SHOCK TUBE STUDIES OF SOOT FORMATION, STRUCTURE AND YIELD AT PYROLYSIS OF METHANE, ACETYLENE AND PROPANE

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The soot is one of the products formed due to pyrolysis of hydrocarbons. Shock tubes are frequently used for pyrolysis studies, since they permit synthesizing different substances from gas mixtures over a wide range of temperatures and pressures [1-3]. In addition, the use of shock tubes enables one to adopt a great variety of recording techniques in order to obtain accurate information on current concentrations of molecules and radicals that are formed during chemical reactions. There are many works, in which pyrolysis is considered along with the oxidation process [4,5]. The presence of oxygen exerts an essential influence on the kinetics of chemical reactions, at the same time shifting considered processes into a lower temperature region. In our case, the consideration of the pyrolysis without the oxygen participation makes it possible to investigate it only under pure thermal excitation conditions, which requires higher initial temperatures. The objective of the present work was to investigate the soot formation, structure and yield at the pyrolysis of methane, acetylene and propane within a wide range of post-shock temperatures.

The square shock tube of 4.5 m long has been applied for these studies (fig. 1). The gas emission behind the reflected shock wave was registered at two wavelengths corresponding to the radiation of the C_2 (λ = 517 nm, $\Delta\lambda$ = 5 nm) and CH radicals (λ = 430.8 nm, $\Delta\lambda$ = 2.5 nm). Emissions were detected by interference filters and recorded by photomultipliers. An attenuation signal of laser radiation was recorded by a photoresistor in combination with narrow-band interference filters (λ_1 = 632.2 nm, λ_2 = 632.8 nm, $\Delta\lambda_2$= 2.6 nm) and optical glasses (KC-10, KC-13) that used for cutting off a parasitic spectrum caused by gas self-emission at high temperatures. Since the measuring gas volumes behind the reflected shock wave in recording of the laser absorption and emissivity were equivalent (fig. 1), the moment of luminosity peak was chosen as a time for measurements of laser absorption coefficients. This criterion corresponded to the maximum of production of C_2 and CH radicals during pyrolysis. The concentration of condensed soot [C_{soot}] was calculated by means of Lambert-Beer's law.

Fig. 2 plots obtained temperature dependences for the induction time of soot condensation and yield at pyrolysis of the mixture C_3H_8/Argon (4 : 96) behind the reflected shock wave. It is seen that the temperature dependence for soot yield as in the case of acetylene [1, 2] has a typical "bell-shaped" form. However, a maximal soot yield in propane is in the higher temperature range 2400 – 2800 K, and unlike acetylene its profile is essentially broadened. Analysis of soot microphotos and electron photographs (fig. 2) for the most characteristic parts of the curve for soot yield has not revealed the presence of fullerens and nanotubes in sample materials. It is found that fine-dispersed graphite is the main component of the soot structure over the investigated range of pressures and temperatures.

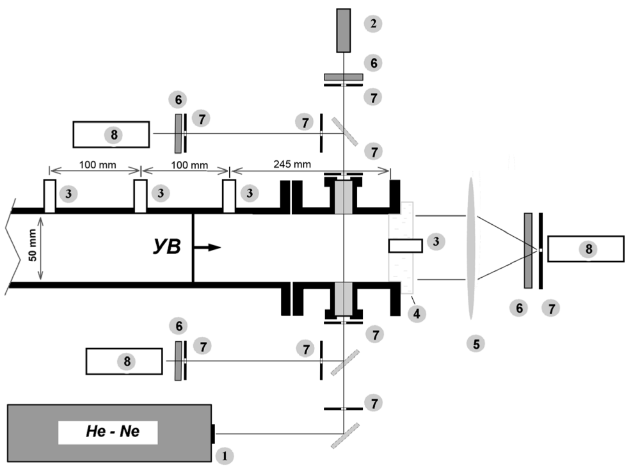
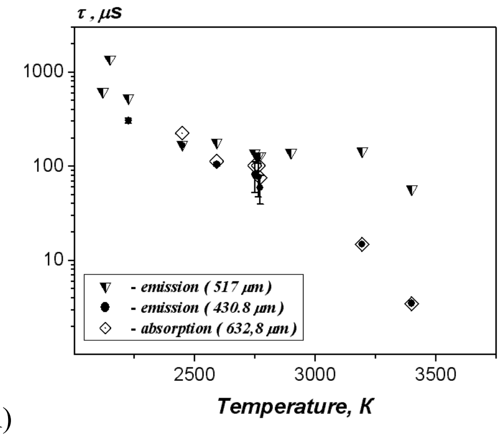
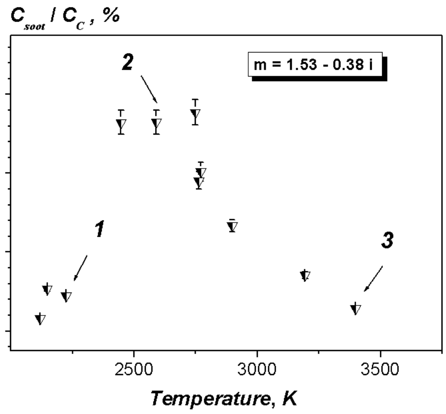


