UDC 661.66.
IRSTI 61.67.09 DOI 10.55452/1998-6688-2021-18-1-52-56

PRODUCTION OF POROUS CARBON MATERIALS BASED ON SUPERHYDROPHOBIC SOOT AND OIL SLUDGE

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Abstract: *In this work, a porous carbon material (PCM) was obtained on the basis of superhydrophobic soot synthesized during combustion of a propane-butane mixture and an oil sludge from the Zhanaozen field. The resulting PCM was used as a carrier catalyst for the synthesis of carbon nanotubes. Synthesized samples of multi-wall carbon nanotubes (MWCNT) were confirmed by Raman spectra and a scanning electron microscope (SEM).*

Key words: *synthesis, porous carbon materials, superhydrophobic soot, oil sludge, multiwalled carbon nanotubes*

АСА ГИДРОФОБТЫ КҮЙЕ ЖӘНЕ МҰНАЙ ШЛАМЫ НЕГІЗІНДЕ КЕУЕКТІ КӨМІРТЕКТІ МАТЕРИАЛДАРДЫ ӨНДІРУ

Аңдатпа: *Бұл жұмыста кеуекті көміртекті материал (ККМ) пропан-бутан қоспасы жанғанда синтезделген аса гидрофобты күйе мен мұнай қалдығы негізінде алынған. Мұнай қалдығы ретінде Жаңаөзен кен орнының шламы таңдалды. Алынған ККМ тасымалдаушы катализатор ретінде нанотүтікшелер синтезі үшін қолданылды. Енді осы көпқабатты көміртекті нанотүтікшелердің (КҚКНТ) үлгілері спектрлерді комбинациялық шашырату әдісі және сканерлеуші электронды микроскоп арқылы зерттелді.*

Түйінді сөздер: *синтез, кеуекті көміртекті материал, аса гидрофобты күйе, мұнай шламы, көпқабатты көміртекті нанотүтікшелер*

ПРОИЗВОДСТВО ПОРИСТЫХ УГЛЕРОДНЫХ МАТЕРИАЛОВ НА ОСНОВЕ СУПЕРГИДРОФОБНОЙ САЖИ И НЕФТЯНОГО ШЛАМА

Аннотация: *В данной работе пористый углеродный материал (ПКМ) был получен на основе супергидрофобной сажи, синтезированной при сжигании пропан-бутановой смеси и нефтешлама с месторождения Жанаозен. Полученный ПКМ использовали в качестве катализатора-носителя для синтеза углеродных нанотрубок. Синтезированные образцы многостенных углеродных нанотрубок (MWCNT) были подтверждены рамановскими спектрами и сканирующим электронным микроскопом (SEM).*

Ключевые слова: *синтез, пористые углеродные материалы, супергидрофобная сажа, нефтешламы, многостенные углеродные нанотрубки*

Introduction

Nowadays, the amount of global production of porous carbon materials is about one million tons per year and continues to grow. The main areas of PCM use are systems for adsorption cleaning and separation of gas and liquid media. The use of pumas as hemosorbents, carriers for catalysts, adsorbents for chromatography, energy and gas storage systems, etc. is expanding [1, 2]. Their application areas are constantly expanding due to the development of methods for producing PCM with fundamentally new properties: carbon composite materials, molecular sieves, fibers, fullerenes, hollow nanotubes, etc. [3]. In this paper, it was considered methods for synthesizing the structure of the core shell from a porous carbon material, methods for the functionalization of a porous carbon material by direct inclusion of a heteroatom in carbon synthesis, halogenation, sulfonation, surface oxidation, and grafting [4, 5]. Over the past decade, the rapid development of nanotechnology has strengthened the carbon field and a number of new carbons with unique morphologies (such as carbon nanospheres, graphene or leaf-like carbon and ordered mesoporous carbon) and compositions (such as nitrogen-doped carbon) and carbon nitride, which are at the same time very useful for heterogeneous catalysis. Metal nanoparticles or metal oxides deposited on these carbon carriers provide interesting and exceptional characteristics in various catalytic processes, such as selective oxidation, hydrogenation, and oxygen reduction reactions [6].

