Journal of Molecular Liquids 253 (2018) 340–346

Contents lists available at ScienceDirect

Journal of Molecular Liquids

journal homepage: www.elsevier.com/locate/molliq

Novel and economic method of carbon nanotubes synthesis on a nickel magnesium oxide catalyst using microwave radiation

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ARTICLE INFO

Article history:
Received 13 October 2017
Received in revised form 23 November 2017
Accepted 9 January 2018
Available online 10 January 2018

Keywords:
Economic carbon nanotube synthesis
Large surface area
Microwave radiation
Catalysts

ABSTRACT

Carbon nanotubes (CNTs) are gaining increased importance in many fields of science and technology due to their unique properties of greater surface area, mechanical strength, electrical and thermal conductivity. A novel method of carbon nanotubes synthesis on a nickel magnesium oxide catalyst using microwave radiation was developed and presented. In the present paper, the possibility of modifying a Ni-MgO catalyst, for carbon nanotube synthesis with microwave radiation (0.8 kW and 2.45 GHz) at the production stage, is studied. The effect of this radiation on the catalyst characteristics (specific surface area, catalytic activity, etc.) is experimentally considered. It is shown that the use of short term exposure to the microwave radiation in preparing the catalyst made it possible to increase its specific surface area from 5.2 to 9.1 m^2/g. The implementation of chemical vapor deposition of the catalyst, modified with the microwave radiation for 30 s, contributed to an increase in the yield of a nanostructured material by 40–45%; making carbon nanotubes inexpensive in production. The synthesized carbon nanostructured material predominantly represented multilayered nanotubes with a diameter of 10–40 nm. The developed method was capable to produce 40–45% yield with almost two time's greater surface area. The synthesized carbon nanotubes may be used for various purposes including water treatment due to the economy in production and large surface area.

1. Introduction

The different nanomaterials including carbon nanotubes (CNTs) are gaining increased importance in many fields of science and technology [1–9]. These comprise nanoelectronics, photovoltaics, nanosensors and chemical/biological sensing. It is due to their unique properties of greater surface area, mechanical strength, electrical and thermal conductivity [10–14]. CNTs are often used as modifiers for various materials (polymers, concrete, rubber, etc.). For each application field, commercially produced CNTs with certain characteristics are required. The CNT characteristics are affected by both the synthesis parameters and the catalyst compositions. The formation and further growth of nanostructures take place on active centers of the catalysts. Therefore, the development of technologies for obtaining an efficient catalyst appears to be an urgent issue. The activity of catalytic systems may be increased using chemical or physical and mechanical processes, but these do not always ensure achievement of the required degree of activity. The non-conventional methods for the catalyst modification prove the relevance of developing and studying new methods for increasing the catalytic activity [15]. At present, microwave radiation is widely used to intensify various processes. It ensures rapid and efficient heating of the systems [16,17]. The application of this method is extensively covered in scientific publications of the last decade [18–20]. However, most of the papers are fragmentary. There is a gap between research and their practical use in manufacturing. The literature search of scientific and technical papers has shown that researchers often pay attention to the possibilities of using microwave radiation in various fields of science and technology, including catalysis processes [21–23]. Thus, it was proved that that the effect of microwave radiation on a metal catalyst slowed down its carbonization, thereby, increasing the degree of its chemical activity [24]. Besides, the use of microwave treatment makes it possible to obtain well-crystallized powders of oxide materials with low defectiveness.

The application of microwave treatment of impregnated catalysts to produce alkene oxide allows for obtaining a finely dispersed layer of metallic silver over aluminum oxide with a particle size of 2 to 100 nm [25]. Thus, the use of microwave radiation leads for synthesizing catalysts with a more uniform particle distribution [26]. Under the

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https://doi.org/10.1016/j.molliq.2018.01.062
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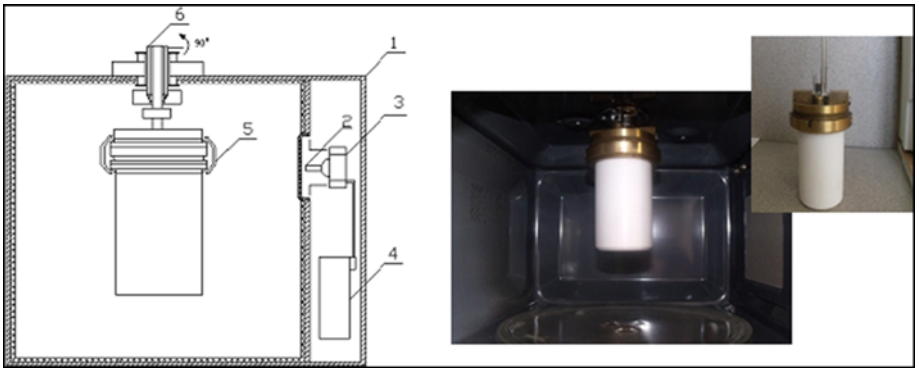

