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Effect of Sol gel and hydrothermal catalyst synthesis methods in production of CNTs

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***Abstract*— In this study CNTs have been produced in methane decomposition method successfully, however prior to the production of CNTs the iron catalyst has been prepared by different sol-gel and hydrothermal methods to investigate their effect on the obtained CNTs. It was concluded that the sol gel method catalyst lead to the CNTs with better morphology but having less yield, however those produced by hydrothermal methods lead to the higher yield of the CNTs.**

***Index Terms*— CNTs, synthesis, MWNTs**

I. INTRODUCTION

Among the most applicable carbon nano structures Graphene[1] and Carbon nanotubes are very popular. Carbon nanotubes (CNTs) were firstly observed and described in 1952 by Radushkevich and Lukyanovich1 and later in 1976 the single (or double) walled carbon nanotubes were observed by Oberlin et al[2]. In more recent history the discovery of CNTs is attributed to Iijima as the first scientist who described the multiwall carbon nanotubes (MWNTs) preparation process after a random event during the test of a new arc evaporation method for C60 carbon molecule fabrication in 1991[3].

They have excellent properties that make them potentially useful in a wide variety of applications in mechanical, structural, textile, thermal, electrical.[4]. They can be produced via different methods of laser ablation[5], CVD[6] , Arc discharge[7] , high pressure carbon monoxide[8] . in such methods carbonous gases such as hydrocarbons like methane [9], acetylene[10], or carbon dioxide[11] can be used.

The effect of synthesis method is one of the important factors in catalyst preparation[12] for different applications . In the present work this effect has been investigated on the synthesis of CNTs.

II. METHODOLOGY

The methods for sol gel and hydrothermal methods are taken from Allaedini et al with some modification [13, 14]. The materials were bought from sigma Aldrich and were used without any further purification. The chemicals used in the synthesis are of above 99% purity.

Iron nitrate hexahydrate ($Fe(NO_3)_2.6H_2O$), and oleic acid ($C_{18}H_{34}O_2$) were used as precursors. 20 g of iron nitrate hexahydrate was added slowly to 100 ml of ethyl alcohol and stirred at 100◦C until complete dissolution was achieved. Oleic acid was then added to the resulting solution slowly with a constant stirring to produce a thick white gel. The product was dried at 100◦C for 24 h. The samples were calcined at 600 ◦C for 4 h.

Ferric nitrate nanohydrate ($Fe(NO_3)_3 9H_2O$), and NaOH were used as precursors and dissolved in deionized water (50 ml). An aqueous solution of 3 M NaOH was used as the precipitating agent. The as prepared metal nitrate solutions were added to the boiling solution of 3 M NaOH (25 ml) at 150 C. The obtained solution was stirred for 6 h while maintaining the reaction temperature at 150 C. The pH of the mixture was adjusted to 9 to control the nucleation and was further stirred for 3 more hours[15]. The prepared solution was then transferred to a 100 ml Teflon-lined stainless autoclave. The autoclave was sealed and heated to 180 C for 10 h. The pressure in the sealed autoclave became lower than the equilibrium vapor tension of the pure water at this temperature due to the presence of NaOH in the solution. The nano powders were formed .

In order to synthesize CNTs 200 mg of the catalysts were placed in a reactor, heated in Ar up to 200 C when purged and then up to 800 C and after the temperature was stabilized the methane was introduced with a flow rate of 100 cm/m3 for 5 hrs[16]

III. RESULT

Looking at the morphologies of the SEM it can be seen that the iron nano powders in both methods are homogenous in shape, however a bit more spherical in sol gel methods, but generally both have a range of spherical distribution shown in figure 1 .

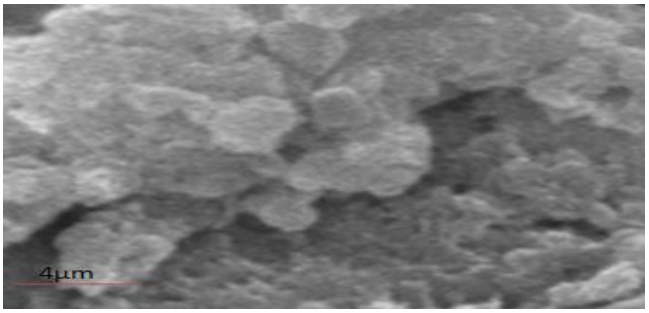

Figure 1-SEM images of the iron nano powders by (left) sol gel and (right) hydrothermal