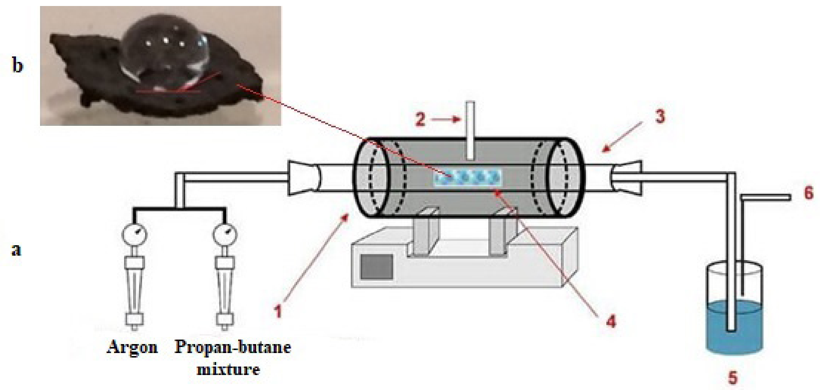
Carbon in dense form use its remarkable characteristics, as mechanical, thermal and electrical in various applications from common lead pencil to advanced management systems for spacecraft's. Simultaneously, its affinity for oxygen at high temperature can be beneficially used in making porous carbons. Porous carbons are prepared through controlled pyrolysis of carbonaceous materials, naturally occurring woods or synthetic polymeric materials [7-8].

However, the scale of consumption of porous carbon materials is largely limited by their relatively high cost. Therefore, an urgent task is to develop new methods for obtaining porous carbon materials with the required set of properties from cheap natural raw materials. As the last can be used the oil sludge and superhydrophobic soot [9] obtained during combustion of a propane-butane mixture.

Experimental part

Prelimenary mixed oil sludge and superhydrophobic soot were used to as a matrix to produce a carbon-based porous material. Then the mixed mixture in a special container was placed in a furnace to produce a porous material during carbonation at a temperature of 700°C for 40 minutes with argon purging.

The resulting porous material was impregnated with nickel nitrate crystallohydrate, dissolved in various concentrations in alcohol (ethanol 95%), and heated in a drying cabinet at a temperature of 100°C within 30 minutes. The obtained sample was heated in an argon atmosphere in a furnace at a temperature of 400°C during 1 hour. Then resulting sample was processed in furnace during combustion propane-butane mixture at different temperatures 650, 700, 750, 800°C.



1 - furnace; 2 - thermocouple;
3 - quarts tube;
4 - catalyst;
5 - controller of gas;
6 - emitted smoke

a – experimental installation;
b - porous carbon material

Fig. 1. – Photo of the experimental setup

Results and discussion

In this work samples of a porous carbon material containing nanotubes were studied at the Raman spectrometer NTegra Spectra (NT-MDT, Russia), whose excitatory radiation wavelength is 473 nm. The laser power is 20 mW. The signal accumulation time for all the spectra presented in this paper is 30 seconds.

Figure 2 shows scanning electron microscopic image of a porous material impregnated with nickel nitrate crystallohydrate, which was obtained at a temperature of 400°C.

