INVESTIGATION OF THE SYNGAS IGNITION BEHAVIOR IN REFLECTED SHOCK WAVES

P. A. Vlasov[1], V. N. Smirnov[1], O. B. Ryabikov[1], and A. R. Ahunyanov[2]

[1]N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences, 4 Kosygin Str., Moscow 119991, Russian Federation

[2]National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 31 Kashirskoe Sh., Moscow 115409, Russian Federation

Abstract: Experimental measurements of time profiles of the intensities of the absorption by ground-state hydroxyl radicals OH at $\lambda = 306.77$ nm, and the emission from electronically excited OH* ($A^2\Sigma^+ \to X^2\Pi$ transition, $\lambda = 310 \pm 4$ nm) were carried out. Based on these measurements, the ignition delay times were determined as the time interval between the time of arrival of the reflected shock wave and the time of reaching the maximum intensity of OH* emission. The corresponding temperature dependences of the ignition delay times were plotted for various H_2/CO/O_2/Ar mixtures. Detailed kinetic simulations of the profiles of electronically excited OH* radicals were performed and compared with the experimentally measured profiles to gain insights into the mechanism of the electronic excitation and quenching of these species. Pressure variability behind the front of the reflected shock wave was taken into account in the kinetic simulations. This made it possible to significantly improve the coincidence of calculated and experimentally measured ignition delays.

Keywords: kinetics of self-ignition of syngas; ignition delay time; chemiluminescent emission of hydroxyl radicals; numerical simulations; shock tube; reflected shock waves

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Figure Captions

Figure 1 A typical experimental record with a mixture of 1.5% H_2 + 1.5% CO + 1.5% O2 + 95.5% Ar, $\varphi = 1.0$, at $T_5 = 1380$ K and $p_5 = 1.2$ bar. Along the abscissa, time is plotted. One coarse division consisting of five smaller ones corresponds to a time interval of 200 μs. The total time from the beginning up to the end of the oscilloscope sweep corresponds to 2 ms. Along the ordinate, the following signals are plotted: *1* — pressure sensor signal (sensitivity 20 mV per division); *2* — absorption signal of OH radicals (sensitivity 200 mV per division); and *3* and *4* — chemiluminescence signals of electronically excited OH* radicals obtained at different sensitivities of the oscilloscope (100 and 50 mV per division, respectively)

Figure 2 Oscillograms of experiments with a mixture of 1.5% H2 + 1.5% CO + 1.5% O2 + 95.5% Ar, $\varphi = 1.0$, at different temperatures behind the reflected shock wave at a pressure close to atmospheric: (*a*) $T_5 = 1100$ K, $M_5 = 1.07 \cdot 10^{-5}$ mol/cm^3; (*b*) $T_5 = 1380$ K, $M_5 = 1.04 \cdot 10^{-5}$ mol/cm^3; and (*c*) $T_5 = 1610$ K, $M_5 = 1.14 \cdot 10^{-5}$ mol/cm^3; *1* — CO_2 radiation; *2* — OH adsorption; *3* — OH* radiation; and *4* — pressure

Figure 3 Intensity of chemiluminescent radiation of electronically-excited OH* radicals: *1* — signal of pressure sensor (P_5); *2* and *3* — signals of chemiluminescent radiation of OH* radicals obtained experimentally in [11] and as a result of the authors' kinetic simulation, respectively, using Aramco 1.3 mechanism [13] for a mixture of 0.30% H_2 + 5.62% CO + 2.96% O_2 + 91.12% Ar ($\varphi = 1.0$) for $T_5 = 1240$ K and $p_5 = 3.79$ bar

Figure 4 Calculated values of T_5 temperatures behind reflected shock waves and the concentrations of H_2, CO, and electron-excited OH* radicals for 0.30% H_2 + 5.62% CO + 2.96% O_2 + 91.12% Ar ($\varphi = 1.0$) and 0.30% H_2 + 2.96% O_2 + 96.74% Ar ($\varphi = 0.051$) mixtures at $T_5 = 1240$ K and $p_5 = 3.79$ bar. For a mixture containing CO, solid lines represent the results from [11] and dot-and-dash lines the results of the authors' kinetic simulations. For a mixture without CO, dashed lines represent the results from [11] and dotted lines the results of the authors' kinetic simulations

