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Synthesis of biomorphic SiC ceramics

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This paper deals with a new method for producing non-oxide ceramic using wood as a template. Highly porous silicon carbide (SiC) ceramic with woodlike microstructure has been prepared by carbothermal reduction reactions of Tilia wood/TEOS composite at 1873K. Wood specimens were carbonized at 1273K in Ar atmosphere for 2 hour. The porous carbon preform was infiltrated with TEOS ($Si(OC_2H_5)_4$), as a source of silica, without pressure at 298K. The infiltration/annealing processs was repeated up to five times to increase the SiO_2 content in the biomorphic samples. The morphology of resulting porous SiC ceramics, as well as the conversion mechanism of wood to porous SiC ceramics, have been investigated by mean of scanning electron microscopy (SEM/EDS) and X-ray diffraction analysis (XRD). Experimental results show that the biomorphic cellular structure of wood is preserved after thermal treatment as a porous SiC ceramics. Obtained SiC ceramics consists of β-SiC with traces of α-SiC.

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1. Introduction

Manufacturing of ceramic materials from biological templates such as wood has become a matter of increasing interest because of the possibility of producing novel ceramic materials with a unique microstructure pseudomorphous to wood [1-5].

Woodceramics are promising new porous materials with large potentiality for a variety of industrial applications.

Anisotropy in wood is the result of the orientation and alignment of cells and cell walls, as well as variation in density. The highly anisotropic cellular structure of wood tissue may serve as a hierarchically structured template to generate novel cellular ceramics with micro-, meso-, and macro-structure. Related to the anatomical structure tree can be classified as a hardwoods and as a softwoods. Wood is natural composite with cellulose, hemicellulose, and lignin as the major constituents [6].

The morphology and the arrangement of the different cells may vary in a wide range between the different kinds of wood with large vessel cells dominanting in hardwood and tracheids dominanting in softwood. The diameter of the vessels and tracheids (named as pores) varies between 5 and 50 μm in softwood and between 1 and 300 μm in hardwood. These cells with a preferential orientation in axial direction offer the possibility to use various infiltration techniques to transform the bioorganic wood structure into an inorganic ceramic material with tailored physical and mechanical properties. High temperature (above 873K) pyrolyzing wood results in decomposition of the polyaromatic constituents to form carbon template with original cellular structure. These carbon performs can be used for producing SiC with highly anisotropic porosity, by liquid infiltration with tetraethylorthosilicate (TEOS) and subsequent carbothermal reduction reaction at elevated temperatures.

In the present work, we demonstrated the possibility of producing porous SiC ceramics with biomorphic microstructure by carbothermal reduction of premineralized Tilia wood with silica.

2. Experimental

Rectangular specimens (10x5x5mm) of Tilia wood were dried at 343K for 2 days. Carbon preform were prepared by carbonizing the dried wood at 1273K in Ar atmosphere for 2 hour with heating rate of 5 K/min. Specimens were soaked in ethanol solution of tetraethoxysilane [$Si(OC_2H_5)_4$, TEOS] (molar ratio TEOS:EtOH =1:4) and stirred for 2 hour at room temperature.

Distilled water (12 parts per one part of TEOS) and few drops of acetic acid were added to obtain the maximum gelling rate (pH=4). The silica sol contained in carbon perform was gelled at room temperature for 2 days and dried at 293K for 8 hour to remove other solvents. Samples are then again heat treated at 1273K (Ar) to obtain C/SiO_2 composites (CWS in further text). The treatment procedure of impregnation, gelling, drying and 1273K treatments were repeated several times, to increase the silica content in C/SiO_2 composites. Samples after first, second and third infiltrations will be denoted as CWSI, CWSII and CWSIII.

Carbothermal reduction reaction (CRR) of C/SiO_2 composite was carried out at 1873K in fluctuate Ar atmosphere in furnace with graphitic heating element for 1 hour with heating rate of 40 K/min to form porous SiC ceramic. The morphology changes and carbothermal conversion of the C/SiO_2 composite into SiC ceramic were investigated by scanning electron microscopy (SEM/EDS) and X-ray diffraction analysis (XRD).

3. Results and Discussion

Fig. 1. shows the XRD pattern of CWSIII that indicates semi-crystalline structure with characteristic (002) and (101) reflections of graphite. The XRD patterns of samples after carbothermal reduction (Fig. 2. depicts, that obtain SiC ceramics consists of β-SiC whit traces of α-SiC. The amount of β-SiC increases with increasing number of infiltration.

