Temperature hysteresis in the unstable combustion mode of SHS: experiment with high-speed micro-pyrometry

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Abstract. The aim of this study is to report a new experimental regularity of the change in the propagation velocity of SHS as a function of the adiabatic temperature at the local point of the combustion front. The paper presents the results of measuring the temperature and velocity in the propagation of a combustion wave, obtained using a special television micro-pyrometer (1200×800 pixels) with high spatial (5.85 μm/pixel) and resolution time (1000 fps). High accuracy of temperature measurement was provided by using a new method of spectrally-bright pyrometry (patent RUS 2616937) from 800 to 2000°C with an error of less than 1%. This paper shows the experimental procedure and statistical data on the hysteretic dependence of velocity on temperature.

1. Introduction
The theoretical possibility of loss of diffusion-thermal stability of laminar combustion was studied in [1]. The main result is that if the diffusion coefficient of fuel D is greater than the thermal diffusivity of mixture α, then for D>α the flame proves to be unstable.

The phenomenon of self-propagating high-temperature synthesis of materials (SHS), discovered in 1967 by Academician A.G. Merzhanov is the process of gasless combustion of a solid powder localized in a narrow reaction zone and propagating at the same speed throughout the initial volume of products [2]. Traditionally, SHS was considered as an autowave process in a quasihomogeneous medium with heat sources, described by a system of heat conduction and reaction diffusion equations [3]. Modern ideas about such a model contradict the experimental data on the discrete nature of SHS [4], but can be explained by the hysteresis dependence of the reaction rate on temperature [5]. For the first time with the development and application of new methods of high-speed micro-pyrometry [6-8], and then special electron-optical complexes with synchronous scanning of streak-cameras with nanosecond time resolution [9-11]. Particular attention is paid to recording the velocity and temperature in local foci of discrete combustion, which makes it possible to distinguish between the quasihomogeneous and microheterogeneous regimes of such processes [12-15]. It should be noted that the nonlinear hysteresis effect of diffusion combustion, explaining the effect of "heat localization" and the appearance of metastable thermal structures of finite, so-called "fundamental" thickness, is now little studied and therefore rarely used in theoretical models of discrete combustion of SHS.

Aim of the work: it was previously believed that the effect of heat localization is not feasible in solids due to a small change in the thermal conductivity and the linearity of the internal heat sources of the burning medium. As we showed earlier [16], in the SHS processes considered by us this restriction
does not exist. The purpose of this study is to establish the experimental dependences of the local combustion wave velocity of SHS on the local adiabatic temperature of the discrete focus.

2. Experiment and Equipment
To record the high-speed propagation of the SHS combustion wave, an optoelectronic micro-pyrometric complex was used [10], based on the nanosecond video resolution system "Video Sprint NanoGate" (Videoscan, Russia) with the image analysis and image processing program Fiji-ImageJ (NIH, USA) [11]. At the combustion of reacting mixture, we observed a rapid propagating of high temperature along the tablet after its ignition. Figure 1 shows an example of recording successive frames of high-speed (1000 frames per second) shooting of the combustion wave.

![Figure 1](image-url)  
**Figure 1.** Example of recording a combustion wave: serial frames of the solid state burning process; a) chronogram of the combustion wave velocity; b) thermogram of burning.

Figure 1a demonstrates serial frames of records of the solid state burning process. The basic stages of the process can be observed (figure1b). The warming up of the lowest cold layer is presented in zone 1. Here we do not observe any chemical reactions. In the zone 2 the rapid ignition and exothermic combustion reaction as local thermal explosion take place. The effective thermal width XT varies from 0.15 to 1 mm. However the necessary structural and phase changes of the crystal lattice still do not have time to occur. Zone 3 presents the disintegration process of high-temperature site to the small ones due to internal heat outflow. New sites form a vast "heat cloud", where the temperature approaches the adiabatic value and promotes complete burn-out of the initial products. The next stage is presented in the zone 4. The next step is presented in the zone 4, where the heat sink reduces the temperature of the final product and the diffusion slowly sets the required stoichiometric proportion. The micropyrometry measurements are presented in figure1b as the 1-D scanning thermal chronoscope along the dashed line of the heat monitoring photo-matrix. The thermal emission time in
the zone 2, the time of heat induction in the zone 3 and the constant of the heat outflow time in the zone 4 can be measured.

3. Results and Discussions
As a result of statistical processing of all chronograms - in our case, their number is equal to the number of lines of the photodetector matrix ($N_Y = 1200$), we can obtain the density of distribution of points along the line of the wave front in the temperatures shown in figure 2.

![Figure 2. Temperature distribution in the SHS wave.](image1)

![Figure 3. Distribution of speed chronograms.](image2)

With the velocity distribution, the situation is much more complicated, because speed is the result of an indirect measurement and in our case it was decided to take the average velocity for each chronoscopy line, i.e. averaged slope of the chronogram in figure 3.

As a result of the analysis of all scan lines, the distribution shown in figure 3 was obtained.

An analysis of the correlation dependence of velocity on temperature was carried out by constructing a phase space ${\{V_x, T\}}$ mapping the points $[V_x(i), T(i)]$ for all $i$ from 1 to $N_Y = 1200$, as shown in figure 4.

![Figure 4. The dependence of the combustion wave velocity of SHS on the temperature of the wave front.](image3)
Obviously, this dependence has a nonlinear character and a pronounced hysteresis. This behavior of the wave, as is known [1, 3, 4], can be explained by a number of physical mechanisms that require additional study. The form of hysteresis, obtained experimentally, clearly indicates the competition of thermal diffusivity (\( \alpha \)) and diffusion (\( D \)).

4. Conclusions
1. An appreciable manifestation of bimodality in the distribution of the temperature and velocity of the combustion wave was experimentally established with an increase in the proportion of the inert additive.
2. There are two main components in the temperature distribution. The first is described by the normal Gaussian distribution of the regime \( T=1950^\circ C \) and the mean square spread of 65-70°C, which corresponds to the afterburner region, where the thermal effect of SHS is absorbed by the internal heat removal to the endothermic reaction. The second one has an anomalous probability density (4 times higher than the normal distribution mode) at \( T_{ad}=2075^\circ C \) and corresponds to the localization of heat in micro-scopes (~10 \( \mu \)m) of local thermal explosions.
3. Temperature-velocity correlation has two stationary values of speed: "fast" – 2.7 mm/s, with a predominance of diffusion over thermal conductivity and "slow" – 2.4 mm/s, where thermal conductivity predominates.

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