ 670-1500 nm. The results show that temperature and SiC yield are proportional to each other, and the orientation of the substrate (111) is more preferable.

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Рентгенівське дослідження наноструктури SiC на плівках Cu

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У статті представлені результати рентгенографічного дослідження наноструктур карбіду кремнію, синтезованих методом мікрохвильового хімічного пароутворення (MWCVD). Мідні плівки, нанесені методом магнетронного розпилення, використовувалися як каталізатори. Експерименти проводили при різних температурах від 600 °C до 900 °C (з кроком 100 °C). SEM дослідження зразків показали, що наноструктури мають діаметр 180-400 нм і довжину від 670 нм до 1.5 мкм. З досліджень скануючою електронною мікроскопією видно, що синтезовані наноструктури мають шорстку поверхню. Аналіз результатів експериментів з синтезу наноструктур карбіду кремнію методом MWCVD показує, що зростання наноструктур починається від 700 °C. Слід підкреслити, що інтенсивність піку SiC зростає зі збільшенням температури синтезу. Крім того, при збільшенні температури синтезу можна помітити, що з'являються піки наступного відбиття, характерні для карбіду кремнію. Результати рентгенівського аналізу показали, що зразки, вирощені при температурі 900 °C, мають найвищу ступінь кристалічності. Результати досліджень з використанням методів скануючої електронної мікроскопії і рентгенівського аналізу показали, що в ході експериментів, крім наноструктур карбіду кремнію, були отримані різні вуглецеві форми. Таким чином, при температурі 600 °C утворюються аморфні вуглецеві плівки, а при 700 °C утворюються наноструктури карбіду кремнію.

Ключові слова: Карбід кремнію, Хімічне осадження парів, Мікрохвильова плазма, Метод скануючої електронної мікроскопії, Рентгенівський аналіз.