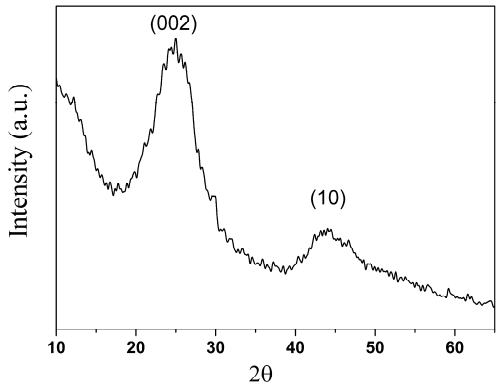


Fig. 1 – XRD pattern of C/SiO2 composite after 1273K treatment

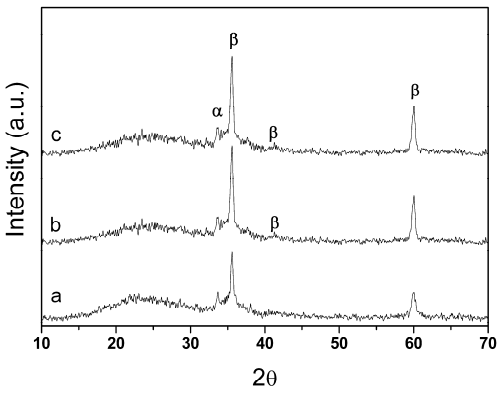
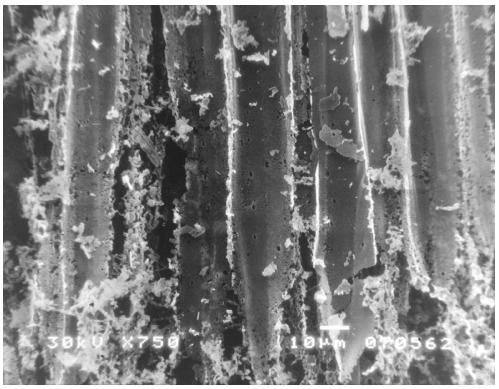
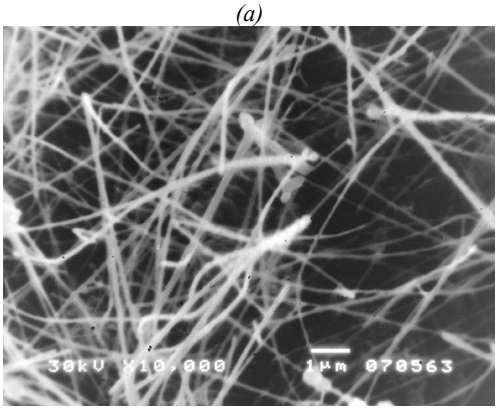


Fig. 2 – XRD patterns of samples treated at 1873K: a) CWSI, b) CWSII, c) CWSIII

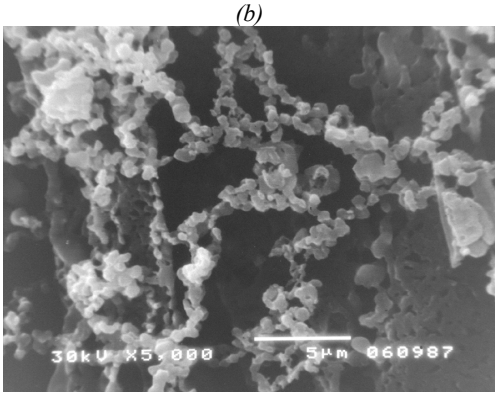
Fig. 3 shows SEM micrographs of highly porous woodlike silicon carbide ceramic obtained after CRR at 1873K and removal of free carbon at 973K (air, 2h). Preserved biomorphic cellular structure can be seen in Fig. 3a, showing channels that originate from tracheid cells that are parallel to the axis of the tree. Corresponding EDS spectra is presented in Fig. 4a, showing that this phase is mainly composed of Si and small amount of O, Ca and K. Unfortunately, carbon cannot be detected due to instrument limitations. At some points in the samples, usually at the end of the channels, fibers are observed (Fig. 3b). They are about 10μm in length with diameters varying in the range from 100 to 300nm. EDS spectra (Fig. 4b) is almost identical to previous spectra, showing only traces of O, Ca and K.



