velocity of the liquid; f, repetition frequency of the drops; λ, wavelength; S_{sp}, surface area of a drop; ℓ_1, distance from the opposite spherical electrode to the edge of the jet; q_{lim}, limiting charge of a spherical particle; L^*_j, length of the undecomposed part of the charged jet; S, distance from the location of outflow to the axis of the needle–sphere system; and D, diameter of the jet.

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INVESTIGATION OF THE KINETICS OF SELF-PROPAGATING SYNTHESIS BY THE METHODS OF COMPUTATIONAL IR THERMOGRAPHY

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The methods, techniques, and results of investigations of the kinetics of self-propagating synthesis of organic reagents performed in order to improve self-propagating high-temperature synthesis (SHS) are described. Data on the structure of the combustion wave are presented; the data were obtained using a system for computational IR thermography, IR spectroscopy of scattering media, contact thermometry and microscopy, and thermogravimetric analysis, all of which made it possible to obtain information about the physical and chemical processes occurring in a wave of SHS.

In the last few years methods for synthesizing refractory compounds and materials based on oxygen-free combustion – self-propagating high-temperature synthesis (SHS) – have been widely used. Wave regimes in which the chemical transformation is concentrated in a zone propagating along a mixture of powders have been obtained with the help of local initiation [1]. The characteristic features of the process are that virtually no gas is released, the reaction product is in the condensed phase, the temperature in the reaction zone is high, the energy consumption is low, the equipment is simple, and the product is pure. In SHS processes surface and layerwise combustion, wave localization, splitting and reflection of waves, self-excited and spin combustion, and wave stratification of the medium are realized.

It is of greatest interest to clarify the mechanism of SHS processes, to model SHS numerically, and to develop efficient technologies. The solution of this problem will make it possible to realize the direct synthesis of multicomponent solid solutions of high purity and the synthesis of metastable phases. It is difficult to study directly the chemical and physical processes occurring in a combustion wave and primarily at the combustion front in classical SHS of systems of the type metal-oxidizer. The short reaction time in the wave

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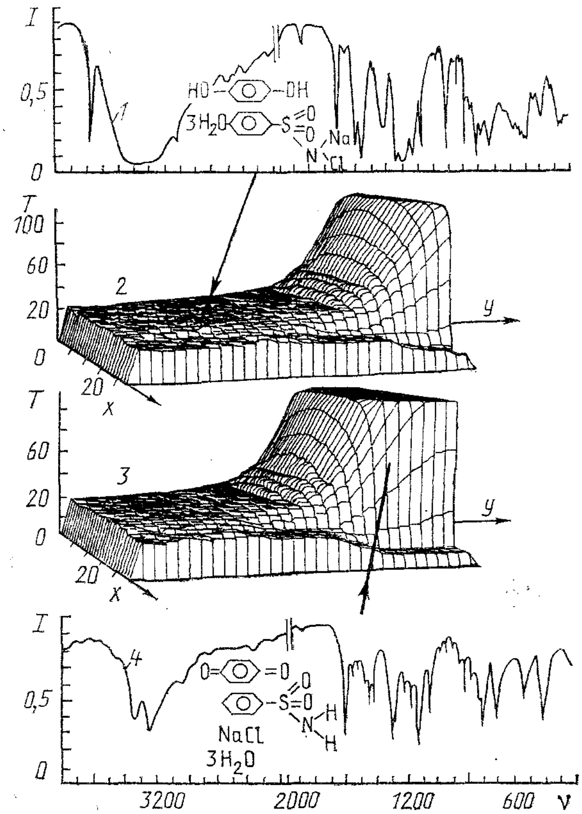

Fig. 1. Dynamics of the propagation of a temperature wave in the process of self-propagating synthesis (the time interval between the measurements is equal to 5 sec) and the IR transmission spectra of the mixture before and after the reaction (2, 3 and 1, 4, respectively). I, arbitrary units; T, °C; x, y, mm; and ν, cm^{-1}.

and the high rates of heating (0.05 sec and $1 \cdot 10^6$ K/sec, respectively) make it necessary to use laborious x-ray-spectral methods for analyzing thin sections of extinguished samples. An alternative method is to study the fronts of chemical reactions, having characteristic temperatures of 300...500 K and initiated in a mixture of powders, as analogs of the SHS processes.

One problem at the stage of experimental studies of the kinetics of self-propagating synthesis is to develop, based on progress in IR spectrophotometry, IR thermometry, and computational IR thermography [2], methods of diagnostics that yield the maximum possible amount of information. Classical methods, such as thermogravimetry, calorimetry, contact thermometry, and chemical analysis, have significant possibilities.

To develop measurement methods we selected reactions occurring at temperature ranging from 200 to 300°C with relatively slow velocities of the reaction front in a mixture of powdered reagents with the simplest chemical structure. Examples of such systems are mixtures of hydroquinone and chloramine B, and a number of other mixtures developed at the Institute of Chemical Physics of the Academy of Sciences of the Armenian SSR.