Figure 5 Calculated values of T_5 temperatures and concentrations of H_2, CO, and OH* for 2.96% H_2 + 2.96% CO + + 2.96% O_2 + 91.12% Ar ($\varphi = 1.0$) and 2.96% H_2 + 2.96% O_2 + 94.08% Ar ($\varphi = 0.5$) mixtures at $T_5 = 1240$ and $p_5 = 3.79$ bar. For a mixture containing CO, solid lines represent the results from [11] and dot-and-dash lines the results of the authors' kinetic simulations. For a mixture without CO, dashed lines represent the results from [11] and dotted lines the results of the authors' kinetic simulations

Figure 6 Temperature dependence of the ignition delay time and its linear approximation for $1.5\%\,H_2 + 1.5\%\,CO + 1.5\%\,O_2 + 95.5\%\,Ar$ ($\varphi = 1.0$) mixture obtained for different temperatures behind reflected shock waves at a pressure close to atmospheric

Figure 7 Temperature dependences of the ignition delay for $0.75\%\,H_2 + 0.75\%\,CO + 1.5\%\,O_2 + 97.0\%\,Ar$ (*a*), $1.0\%\,H_2 + 2.0\%\,CO + 3.0\%\,O_2 + 94.0\%\,Ar$ (*b*), and $2.0\%\,H_2 + 1.0\%\,CO + 3.0\%\,O_2 + 94.0\%\,Ar$ (*c*) mixtures with $\varphi = 0.5$ at a pressure close to atmospheric: *1* and *2* — experiments 1 and 2, respectively; and *3* and *4* — calculations according kinetic schemes [13] and [15], respectively

Figure 8 Experimentally measured and calculated temperature dependences of the ignition delay time for different mixtures: (*a*) $1.0\%\,H_2 + 2.0\%\,CO + 3.0\%\,O_2 + 94.0\%\,Ar$ ($\varphi = 0.5$); (*b*) $0.75\%\,H_2 + 0.75\%\,CO + 1.5\%\,O_2 + 97.0\%\,Ar$ ($\varphi = 0.5$); (*c*) $2.0\%\,H_2 + 1.0\%\,CO + 3.0\%\,O_2 + 94.0\%\,Ar$ ($\varphi = 0.5$); and (*d*) $3.0\%\,H_2 + 3.0\%\,CO + 1.5\%\,O_2 + 92.5\%\,Ar$ ($\varphi = 2.0$); *1* — results of the authors' experiments behind reflected shock waves; *2* — results of detailed kinetic simulations for mechanism [13] at constant pressure behind reflected shock waves; *3* — results of kinetic simulations for the same mechanism [13] obtained for the experimentally measured pressure profile $p_5(t)$ behind reflected shock waves, i. e., for variable pressure behind a reflected shock wave; and *4* and *5* — linear approximations

Figure 9 Time histories of the normalized amplitude of the concentration of electronically excited OH^* radicals for a mixture of $1.5\%\,H_2 + 1.5\%\,CO + 3\%\,O_2 + 97.0\%\,Ar$, $\varphi = 0.5$, obtained at different initial temperatures behind reflected shock waves for a pressure close to atmospheric: (*a*) $T_5 = 955$ K, $p_5 = 0.75$ bar; (*b*) $T_5 = 985$ K, $p_5 = 0.80$ bar; (*c*) $T_5 = 1157$ K, $p_5 = 0.92$ bar; (*d*) $T_5 = 1284$ K, $p_5 = 1.16$ bar; (*e*) $T_5 = 1327$ K, $p_5 = 1.17$ bar; (*f*) $T_5 = 1684$ K, $p_5 = 1.64$ bar; *1* — authors' calculations; *2* — calculation at $P_5 = const$; and *3* — calculation $p_t(t)$

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Contributors

Vlasov Pavel A. (b. 1955) — Doctor of Science in physics and mathematics, leading research scientist, N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences, 4 Kosygin Str., Moscow 119991, Russian Federation; iz@chph.ras.ru

Smirnov Vladimir N. (b. 1950) — Doctor of Science in physics and mathematics, chief research scientist, N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences, 4 Kosygin Str., Moscow 119991, Russian Federation; vns1951@yandex.ru

Ryabikov Oleg B. (b. 1943) — Candidate of Science in physics and mathematics, research scientist, N. N. Semenov Federal Research Center for Chemical Physics of the Russian Academy of Sciences, 4 Kosygin Str., Moscow 119991, Russian Federation; zaslonko@chph.ras.ru

Akhunyanov Artur R. (b. 1994) — magister, National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 31 Kashirskoe Sh., Moscow 115409, Russian Federation; iz@chph.ras.ru