SYNTHESIS OF SILICON NITRIDE NANOTUBES USING NANOSTRUCTURAL CARBON

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Introduction

As we showed in [1,2], in 80th of the last century in the course of synthesis of sialons from kaolin (carbothermal reduction in nitrogen-containing medium) the sort of products strongly depends on the gas phase composition. When the process is performed in ammonia, the by-product of synthesis are silicon nitride fibers.

As noted in [3-23], during the elementary silicon nitriding in the ammonia flow Si_3N_4 forms as fibers already at 1000 °C. The process proceeds through the intermediate stage where $Si_xN_yH_z$ forms. Then, losing hydrogen, this compound transforms into Si_3N_4.

In the work presented the hypothesis of using nanostructural carbon to synthesize Si_3N_4 nanotubes has been verified.

Experiment

All the modifications of carbon have been used as a reducing additive into kaolin. These modifications formed on the cathode as deposit resulted from the arc synthesis of fullerenes.

Kaolin and deposit material were mixed in the ball mill. Then samples were subjected to thermal treatment at 1000-1600 °C in the nitrogen-containing medium. The phase composition has been determined using XRD. Morphology of the product has been observed using a transmission electron microscope.

Results and discussion

The experiment performed has demonstrated that silicon nitride nanotubes begin to form at the temperature above 1100°C (Fig.1). The considerable number of fibers form at 1100-1400 °C when the processes of the carbothermal SiO_2 reduction and the product nitriding to Si_2ON and further to Si_3N_4 begin in the initial charge.

At temperatures above 1400 °C X-ray study on the cotton-like material clearly fixes three phases: α- and β-Si_3N_4 and Si_2ON. α and β phases present in approximately equal amount.

Considering the information above and results of morphological investigations, one can note the following. Silicion nitride nanotubes begin to grow at temperatures above 1200 °C (Fig.2,3). At the temperature above 1400 °C the clear X-ray lines of nitride phases appear in diffractograms of the fibrous product. The halo corresponding to the nanotubes is absent. This suggests that at this temperature nanotubes transform into the fibers and have the clear crystalline structure (Fig.4).

Using some technological methods, we have succeeded to produce the samples of nanotubes from pure α or β phases of Si_3N_4.

Fig.5 (a,b,c) shows three IR transmission spectra for β-Si_3N_4 taken on the one sample in depth of the cotton-like layer (The curve *a* - at the bottom, *b* - middle of fibres, *c* - upper part of the fibre). Analogous spectra have been taken for α-Si_3N_4 (the curve *d* - at the bottom of the fiber, *e* - surface).

Conclusions

When nanostructural carbon is used in the initial charge, the growth of fibers begins from the growth of nanotubes which gradually transform into fibers.

Carbon present in the deposit in the nanostructural state (and consequently in the active form), when mixed with kaolin, allows the decrease in the SiO_2 reduction temperature by ~ 200 °C. This results in the fact that the SiO_2 reduction temperature approaches the temperature for silicon nitride fiber formation on the elementary silicon.

The holding time, the temperature and the medium have a great effect on the formation of nanotubes and their transformation into the fibers.

References

1. Anikina NS, Schur DV, Simanovskiy AP, Zolotarenko AD, Dubovoy AG, Ivanchenko NV; Problem on fullerene production by electric arc method, Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Ukraine,590-591, 2001,
2. Matysina ZA, Schur DV; Hydrogen and solid phase transformations in metals, alloys and fullerites, Dnepropetrovsk: Nauka i obrazovanie, 420p (in Russian),2002,
3. Matysina ZA, Pogorelova OS, Zaginaichenko S Yu, Schur DV; The surface energy of crystalline CuZn and FeAl alloys,Journal of Physics and Chemistry of Solids,56,1,9-14, 1995,Elsevier
4. Schur DV, Lavrenko VA, Adejev VM, Kirjakova IE; Studies of the hydride formation mechanism in