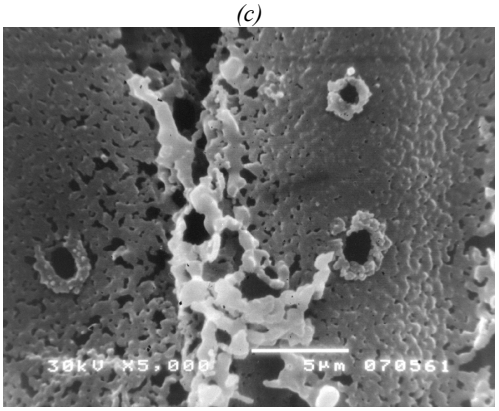
(a)



(b)



(c)



(d)

Fig. 3 SEM micrograph of woodlike SiC ceramics structures: a) cellular, b) fibrous, c-d) granular

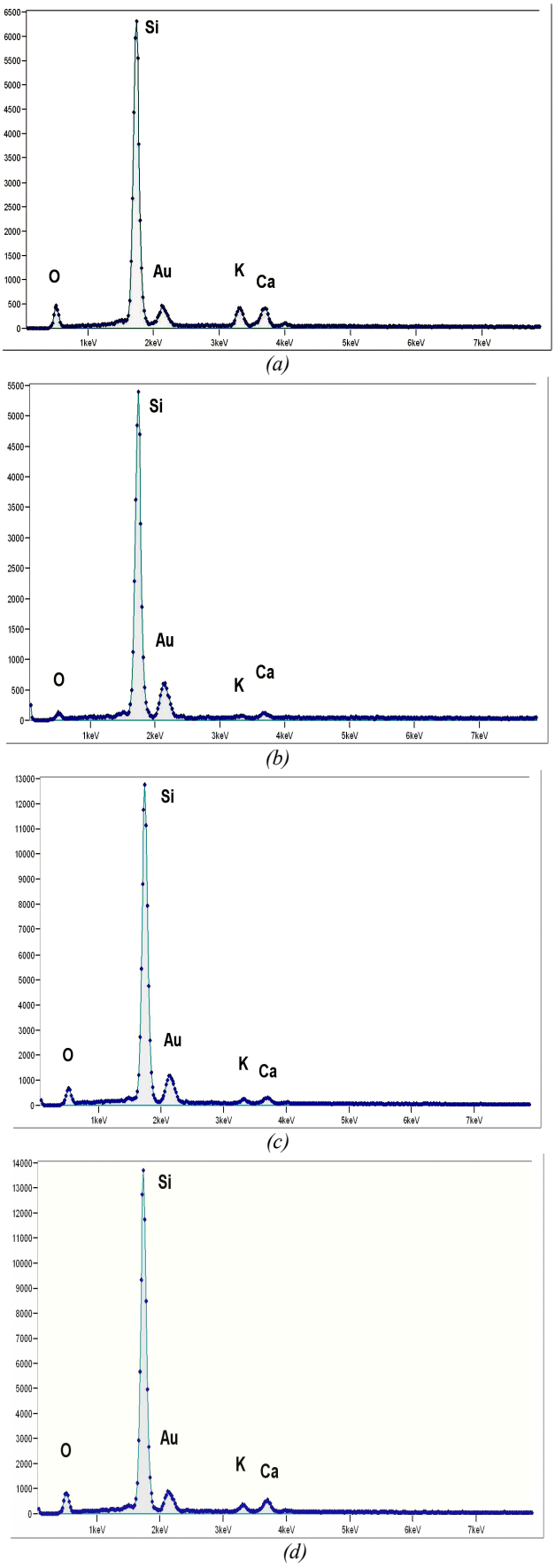


Fig. 4 EDS spectra of woodlike SiC ceramics structures: a) cellular, b) fibrous, c-d) granular

Granular porous structures are presented in Fig. 3c-d, showing chain-like structure (Fig. 3c) and pore structure i.e. open ends of channels, viewed in axial direction (Fig. 3d). EDS spectra (Fig. 4c-d) are also identical to previous ones, confirming homogeneity of sample, i.e. all observed phases have same chemical composition.

From both XRD and EDS results, it can be concluded that all observed phases are mainly composed of SiC. Ca and K are most probably present in form of CaO and K_2O, which are remains of corresponding water-soluble salts originally present in the raw wood material.

4. Conclusions

This paper demonstrated the conversion of biological cellular tissues structure into porous ceramics. This porous SiC ceramic with wood-like microstructure was prepared by sol infiltration of carbonized Tillia wood performs using TEOS as silica source and carbothermal reduction reaction at 1873K. XRD and EDS analysis reveals that β-SiC is the major phase present in the cellular ceramic product. This technique provides promising future applications for designing advanced materials with low density and high porosity.

Acknowledgments

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