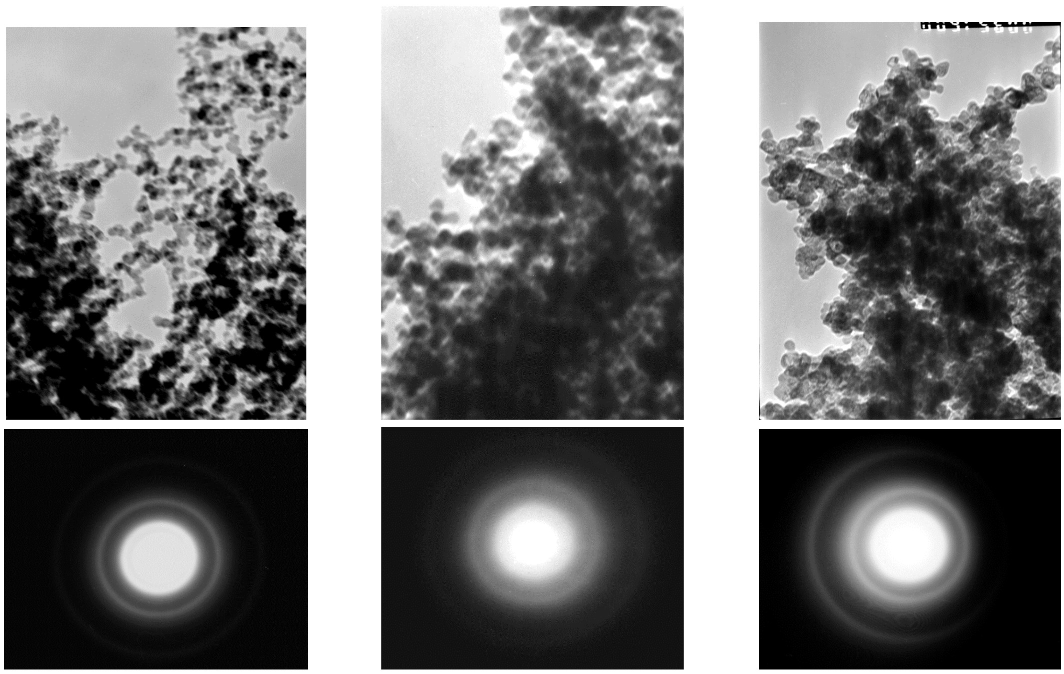
Fig. 1. The shock tube measuring section and the optical schematic of the setup for studying the pyrolysis behind the reflected shock wave. 1 – He-Ne laser; 2 – photoresistor; 3 – pressure sensors; 4 – reflecting end wall; 5 – lens; 6 – interference filter; 7 – diaphragm; 8 – photomultipliers



A)



B)



C) **1** **2** **3**

Fig. 2. Induction time (A) and soot yield (B) vs. gas temperature behind the reflected shock wave in C_3H_8/Argon (4:96) mixture and the microphotos and electron photographs of carbon materials over different sections of dependence (B) during pyrolysis: 1 – 2224 K; 2 – 2590 K; 3 – 3399 K

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