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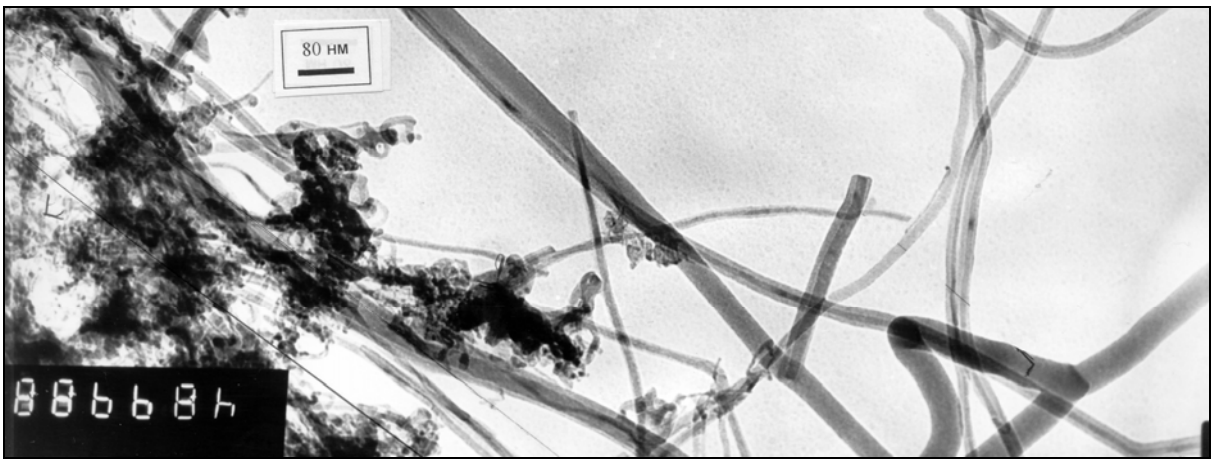
metals,International journal of hydrogen energy,19,3,265-268,1994,Elsevier
5. Schur DV, Dubovoi AG, Anikina NS, Zaginaichenko S Yu, Dobrovol'skij VD, Pishuk VK, Tarasov BP, Shul'ga Yu M, Meleshevich KA, Pomytkin AP; The production of utrafine powders of fullerites by the salting out method,Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Alushta-Cremia-Ukraine, September, 16-22, 2001,
6. Tarasov BP, Shul'ga Yu M, Fokin VN, Vasilets VN, Shul'ga N Yu, Schur DV, Yartys VA; Deuterofullerene C 60 D 24 studied by XRD, IR and XPS,Journal of alloys and compounds, 314,1,296-300,2001,Elsevier
7. Tarasov BP, Fokin VN, Moravsky AP, Shul'ga Yu M, Yartys VA, Schur DV; Promotion of fullerene hydride synthesis by intermetallic compounds, Hydrogen energy progress, 2, 1221-1230,1998,
8. Schur DV, Zaginaichenko S Yu, Matysina ZA, Smityukh I, Pishuk VK; Hydrogen in lanthan–nickel storage alloys,Journal of alloys and compounds,330,70-75,2002,Elsevier
9. Schur DV, Tarasov BP, Shul'ga Yu M, Zaginaichenko S Yu, Matysina ZA; Research of Fullerites Hydrogen Capacity,Hydrogen Materials Science and Chemistry of Metal Hydrides: Proceedings of the NATO Advanced Research Workshop on. Alushta Crimea, Ukraine, 16-22 September 2001,1,2002, Kluwer Academic Pub
10. Lavriv LV, Anikina NS, Simanovskij AP, Zolotarenko AD, Schur DV; Features of fullerene extraction wth toluene, Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Ukraine,596,2001
11. Schur DV, Zaginaichenko S Yu, Adejev VM, Voitovich VB, Lyashenko AA, Trefilov VI; Phase transformations in titanium hydrides, International journal of hydrogen energy, 21,11,1121-1124,1996, Pergamon
12. Schur DV, Tarasov BP, Zaginaichenko S Yu, Pishuk VK, Veziroglu TN, Shul'ga Yu M, Dubovoi AG, Anikina NS, Pomytkin AP, Zolotarenko AD; The prospects for using of carbon nanomaterials as hydrogen storage systems,International journal of hydrogen energy,27,10,1063-1069,2002,Pergamon
13. Shul'ga Yu M, Martynenko VM, Tarasov BP, Fokin VN, Rubtsov VI, Shul'ga N Yu, Krasochka GA, Chapysheva NV, Shevchenko VV, Schur DV; On the thermal decomposition of the C60D19 deuterium fullerite,Physics of the Solid State,44,3,545-547,2002, Nauka/ Interperiodica
14. Pishuk VK, Schur DV, Bogolepov VA, Savenko AF, Zaginaichenko SYu, Zolotarenko AD, Mar'yanchuk IV, Prikhod'ko AB; Problem on production of highly dispersed extra pure powders, Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Ukraine,586-587,2001,
15. Matysina ZA, Zaginaichenko S Yu, Schur DV, Pishuk VK; Theoretical investigation of isopleths of hydrogen solubility in transition metals,Journal of alloys and compounds, 330,85-88,2002,Elsevier
16. Trefilov VI, Schur DV, Pishuk VK, Zaginaichenko S Yu, Choba AV, Nagornaya NR; The solar furnaces for scientific and technological investigation, Renewable energy, 16,1,757-760, 1999, Elsevier
17. Трефилов ВИ, Щур ДВ, Загинайченко СЮ; Фуллерены-основа материалов будущего, 2001, Laboratory 67
18. Schur Dmitry V, Zaginaichenko Svetlana Yu, Veziroğlu T Nejat, Javadov NF; The Peculiarities of Hydrogenation of Fullerene Molecules C60 and Their Transformation, Black Sea Energy Resource Development and Hydrogen Energy Problems,191-204,2013, Springer Netherlands
19. Kharlamov AI, Loytchenko SV, Kirillova NV, Kaverina SN, Vasilev AD, Fomenko VV, Zolotarenko AD, Kazimirov VP; Tubular and filamentous nanostructures of hexagonal silicon carbide, Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Ukraine, 572-574,2001,
20. Slys IG, Berezanskaya VI, Schur DV, Zaginaychenko SYu, Rogozinskaya AA, Adejev VM, Zolotarenko AD; Making the point metal coatings on the particles of hydride-forming intermetallides, Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Ukraine,404-405,2001,
21. Schur DV, Matysina ZA, Zaginaichenko S Yu; Theoretical study of interstitial atoms distribution in the bulk and at the surface of crystal. Surface segregation,Journal of alloys and compounds,330,81-84,2002,Elsevier
22. Schur DV, Matysina ZA, Zaginaichenko S Yu; Study of physico-chemical processes on catalyst in the course of synthesis of carbon nanomaterials,Hydrogen Materials Science and Chemistry of Metal Hydrides: Proceedings of the NATO Advanced Research Workshop on. Alushta Crimea, Ukraine, 16-22 September 2001,235,2002,Kluwer Academic Pub
23. Muratov VB, Meleshevich KA, Bolgar AS, Zolotarenko AD; Application of dynamic c-calorimetry method for investigation of enthalpy at hydride dissociation,Proceedings of VII International Conference "Hydrogen Material Science and Chemistry of Metal Hydrides", Ukraine,342-343,2001,