In the investigations performed the synthesis reaction in a medium consisting of hydroquinone and chloramine B was initiated by a heat source.

Both substances $C_6H_4(OH)_2$ and $C_6H_5SO_2NNaCl \cdot 3H_2O$ are colorless crystals (they melt at 170 and 180...185°C, respectively) and they form in the course of the reaction an opaque mass, consisting of a mixture of quinhydrone, benzenesulfamide, and sodium chloride.

To study heat transfer processes signals from two chromel-copel thermocouples, placed in a vertical quartz cell 10 mm from one another in the direction of propagation of the reaction wave, were recorded in the course of the reaction. Infrared images of the reagents in the cell in the range 2...6 μm were recorded at the same time by the computational IR

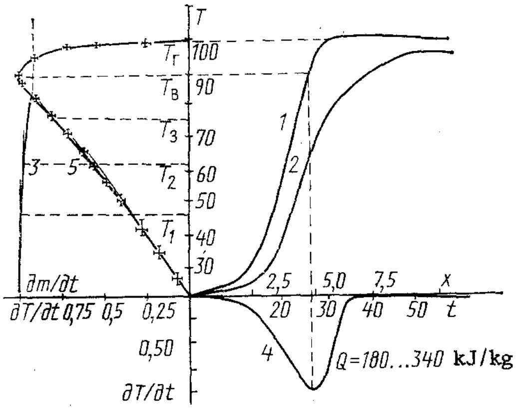

Fig. 2. Dynamics of the temperature change in the reaction wave based on data from optical (1) and contact (2) measurements, the change in mass (3) of the mixture on heating, and the change in the heat flux as a function of the time (4) and temperature (5). t, sec.

thermography system, and a visible image of the reagents was recorded with a television camera on a VCR through a microscope with a magnification of ×98. The experiment was controlled with an Élektronika MS1201 microcomputer and apparatus in the CAMAC standard, connected with an SM-4 central minicomputer. The system also included an IKS-29 IR spectrophotometer and a number of other devices.

Figure 1 shows the dynamics of the temperature change in the reaction zone, recorded by the computational IR thermography system, and IR absorption spectra of the mixture before and after the reaction. The difference in the IR spectra makes it possible to identify the start of the reaction by recording radiation passing from an IR source through the cell with the reagents. It should be noted that the experimental results are highly reproducible. The velocity of propagation of the wavefront remains virtually constant as the compaction density of the powder is varied (from 0 to 20 kg/cm^2) and equal to 0.4...0.18 mm/sec.

It is of interest to study the structure of the combustion wave, since the chemical and physical processes occur in an extended reaction zone. Using for these purposes contact thermometry based on the characteristic IR radiation of the bodies makes it possible to obtain the characteristic temperature profiles, which depend on the presence of phase transformations in the zone of the combustion wave. In the absence of phase transformations the temperature increases smoothly and then decreases smoothly. Analysis of the isothermal sections and extrema in complicated profiles makes it possible to identify phase transition processes occurring in the wave.

Differentiating the temperature curve and then constructing a plot in the coordinates heat flux — temperature makes it possible to determine the temperature of "inflammation," separating the wave into a heating zone and a reaction zone. In the heating zone the dependence of the heat flux on the temperature is linear (the so-called Michelson straight line). The main characteristics of the wave of SHS are the width of the Michelson zone, the total wavelength (the sum of the length of the heating zone and the reaction zone), the reaction time in the wave (t_w), the heating time of matter in the wave (t_h), and the rate of heating of the matter. These parameters were determined numerically in the course of the experiment from the results of contact measurements with the help of microthermocouples or from the results of thermal-vision measurements (Fig. 2). Analysis of the data indicates that there is no endothermal peak when the ratio of the starting materials in the mixture is close to 1:1. The use of contact-free IR diagnostics increases the reliability of measurements, employing a sequence of IR images recorded in a digital form in a format of 64 × 64 pixels or a sequence of separate lines of the image. On the other hand, if the reaction process in the wave is stationary, all of the required information can be obtained based on analysis of one IR image, and this reduces to 62 msec the time required for the study.

The results obtained are indirectly confirmed by data from thermogravimetric analysis, where when the mixture is heated up to 60° part of the mass is lost without any heat being released, and at 75-78° an exothermal reaction is initiated. Analysis of the series of TV images shows that the wavefront does not have a distinct boundary and that "streamers" are present, probably owing to local nonuniformities of the mixture. This can explain the observed profile of the change in temperature.

The method developed, the software, and the apparatus for the experiment in which visible and IR images are simultaneously recorded and analyzed thus make it possible to study the characteristics of physical and chemical processes occurring in a wave of self-propagating synthesis. Additional information can be obtained in further studies using spectral methods for analyzing a spatial zone up to 100 μm wide in which the reaction front may be regarded as uniform.

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