SYNTHESIS OF CARBON NANOTUBES AND NANO-ONIONS IN CH4/C2H4 COUNTERFLOW DIFFUSION FLAMES

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Carbon nanotubes (CNTs) and nano-onions (CNOs) were synthesized in counterflow diffusion flames. The conditions of synthesis and correlation of CNOs and CNTs were studied by methane-ethylene mixed fuel and variation of oxygen concentration. The variation of oxygen concentration affects the morphologies of synthesized products significantly with a threshold of 30% distinguishing the formation of CNO or CNT. CNOs formed at higher oxygen concentrations (30%, 40%, and 50%), and CNTs were synthesized only at lower oxygen concentrations (21% and 30%). The fuel composition has minor effects on the morphologies. When the oxygen concentration is 30%, both CNTs and CNOs can be synthesized. More carbon sources (additional fuel) are required for CNOs synthesis than for CNTs, but the temperatures are similar (1146-1164K for CNTs, 1096-1165K for CNOs). The absence of CNTs and CNOs were found at high ethylene concentration.

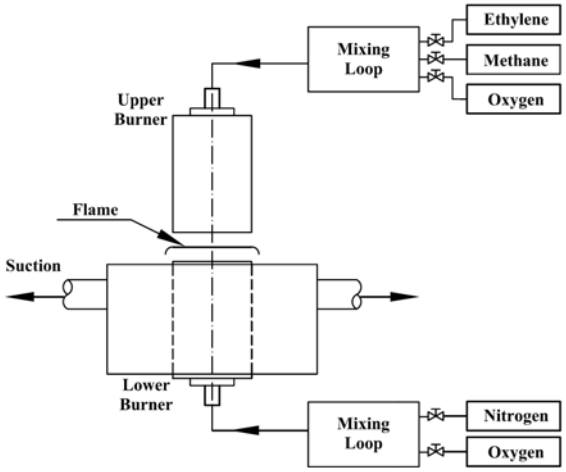

Fig. 1. Schematic of counterflow configuration.

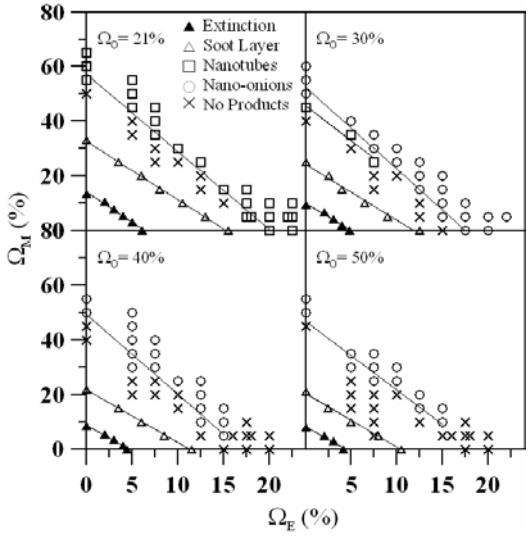

Fig. 2. Flame and synthesized products characteristics for different methane (Ω_M), ethylene (Ω_E) and oxygen (Ω_O) concentrations.

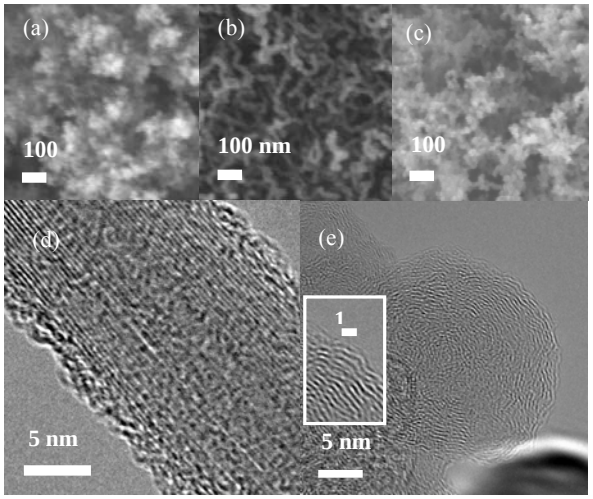

Fig. 3. SEM images of synthesized products (a) CNOs at [Ω_O, Ω_E, Ω_M] = [30%, 7.5%, 30%]; (b) CNTs at [Ω_O, Ω_E, Ω_M] = [30%, 7.5%, 25%]; (c) CNOs at [Ω_O, Ω_E, Ω_M] = [30%, 12.5%, 25%]. HR-TEM images of (d) CNTs; (e) CNOs.

