ABSTRACT

Carbon is the most versatile element in the periodic table, owing to the type, strength and number of bonds it can form with many different elements. The diversity of bonds and their corresponding geometries enable the existence of structural isomers, geometric isomers and enantiomers. Carbon nanostructures have attracted a great deal of interest due to the discovery of fullerenes. Widely recognized as the quintessential nanomaterial, carbon nanotubes (CNTs) have already compiled an impressive list of superlatives since their discovery in 1991 by Sumio Iijima. The basic structural unit of CNT is a graphitic sheet rolled into a cylinder, with the tube tips closed by hemispherical or polyhedral graphitic domes. There are different types of CNTs such as SWCNTs and MWCNTs. Further, the SWCNTs were classified as zig-zag, armchair and chiral nanotubes by the way of rolling the graphene sheet.

CNTs can be synthesized by various methods such as Arc discharge, Laser ablation and Chemical vapour deposition (CVD). The first two methods, solid-state carbons are used as a carbon source and needed high temperature to evaporate it. In that sense, all eyes are turning to the CVD of hydrocarbons over metal catalysts. CVD is considered as the best method for producing CNTs with varying diameters and structures on a large scale, the products formed were of high purity, allows the growth of large amount of CNT at lower cost and reaction occurs at moderate temperature.

The catalyst is an important issue for efficient transformation of carbon clusters into CNTs. Catalytic synthesis is known to be one of the most effective ways to control the selectivity to SWCNTs and MWCNTs with high yield and it has the potential to be scaled-up at relatively low cost. Mesoporous MCM-41 and KIT-6 materials are best appreciated in systems where molecular recognition is needed for selective adsorption and separation processes, chemical sensors and nanotechnology. Transition metal particles can be incorporated into the 2D hexagonal MCM-41 and 3D cubic KIT-6 molecular sieves, stabilizing dispersed catalytic sites for the formation of CNTs. Among the mesoporous materials, KIT-6 have been several advantages such as high thermal stability due to construction of walls, large tunable pores and finally KIT-6 exhibits a three-dimensional cubic Ia3d symmetric structure with interpenetrating bicontinuous network of channels. The metal incorporated and impregnated mesoporous 3D cubic KIT-6 molecular sieves are suitable candidate for the production of well graphitized MWCNTs. Hence in the present investigation, different metal substituted MCM-41 and KIT-6 materials are proposed as the most suitable catalysts for the formation of CNTs by decomposition of acetylene through CVD method.

The present study involves hydrothermal synthesis of Si-MCM-41, Cr-MCM-41, Fe-MCM-41, Zn-MCM-41 and Zn-Fe-MCM-41 molecular sieves with Si/M ratios of 50, 75, 100 and 125, where M = Cr, Fe, Zn and Zn-Fe. The transition metals such as Fe, Co, Mn, Ni, Ti, Ru and Pd were loaded with 0.2 wt. % individually over Cr-MCM-41 by wet impregnation method. Antimony with different wt. % of 1.0, 2.0, 3.0, 5.0, 10.0, 15.0 and

20.0 was loaded on Si-MCM-41 by wet impregnation method. Mesoporous 3D cubic Fe-KIT-6 with various Si/Fe ratios (50, 75 and 100) were synthesized hydrothermally and Zn with 0.25, 0.50 and 0.75 wt. % was loaded on them by wet impregnation method. Fe and Zn, the ratio of 3:1 with 1.0, 2.0 and 3.0 wt. % was loaded on MgO by wet impregnation method.

The above synthesized catalytic materials were characterized by various physico-chemical techniques such as ICP-AES, XRD, N_2 sorption isotherms, TGA, FT-IR Spectroscopy, DRS-UV, SEM and TEM. The growth of SWCNTs or MWCNTs was carried out using the above synthesized catalytic materials by CVD method using acetylene as carbon precursor. The reaction parameters such as metal concentration over the catalytic template, flow rate of carbon precursors, growth time and temperature were optimized individually for the production of well graphitized CNTs over the above said catalytic materials. The as-synthesized carbon deposits formed over different catalytic materials were purified by acid treatment followed by air oxidation. The purified CNTs were characterized by various physico-chemical techniques such as XRD, TGA, SEM, TEM and Raman spectroscopy. From the observations, the mesoporous metal containing KIT-6 produced MWCNTs were found to possess high thermo-mechanical properties due to their wall thickness (number of graphene layers). The well graphitized MWCNTs were functionalized and confirmed by SEM and FT-IR spectroscopy. The extraordinary mechanical properties of CNTs and their reinforcements are potential for the production of ultra-strong nanocomposites that are in-turn compatible for aerospace applications.

In general, neat epoxy resins pose some major disadvantages such as low peel, low impact and tensile strength at elevated temperatures and also low chemical resistance. The thermo-mechanical properties of the neat epoxy resin have been improved by the addition of MWCNTs or f-MWCNTs. The fabricated epoxy resins with MWCNTs and f-MWCNTs were compared with one another to study the dispersion behaviour. MWCNTs/Epoxy and f-MWCNTs/Epoxy nanocomposites were fabricated with different wt. % of MWCNTs and f-MWCNTs respectively. Thermo-mechanical properties such as flame retardancy, thermal stability, tensile strength, flexural strength and hardness were investigated for fabricated nanocomposites. 1 wt. % of MWCNTs/f-MWCNTs loaded epoxy resin showed the highest improvement in tensile strength, flexural strength and hardness as compared to neat and other epoxy systems. The rate of burning is decreased with respect to the wt. % of MWCNTs/f-MWCNTs. Moreover, f-MWCNTs/epoxy nanocomposites exhibited high thermo-mechanical properties than others.

The experimental research work reveals that the above said metal containing mesoporous MCM-41 and KIT-6 materials can be a convenient eco-friendly alternative to all other catalyst for the formation of SWCNTs and MWCNTs with high quality and large quantity. Especially, metal containing mesoporous KIT-6 materials were suitable candidate for the large scale production of well graphitized MWCNTs. Fabricated nanocomposites by using MWCNTs/f-MWCNTs exhibited good thermo-mechanical properties which might be a good opening for new avenues to various industrial applications.