Abstract In the given work oil residue, the oil sludge of Zhanaozen field and soot obtained during the combustion of propane-butane mixture with applying an electric field were selected among various types of natural materials to obtain porous carbon material (PCM). The PCMs obtained were used as a catalyst-carrier. The result of the research is synthesized multiwall nanotubes (MWNT), the identification of which is confirmed by the study of Raman spectra of the obtained samples. The search for new methods of creating catalytic systems (catalyst/carrier), which allow controlling the structure of carbon particles is an important problem, the solution of which may lead to the development of the approaches to the synthesis of carbon nanotubes (CNTs) with specific functional properties.

Keywords Carbon Porous Material, Oil Sludge, Soot, Carbon Nanotubes

1. Introduction

The total world production of porous carbon materials is, currently, about 1 million tons per year and it continues growing. The main practical applications of PCMs are the systems of adsorption purification and separation of gas and liquid media. The application of PCMs as hemosorbsents, carries for catalysts, adsorbents for chromatography, systems of the storage of gases and etc. [1-3] is extended. However, the scales of using PCMs, largely, are restricted by their sufficiently high cost. So, the urgent problem is the development of new methods to obtain porous carbon materials with a required set of properties from cheap types of raw materials. Oil sludge and soot obtained during the combustion of propane-butane mixture can be as cheap raw materials.

The activated carbons have been prepared by carbonization of precursors, i.e., raw materials with subsequent activation by physical treatment and/or chemical treatment. The activation procedure is conducted in order to enlarge the volume and size of the pores which were already created during the carbonization process and to create some new porosity. Most commonly employed activation methods are divided into physical and chemical activations. [3].

2. Use of Raman Spectroscopy to Characterize Carbon Nanotubes

Raman spectroscopy allows obtaining the information about purity, defects, and mutual alignment of carbon nanotubes (CNT) and also it helps to distinguish nanotubes from other carbon allotropes. This method is a quite successful in the description of structural properties of single-wall carbon nanotubes [4]. Raman spectroscopy can be successfully used for qualitative and quantitative analysis of the samples containing carbon tubular structures. The method of Raman spectroscopy allows identifying also multiwall carbon nanotubes (MWCNT). In the studies of Raman combination scattering MWCNTs are considered as a set of CNTs with a wide range of the variation of a diameter.

The interpretation of experimental spectra is always based on the well-studied results which were obtained for SWNCTs (single-wall carbon nanotubes) because of the not sufficient amount of theoretical works directed to understanding Raman spectrum of MWCNTs. This approach is useful, but it has a number of restrictions due to a number of effects being absent in SWNCTs which are observed in MWCNTs.

The typical spectrum of CNT has the following main characteristics:

1. Low-frequency band corresponding to radial mode is a main characteristic of SWNCT. The position of this mode depends on the diameter of a nanotube [5];
2. The group of peaks in the region of 1340 sm\(^{-1}\) which is called D-line is linked with a presence of the disorder in graphite materials. The appearance of D-peak in nanotubes is often considered as a measure of “disorder” like in graphite [6];

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3. G-line is composed of the group of peaks in the region of 1550-1600 sm\(^{-1}\). In graphite this region is represented by one peak at 1582 sm\(^{-1}\). This peak corresponds to tangent fluctuations of carbon sp\(^2\) atoms. This peak is a measure of graphitization of a sample;

4. The line in the region of 2700 sm\(^{-1}\) is the second harmonic of D-mode. It is noted as G' or 2D. This line indicates the distant of order in the sample structure and it appears in the process of double-background scattering;

5. The line in the region of 1610 sm\(^{-1}\) (and its second order of 3240 sm\(^{-1}\)) - intensity of this peak increases with increasing defects and it is connected with amorphous inclusions in the lattice of sp\(^2\) hybridized carbon.

Raman spectroscopy of MWCNTs is focused on using the presence or absence of these lines in the corresponding spectra to make conclusions about electron and structural characteristics of the sample of nanotubes [4].

As a rule the spectra of multiwall nanotubes are represented with G peak, the frequency and shape of which are comparable with a corresponding mode of graphite. The characteristic of single wall fluctuations is expressed in the presence of low-frequency RBM (radial breathing modes) and in splitting of G-peak. In many cases these two effects are not observed in the spectra of high-oriented samples. Thus, typical spectra of MWCNTs can be concluded to have similarities with a graphite spectrum and show small or lacking effect of cylindrical geometry.

3. Experimental

To obtain carbon porous material pre-mixed oil sludge and soot were used. Then, stirred mixture in a special container was placed into the furnace to obtain porous material in the process of carbonization at 700 \(^\circ\)C for 40 minutes with argon blowing off.