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Abstract

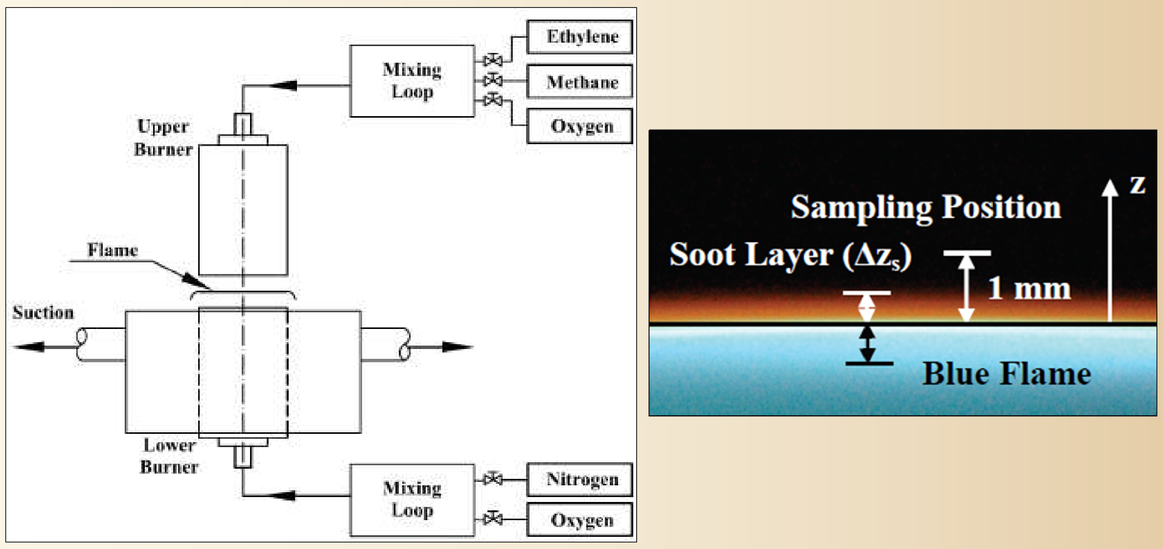
Carbon nanotubes (CNTs) and nano-onions (CNOs) were synthesized in counterflow diffusion flames. The conditions of synthesis and correlation of CNOs and CNTs were studied by methane-ethylene mixed fuel and variation of oxygen concentration. The variation of oxygen concentration affects the morphologies of synthesized products significantly with a threshold of 30% distinguishing the formation of CNO or CNT. CNOs formed at higher oxygen concentrations (30%, 40%, 50%), and CNTs were synthesized only at lower oxygen concentrations (21%, 30%). The fuel composition has minor effects on the morphologies. When the oxygen concentration is 30%, both CNTs and CNOs can be synthesized. More carbon sources (additional fuel) are required for CNOs synthesis than for CNTs, but the temperatures are similar (1146-1164K for CNTs, 1096-1165K for CNOs). The absence of CNTs and CNOs were found at high ethylene concentration.

Introduction

Flame has been applied in synthesizing products of a great variety of nano-structures. Comparing with other methods (e.g. CVD), flame synthesis is more economical due to simple equipments and low energy consumption. In contrast with CNT, studies of carbon nano-onions (CNOs) by flame synthesis were few. In our previous work, CNTs and a high yield of CNOs were synthesized using methane-ethylene mixed fuel. Though CNO and CNT were synthesized under similar flame conditions, the correlation or critical threshold was unclear. In this study, the correlation between CNO and CNT was investigated by examining the synthesized products from counterflow diffusion flames. Parameters of flame conditions included the mixed fuel (methane-ethylene concentrations) and oxygen concentrations. The objective of this study is to distinguish synthesis conditions of CNO and CNT, and to achieve high-yield production of CNOs.

