

Influence of the nature of chelating agents on the activity and structural pre-organization of metal oxide catalysts for the growth of carbon nanotubes synthesized by the method of polymerized complex precursors

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Annotation. Metal oxide catalysts (MOCs) for the growth of carbon nanotubes with the general composition $\text{Fe}_2\text{Co}/\text{CaCO}_3$ were synthesized using the polymerized complex precursor (PCP) method using chelating agents. It has been established that metal oxide catalysts synthesized by the method of polymerized complex precursors are an order of magnitude more effective than MOCs synthesized by the sol-gel technology; the performance of MOCs obtained using two ligands is significantly higher than the performance of monoligand samples. As a result of the study, it was revealed that the use, in addition to carbohydrates, of a wide range of chelating agents of various natures (polyalcohols, hydroxy acids, amino acids, etc.) in the synthesis of metal oxide catalysts for the growth of CNTs using the method of polymerized complex precursors is advisable and makes it possible to influence the stage of preorganization of the structure of the metal oxide precursor growth catalysts, specific productivity of MOCs and qualitative composition of carbon products.

Introduction. The main factors influencing the synthesis of CNTs by CVD are: the nature of the carbon precursor, the catalyst and the catalyst support. The use of the PCP method in the synthesis of MOCs, the use of chelating agents of different structures, makes it possible to obtain preorganized 3d-structures of the CNT growth catalyst with a hierarchical distribution of pore length and diameter. In this case, we are talking about catalysts with high porosity, a maximally developed surface of the support and ions of active metals evenly distributed on it. All other things being equal, the development of such technologies helps to increase the efficiency and selectivity of metal oxide catalysts for the growth of multi-walled carbon nanotubes (MWCNTs) for the CCVD process and is an urgent task.

Results and discussion. Metal oxide catalysts of general composition $\text{Fe}_2\text{Co}/\text{CaCO}_3$ with molar ratio Fe, Co / CaCO_3 equal to 0.12 : 1 were obtained by the PCP method according to the previously described method [1]. The following chelating agents were used: sucrose, fructose, glucose, ethylene glycol, diethylene glycol, glycerin, 1,2-propylene glycol, 1,4-butylene glycol, polyethylene glycol, polyvinyl alcohol, citric acid, glycine, pentaerythritol. MOCs-PCPs were also obtained using two chelating agents, one of which is citric acid. To compare the efficiency of the MOC-PCP sol-gel catalysts, a catalyst for the growth of CNTs of the general composition $\text{Fe}_2\text{Co}/\text{CaCO}_3$ with a molar ratio of Fe, Co / CaCO_3 equal to 0.12:1 was synthesized. CVD synthesis of MWCNTs was carried out in a tubular reactor at $T = 800^\circ\text{C}$ (carrier gas -Ar, 60 cm³/min; carbon precursor gas -commercial propane-butane mixture, 30 cm³/min) according to the previously described method [2].

Analysis of the data allows us to conclude that metal oxide catalysts with a lower bulk density (ρ_n) are more effective compared to metal oxide catalysts with a higher ρ_n . It was revealed that MOCs synthesized by the PCP method have a lower ρ_n compared to MOCs synthesized by the sol-gel method, and the use of a second chelating agent leads to the production of MOCs with low ρ_n , which increases the specific yield of CNTs. Thus, we can conclude that the synthesis of MOC-PCP using two chelating agents simultaneously allows one to vary the physical parameters of precatalysts - bulk density, porosity, dispersity, specific surface area - over a wide range. This approach will make it possible to obtain MWCNTs with specified morphological parameters.

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1. CCVD –синтез углеродных нанотрубок: влияние состава металлоксидных катализаторов / А. П. Прудченко, М.В. Савоськин, С.В. Хрипунов, [и др] // Донецкие чтения 2020: образование, наука, инновации, культура и вызовы современности: материалы V Международной конференции (Донецк, ноября 2020 г.). –Т.2. –Донецк: Дон НУ, 2020. –С. 75-78. 2 Прудченко А.П. Закономерности процесса темплатного синтеза пористых углеродных материалов методом CVD / А.П. Прудченко [и др] // Вестник Луганского национального университета имени Владимира Даля. 2018, №5(11), С. 299–303.

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