The obtained porous material was impregnated with crystal hydrate of nickel nitrite which was dissolved in alcohol in different concentrations (ethanol 95 %) and it was heated in the drying cabinet at 100 \(^\circ\)C for 30 minutes. The sample obtained in argon atmosphere was heated in the furnace at 400 \(^\circ\)C for an hour, Fig.1.

In the Fig.2 presented the obtained carbon porous material.

![Figure 1](image1.jpg)

**Figure 1.** The photo of the experimental setup

Fig. 2 show that obtained carbon porous material has superhydrophobic properties.

The Fig.3 gives the electron-microscopic image of carbon porous material obtained at 400 \(^\circ\)C.

![Figure 2](image2.jpg)

**Figure 2.** The photo of carbon porous material

Then the sample obtained was treated in the furnace with propane supply at various temperatures of 650, 700, 750, 800 \(^\circ\)C.

The samples obtained in this way were investigated in the Laboratory of an engineering profile at Al-Farabi Kazakh National University.

4. Result Discussion

In the given work the samples of porous carbon material containing nanotubes were investigated using NTegra Spectra Raman spectrometer (NT-MDT, Russia) with a the wave length of excitation radiation of 473 nm. The laser power is 20 mW. The time of signal accumulation for all spectra presented in the work is 30 seconds, Fig.4.
As figure 4 shows, the spectra of all the investigated samples are represented by two characteristic carbon (graphite) G peaks in the region of 1570-1600 cm⁻¹ and D at ~1360 cm⁻¹.

In the samples carbonized at lower temperatures (400-700 °C) a wide band (G peak) is observed in the region of 1590 cm⁻¹ that says about the great contribution of an amorphous phase to graphite structure. D peak with a shift of 1360 cm⁻¹ is a wide band with an intensity which exceeds the intensity of G peak for some temperatures of carbonization that also evidences about quite high degree of disorder in the structure of sp² hybridized carbon.

It important to notice that with increasing temperature the shift of G peak into low-frequency region is observed. The values of 1575-1580 cm⁻¹ are characteristic of the fluctuations of C-C carbon atoms in the plane, and a narrow and intensive peak can evidence about the presence extended graphite carbon structures. Besides, in the spectra of the samples obtained at 750 and 800 °C the appearance of 2D peak can be noticed at 2710 cm⁻¹. As it was said above the given peak indicates the distant order in the sample structure.

Besides the samples which were obtained at temperatures of pyrolysis 650, 700, 750 and 800 °C investigated by transmission electron microscopic JEM 1011.

Thus, electron microscopy and Raman studies of the samples obtained at temperatures of 750 and pyrolysis 800 °C confirm the high content in the obtained samples of carbon nanotubes. The length of the nanotubes consist to 2 microns diameter ranging from 30 to 80 nm.
5. Conclusions

Thus Raman spectra being observed for high temperatures can be asserted to correspond to the samples containing multiwall carbon nanotubes with a small amount of the defects of the structure at carbonization temperature of 800 °C.

REFERENCES

[1] Marsh H., Rodriguez-Reinoso F. Activated Carbon. Elsevier, 2006, p. 537.

[2] Manocha S. Porous carbons. Sadhana. Vol.28, Parts1&2, 2003, pp. 335–348. India

[3] Gyoung-Ja Lee and e.t.c. Synthesis and Characterization of Nanoporous Carbon and Its Electrochemical Application to Electrode Material for Supercapacitors. Modern Aspects of Electrochemistry, Number 41, pp. 139–195. Edited by C. Vayenas et al., Springer, New York, 2007.

[4] Z.A. Mansurov. Producing Nanomaterials in Combustion // Combustion, Explosion, and Shock Waves. - 2012. - V. 48, No.5. P.561-569.

[5] A. R. Kerimkulova, B. B. Mansurova, M. K. Gil’manov and Z. A. Mansurov. Nanoporous Carbon Sorbent for Molecular-Sieve Chromatography of Lipoprotein Complex // Russian Journal of Physical Chemistry. - 2012. - V.86, No.6, pp.1004-1007.

[6] Jorio A, Pimenta MA, Souza AG, Saito R, Dresselhaus G, Dresselhaus MS. Characterizing carbon nanotube samples with resonance Raman scattering. New J Phys. 2003;5:139.1–17.

[7] Rao AM, Richter E, Bandow S, Chase B, Eklund PC, Williams KA, et al. Diameter-selective Raman scattering from vibrational modes in carbon nanotubes. Science 1997;275(5297):187–91.

[8] Maultzsch J, Reich S, Thomsen C. Chirality-selective Raman scattering of the D mode in carbon nanotubes. Phys Rev B 2001;64(12):121407-1–4.

[9] John H. Lehan, Mauricio Terrones, Elisabeth Mansfield, Katherine E. Hurst, Vincent Meunier, Evaluating the characteristics of multiwall carbon nanotubes, Carbon, 49 (2011) 2581 –2602.