Experimental

Synthesis of CNTs and CNOs were conducted by the counterflow system (Fig. 1a). Two identical burners with an inner diameter of 46mm were aligned vertically facing each other and the separation distance of burner exits was 22mm. A mixture of methane, ethylene and nitrogen was supplied to the upper burner; while a mixture of oxygen and nitrogen (the oxidizer) to the lower burner. The exit velocities of both burners were kept constant at 15 cm/s. The volume percentages of methane, ethylene and oxygen are denoted by Ω_M, Ω_E and Ω_O, respectively. Flame characteristics of mixed fuel for Ω_O = 21%, 30%, 40% and 50% were measured for comparison of synthesis characteristics. When the fuel concentration was sufficiently high, a soot layer formed above the blue flame on the fuel side (Fig. 1b). The thickness of soot layer is denoted by Δz_s. Z-axis is the axisymmetric axis which originates from the upper edge of the blue flame toward the upper burner. The sampling position was at z = 1 mm. Pure nickel grids were employed for collecting combustion products. The deposition time was 120 s. The gas temperature was determined by a silica-coated R-type thermocouple. Field emission scanning electron microscopy (FE-SEM), and high resolution transmission electron microscopy (HR-TEM) were employed to characterize the deposit materials.



(a) (b)

Fig. 1. (a) Experimental setup. (b) Flame structure and sampling position.

Conclusions

Synthesis conditions and the correlation of CNOs and CNTs were studied by counterflow diffusion flames with mixed fuel and constrained sampling conditions. Parameters of flames included methane-ethylene concentrations and oxygen concentrations. In our experiments, the variation of Ω_O affects the synthesis of CNTs significantly, and the synthesis of CNTs or CNOs dominates by Ω_O with a threshold of 30%. CNOs formed at higher Ω_O (30%, 40%, 50%), and CNTs were synthesized only at lower Ω_O (21%, 30%). The fuel composition has minor effects on the morphologies of synthesized products. At Ω_O = 30%, both CNTs and CNOs were synthesized, and the effects of fuel composition were examined. More carbon sources (Ω_E or Ω_M) are required for CNOs synthesis than for CNTs, but the temperatures required are close (1146-1164K for CNTs, 1096-1165K for CNOs) at Ω_O = 30%. The absence of CNTs and CNOs were found at high Ω_E, which may suggest suppressing effects on both CNTs and CNOs synthesis at high concentration of unsaturated hydrocarbons. But more studies of concentration of local species and their effects are required.

Results and Discussion

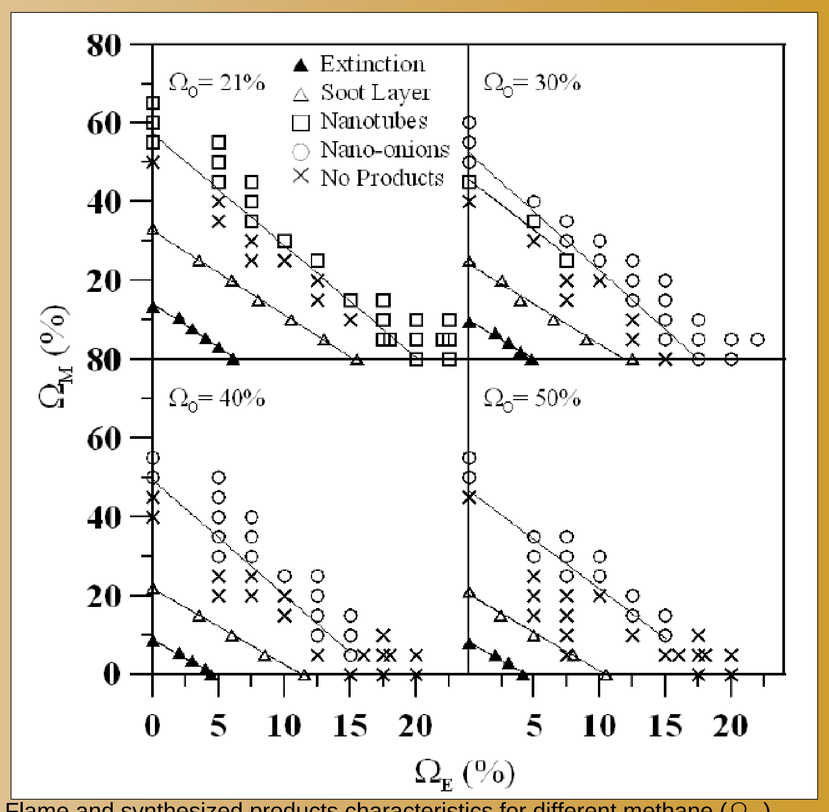
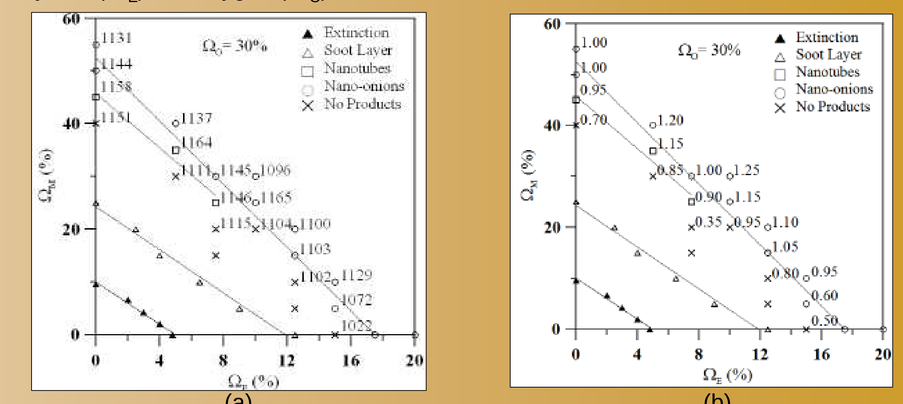