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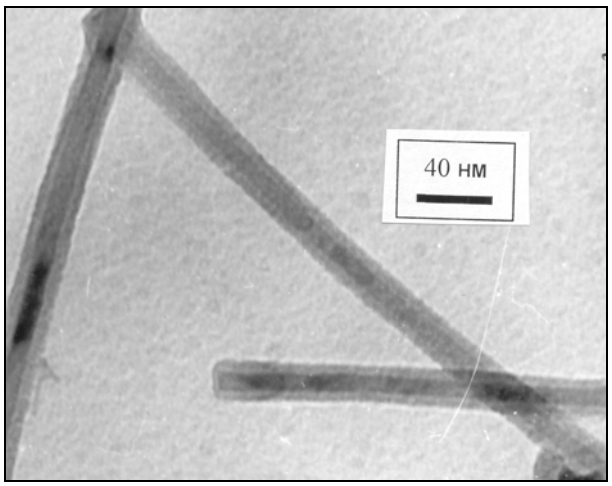

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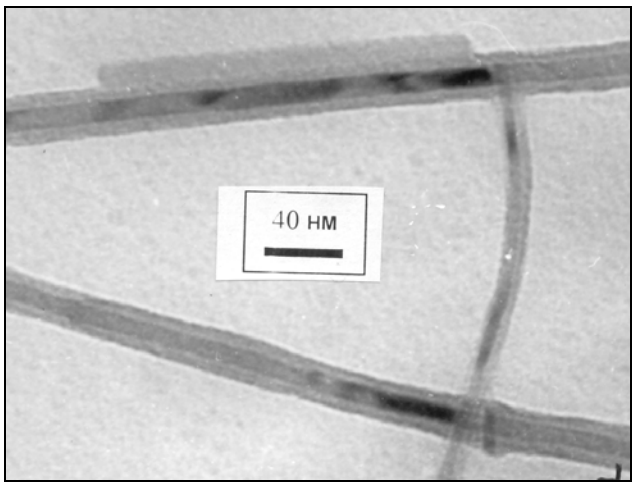

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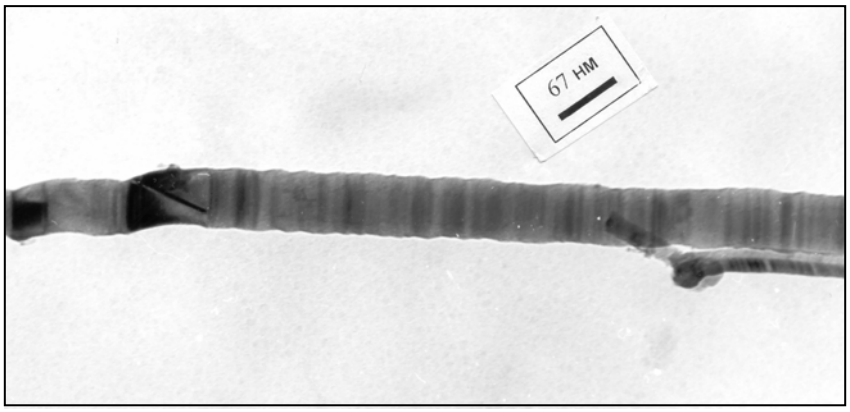

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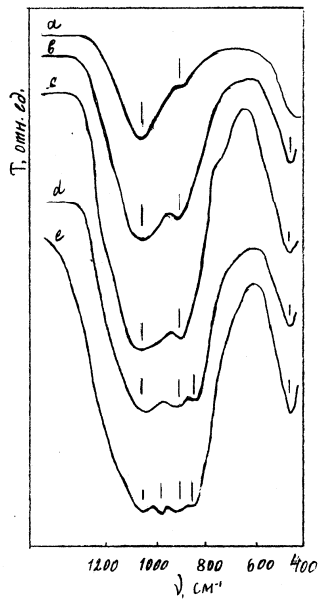

Рис. 5.