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Catalytic carbonization of polypropylene by the combined catalysis of activated carbon with Ni_2O_3 into carbon nanotubes and its mechanism

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ABSTRACT

A one-pot approach was established to prepare carbon nanotubes (CNTs) through the carbonization of polypropylene (PP) by the combined catalysts of activated carbon (AC) with Ni_2O_3. The combination of AC with Ni_2O_3 showed a synergistic catalysis on the catalytic conversion of PP to form CNTs. The effects of the content of AC and carbonization temperature on the yield of CNTs were studied. The morphology, phase structure and thermal stability of the obtained CNTs were analyzed by means of SEM, TEM, XRD, TGA and Raman. In this synergistic catalysis, the surface functional groups (especially carboxylic groups) of AC were proved to be the key factor. AC not only promoted the cracking of PP fragment radicals into light hydrocarbons and the dehydrogenation and aromatization of the resultant light hydrocarbons into aromatic compounds, but also promoted the formation of the intermediate aromatic compounds or polycyclic aromatic hydrocarbons (PAHs) from the reaction of light hydrocarbon products and aromatic compounds. Additionally, AC assisted *in situ* Ni catalyst (originated from the reduction of Ni_2O_3) catalyzing the dehydrogenation and aromatization of intermediate aromatic compounds or PAHs products to form CNTs. At last, a layer-by-layer assembled mechanism based on benzene rings for the growth of CNTs using PP as carbon source and combined AC/Ni_2O_3 as catalysts was proposed. This mechanism will help to understand the growth mechanism of CNTs using virgin or waste plastics as carbon sources. More importantly, this approach offers a new potential way for large-scale production of CNTs from waste plastics using cheap AC as a cocatalyst.

1. Introduction

The tremendous applications of plastic materials in our modern society have caused a severe environmental pollution. Nowadays, waste plastics are treated predominantly by landfill, incineration, mechanical recycling and chemical recycling. Landfill and incineration of waste plastics waste resources and harm the environment. Mechanical recycling is a preferred recovery route for relatively clean waste plastics, but it is sensitive to unsorted or contaminated waste plastics. Chemical recycling has the advantage of recovering the petrochemical components of waste plastics, which can then be used to re-manufacture plastic or to make other synthetic chemicals. Nevertheless, the development of a new technically and economically feasible chemical recycling process for the waste plastics has been a hot topic.

Among these waste plastics, waste polyolefins are the main components. The content of carbon in polyolefins is about 85.7 wt%, hence waste polyolefins are affluent resources for the production of carbon nanomaterials, such as carbon nanotubes (CNTs). Recently, extensive studies have been conducted to transform virgin or waste polyolefins such as polypropylene (PP) and polyethylene (PE) into CNTs [1–10]. In our previous work [3–6], the combination of solid acids such as organically modified montmorillonite (OMMT) or zeolite with nickel catalysts (including Ni/SiO_2–Al_2O_3, Ni_2O_3, NiO and $Ni(OH)_2$) has been found to be efficient for high-yielded conversion of PP into CNTs through "carbenium ions" mechanism. The carbenium ions from OMMT or zeolite not only promote the degradation of PP to predominantly form products with lower carbon numbers that can be easily catalyzed by nickel catalysts for the growth of CNTs, but also promote the growth of CNTs from the degradation products with higher carbon numbers through hydride-transfer reactions [5]. In addition, very recently, our group has found that the combined catalysts of halogenated compounds and Ni_2O_3 can also catalyze carbonization of PP into CNTs [7,8]. In this case, the halogen

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