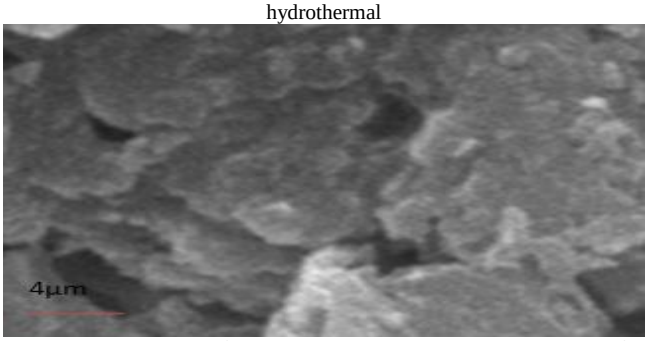

Figure 2 shows the yield of the carbon deposition which was calculated after the reaction by:

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Depositions= weight after reaction- catalyst weight before reaction / weight before reaction *%

It can be seen that higher deposition was occurred when samples were prepared by sol gel method which is about 63% but for the hydrothermal method, it's about 43%.

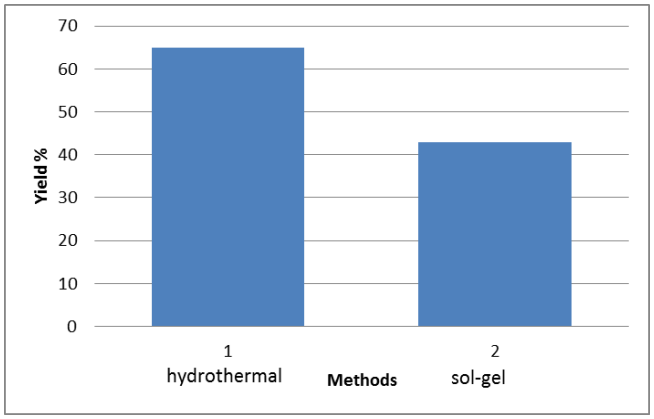

Figure 2 – Deposition yield of carbon

In terms of the obtained morphology the SEM micrographs of the CNTs are shown in figure 3. The CNT which were obtained by sol gel method have better morphology and longer and more obvious tubes than the ones obtained from hydrothermal method.

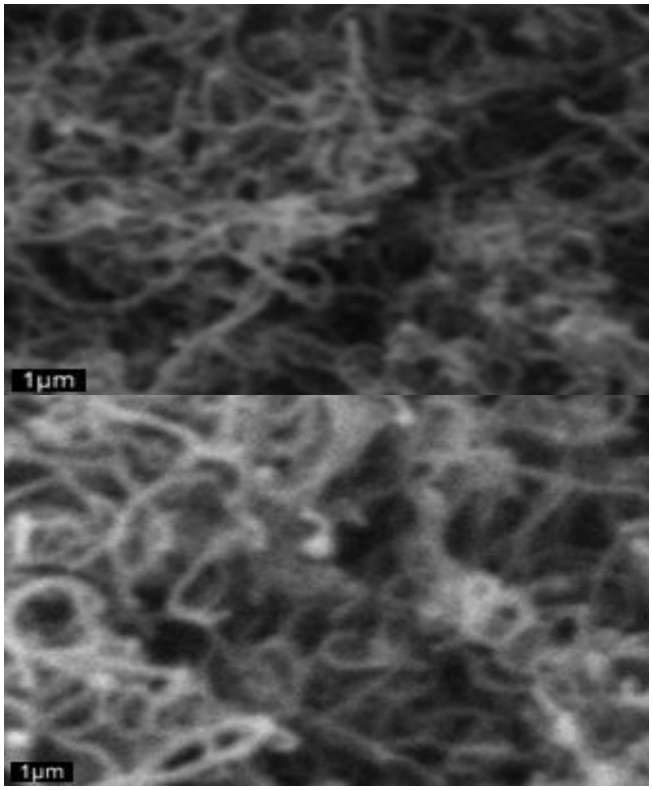

Figure 1-SEM images of the CNT from (left) sol gel and (right) hydrothermal catalyst methods

IV. Conclusion

The effect of catalyst methods of preparation on the CNTs formation was investigated. It was observed that sol gel methods lead to the production of CNT with better morphology but fewer yields; however those CNTs obtained from hydrothermal method resulted in higher yield. These finding can be investigated in details and can be a research topic for optimization of CNT production in terms of increasing yield and at the same time improving morphology of CNTs.