Fig. 1. A microwave batch setup for catalyst modification at the preparation stage. 1. Housing, 2. waveguide, 3. magnetron, 4. control unit, 5. solution reactor and 6. rotary mechanism.

influence of microwave radiation, it is possible to carry out physical and chemical processes such as dehydration, decomposition of salt and hydroxyl precursors, and synthesis and sintering of heterogeneous systems. The use of microwave heating allows not only to shorten the duration of synthesis of multi-component oxide products with different crystal structures, but also to lower the temperature of the synthesis [26]. Oxide phases obtained using the microwave radiation is not inferior to reference samples obtained by conventional heat treatment in terms of the functional properties. It is advisable to employ the observed effects in obtaining catalysts for carbon nanostructured material synthesis. Based on the existing scientific and technical literature available, it may be assumed that the use of the microwave radiation in obtaining catalysts for synthesis of carbon nanostructured materials (carbon nanotubes) can significantly be improved their efficiency. Thus, the modification of heterogeneous metal oxide catalysts, at the stage of their preparation, using non-conventional methods (such as exposure to ultrasound, magnetic field, microwave radiation, etc.) promotes an increase in the activity of carbon crystallization centers [27,28]. It is supposed that the microwave radiation will allow for rapidly and uniformly raising the temperature of the substance in the entire volume at the catalyst preparation stage, thereby, excluding local overheating of some regions in the catalytic system under modification. Considering the above facts, the aim of the present research was to study the efficiency of microwave radiation in obtaining a Ni/MgO catalyst for the CNT synthesis through thermal decomposition, and to investigate its effect on the characteristics of the catalyst and the nanoproduct (synthesized carbon nanostructured materials). The results are presented herein.

2. Experimental

2.1. Chemicals used

All the chemicals used were of reagent grade. The used chemicals were $Ni(NO_3)_2.6H_2O$, $Mg(NO_3)_2.6H_2O$ and NH_2-CH_2-COOH. All the chemicals were supplied by Laverna Story Engineering Ltd., Moscow, Russia. The purified water was prepared using a Millipore Milli-Q, Bedford, M.A., USA system.

2.2. Instruments used

The various major instruments used were Microsizer 201-C laser particle analyzer (Granat-E JSC, St. Petersburg, Russia), Sorbptometr-M specific surface analyzer (Katakon CJSC, Novosibirsk, Russia), STA 449 F3 Jupiter synchronous thermal analyzer (Netzsch, Selb, Germany), two beam scanning electron microscope (Carl Zeiss, Jena, Germany), a DXR confocal Raman microscope (Thermo Fisher Scientific, Waltham, MA, USA).

2.3. Procedure

A Ni/MgO metal oxide catalyst (composition - 80:20) was obtained via thermal decomposition. It was chosen as a material under study. The catalyst precursors were crystal hydrates - $Ni(NO_3)_2{\cdot}6H_2O$ and $Mg(NO_3)_2{\cdot}6H_2O$, and amino acetic acid - (NH_2—CH_2—COOH). The thermal decomposition method, representing a way of preparing catalyst for the synthesis of carbon nanostructured materials, included the stages of intermixing, dissolving components in distilled water at a temperature of ≤60 °C, and thermal decomposition at 550 °C. The mixture was not overheated at >60 °C during the first stage due to the possibility of side reactions of nitrate ion reduction with organic compounds to nitrogen oxides, and chances of release of nitric acid vapors.

In order to increase the efficiency of the catalyst prior to the thermal decomposition stage, the solution of the initial Ni/MgO catalyst components (electrical conductivity 847 μS/cm and pH = 2.17) was exposed to microwave radiation with a frequency of 2.45 GHz and a power of 800 W. To this end, a laboratory batch setup was used (Fig. 1). The setup consists of housing (1), waveguide (2), magnetron (3), control unit (4), solution reactor (5), and rotary mechanism (6) required for

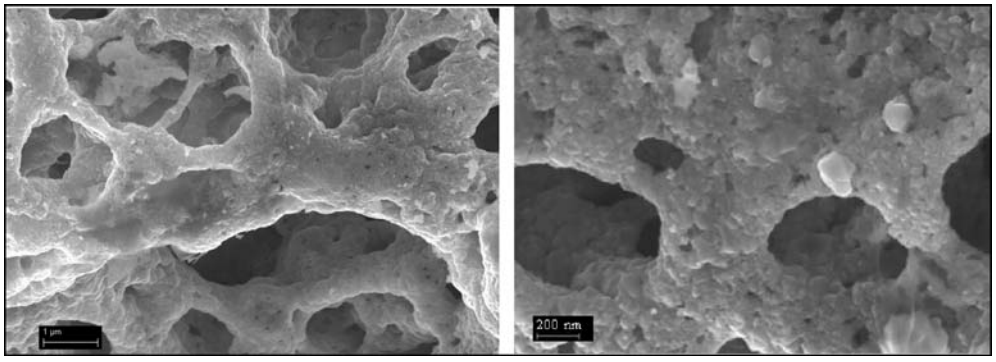

Fig. 2. SEM images of the initial Ni/MgO catalyst.