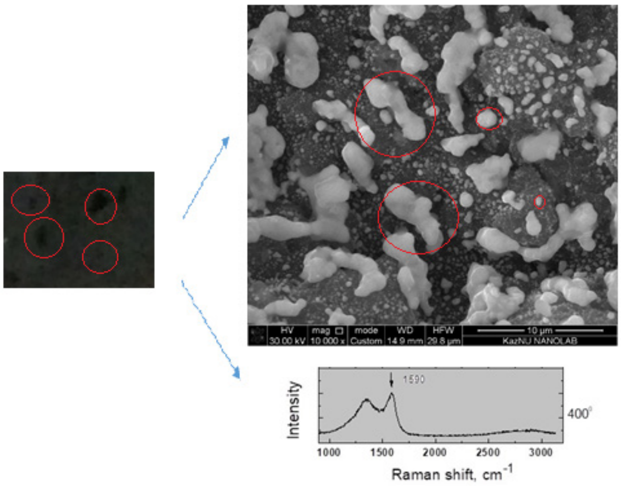


Fig. 2. – Electron microscopic image and Raman spectra of PCM obtained at a temperature of 400°C

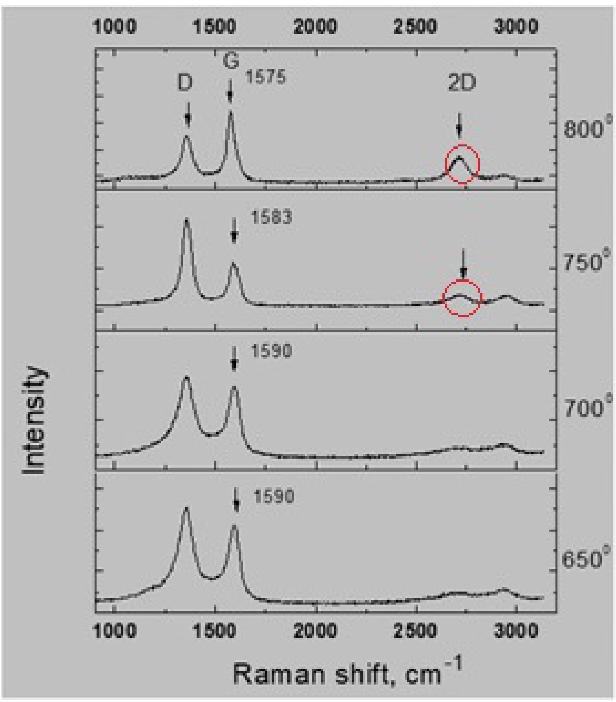
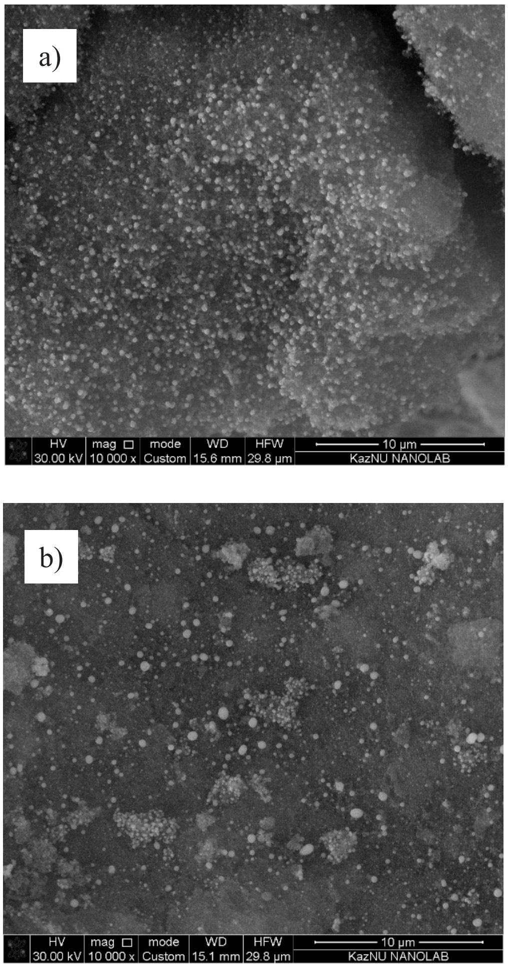
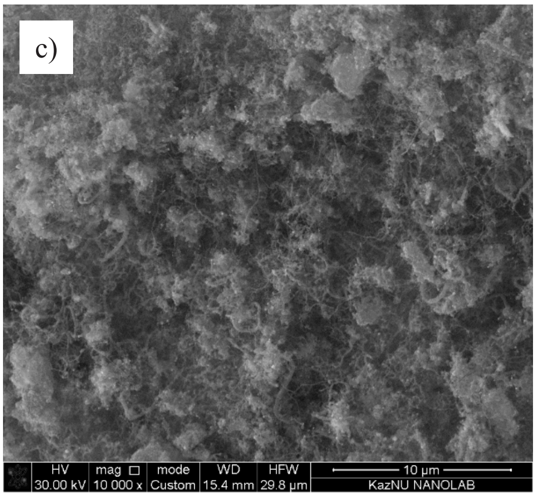