V. References

[1] G. Allaedini, P. Aminayi, S. M. Tasirin, and E. Mahmoudi, "Chemical Vapor Deposition of Methane In The Presence of Cu/Si Nanoparticles as a Facile Method for Graphene Production," Fullerenes, Nanotubes and Carbon Nanostructures, pp. 00-00, 2015.

[2] N. Jawahara, E. Surendraa, and K. R. Krishnaa, "A Review on Carbon Nanotubes: A Novel drug Carrier for Targeting to Cancer Cells."

[3] M. Terrones, "Science and technology of the twenty-first century: synthesis, properties, and applications of carbon nanotubes," Annual review of materials research, vol. 33, pp. 419-501, 2003.

[4] G. Allaedini, S. M. Tasirin, P. Aminayi, Z. Yaakob, and M. Z. M. Talib, "Bulk production of bamboo-shaped multi-walled carbon nanotubes via catalytic decomposition of methane over tri-metallic Ni–Co–Fe catalyst," Reaction Kinetics, Mechanisms and Catalysis, pp. 1-12.

[5] Y. Zhang, H. Gu, and S. Iijima, "Single-wall carbon nanotubes synthesized by laser ablation in a nitrogen atmosphere," Applied physics letters, vol. 73, pp. 3827-3829, 1998.

[6] G. Allaedini, P. Aminayi, and S. M. Tasirin, "The Effect of Alumina and Magnesia Supported Germanium Nanoparticles on the Growth of Carbon Nanotubes in the Chemical Vapor Deposition Method," Journal of Nanomaterials, vol. 501, p. 961231, 2015.

[7] M. Glerup, J. Steinmetz, D. Samaille, O. Stephan, S. Enouz, A. Loiseau, et al., "Synthesis of N-doped SWNT using the arc-discharge procedure," Chemical Physics Letters, vol. 387, pp. 193-197, 2004.

[8] V. Georgakilas, D. Voulgaris, E. Vazquez, M. Prato, D. M. Guldi, A. Kukovecz, et al., "Purification of HiPCO carbon nanotubes via organic functionalization," Journal of the American Chemical Society, vol. 124, pp. 14318-14319, 2002.

[9] J. Kong, A. M. Cassell, and H. Dai, "Chemical vapor deposition of methane for single-walled carbon nanotubes," Chemical Physics Letters, vol. 292, pp. 567-574, 1998.

[10] K.-E. Kim, K.-J. Kim, W. S. Jung, S. Y. Bae, J. Park, J. Choi, et al., "Investigation on the temperature-dependent growth rate of carbon nanotubes using chemical vapor deposition of ferrocene and acetylene," Chemical physics letters, vol. 401, pp. 459-464, 2005.

[11] G. Allaedini, S. M. Tasirin, and P. Aminayi, "Synthesis of CNTs via Chemical Vapor Deposition of Carbon Dioxide as a Carbon Source in the Presence of NiMgO," Journal of Alloys and Compounds, 2015.

[12] G. Allaedini, S. M. Tasirin, M. Talib, M. Zainal, P. Aminayi, and I. Puspasari, "Comparative Study on the Microstructure and Photoluminescence Properties of SnO2 Nano Particles Prepared by Different Methods," in Applied Mechanics and Materials, 2015, pp. 132-136.

[13] G. Allaedini, S. M. Tasirin, and P. Aminayi, "Magnetic properties of cobalt ferrite synthesized by hydrothermal method," International Nano Letters, pp. 1-4.

[14] G. Allaedini, P. Aminayi, and S. M. Tasirin, "Structural properties and optical characterization of flower-like Mg doped NiO," AIP Advances, vol. 5, p. 077161, 2015.

[15] G. Allaedini, S. M. Tasirin, and M. Pudukudy, "Effect of PH on Cobalt Oxide Nano Particles Prepared by Co-Precipitation Method," 2014.

[16] G. Allaedini, S. M. Tasirin, J. Sahari, M. Talib, and M. Zainal, "The Effect of Co/Pd MgO Supported Catalyst Calcination Temperature on the Yield and Morphology of CNTs via Methane Decomposition," in Advanced Materials Research, 2014, pp. 148-151.