Fig. 2. Flame and synthesized products characteristics for different methane (Ω_M), ethylene (Ω_E) and oxygen (Ω_O) concentration.



(a) (b)

Fig. 3. (a) Temperature (in kelvin) and thickness of soot layer of sampling positions for different Ω_M and Ω_E at Ω_O = 30%.

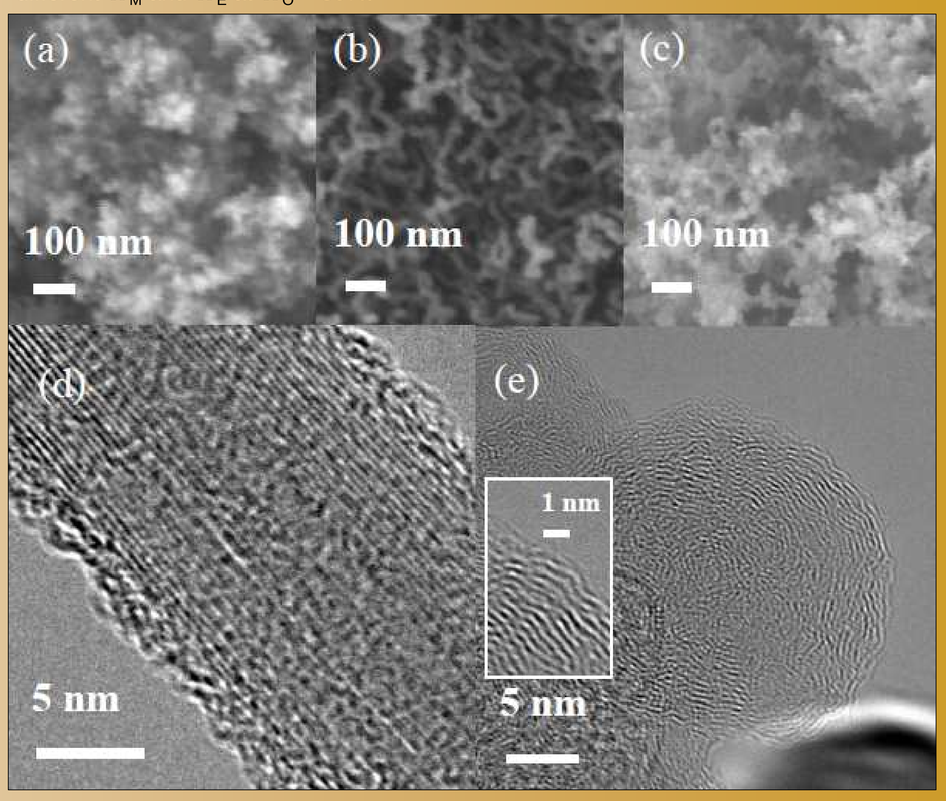


Fig. 4. SEM images of synthesized products (a) CNOs [Ω_O, Ω_E, Ω_M] = [30%, 7.5%, 30%]; (b) CNTs [Ω_O, Ω_E, Ω_M] = [30%, 7.5%, 25%]; (c) CNOs [Ω_O, Ω_E, Ω_M] = [30%, 12.5%, 25%]. HR-TEM images of (d) CNTs; (e) CNOs