Fig.3. – The Raman spectra of samples in different temperatures

Raman spectra of a porous carbon material have shown that represented by two characteristic carbon (graphite) peaks G (1570-1600 cm^{-1}) and D (1360 cm^{-1}). SEM images of samples show presence of nickel particles (white particles of various configurations) on the surface of the sample, which will act as a catalyst, and these particles will contribute for the grow carbon nanotubes. Figure 3 shows that the Raman spectra of samples obtained at temperatures of 650, 700, 750, 800°C.

In samples carbonized at lower temperatures (650-700^{0}C), a wide band (G-peak) is observed in the region of 1590 cm^{-1}, which indicates a large contribution of the amorphous phase in the graphite structure. Peak D with an offset of 1360 cm^{-1} represents a wide band, the intensity for some carbonation temperatures exceeds the intensity of the G peak. Figure 4 depicts SEM images of samples obtained at temperatures 650, 700, 750, 800°C.





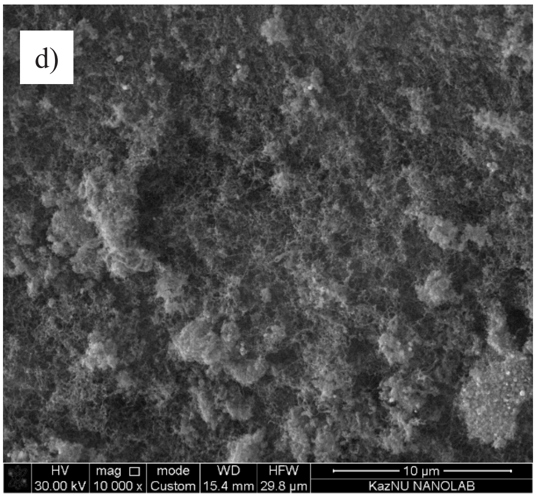


Fig.4. – SEM images of samples obtained at different temperatures

SEM images of samples obtained at temperatures 650, 700, 750, 800°C are fully consistent with the results of Raman spectroscopy.

It is important to note that with increasing temperature, the shift of the peak G to the low-frequency region is observed. The values of 1575-1580 cm^{-1} are typical for vibrations of C-C carbon atoms in the plane, and a narrow and intense peak in this region can indicate the presence of extended graphite carbon structures. In addition, in the spectra of samples obtained at 750 and 800^0C, the appearance of a peak of 2D 2710 cm^{-1} is noticeable.

Conclusion

To sum up, a porous carbon material was obtained based on superhydrophobic soot synthesized during combustion of a propane-butane mixture and an oil sludge. The resulting PCM was used as a carrier catalyst for the synthesis of carbon nanotubes. Synthesized samples of multi-wall carbon nanotubes (MWCNT) were confirmed by Raman spectra and a scanning electron microscope (SEM). Thus, it can be argued that the observed Raman spectra for high temperatures correspond to samples containing multi-walled carbon nanotubes with a small number of structural defects in the case of a carbonation temperature of 800°C.

Acknowledgments

This study was supported by the Ministry of Education and Science of the Republic of Kazakhstan in the framework of the scientific and technical program AR 08856321 of the Republic of Kazakhstan

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