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To cite this article: A V Dolmatov 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **558** 012009

IOP Conf. Series: Materials Science and Engineering **558** (2019) 012009 doi:10.1088/1757-899X/558/1/012009

Chrono-topographical analysis of the laws of the microheterogeneous combustion mode for the production of powders by the SHS method

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Abstract. The paper presents the capabilities of the chrono-topographical analysis method for studying the discrete nature of the propagation of the SHS process on the scale of the combustion focy and the reaction cell. Using the approach of chrono-topographic analysis, thermal imaging studies of the microheterogeneous mode of combustion in the Ni-Al system were carried out. The regularities of the dynamics of individual foci in the SHS wave are revealed. Using chronographic maps of the SHS process, it is shown that there are two types of foci in the combustion wave, which differ in the nature of the propagation dynamics. On the scale of the reaction cells, the experiment investigated the relationship of the combustion dynamics with the moments of the formation of new reaction sites. According to the results, it was concluded that an increase in the heat deficit in microheterogeneous combustion regimes leads to an increase in the dispersion of the resulting product. The work was supported by the Russian Foundation for Basic Research in research projects no. 18-08-01475, 18-08-01152, as well as grant no. 13-01-20/28 "Leading Scientific School of Ugra State University".

1. Introduction

The propagation of a SHS wave in microheterogeneous modes close to unstable combustion is characterized by the presence of areas with significant depression reactions, which contribute to the separation and even granulation of the products of synthesis [1].The use of this phenomenon for the production of powders with a given dispersion determines the relevance of the study of the laws of the microheterogeneous combustion mode.

The historical development of methods for analyzing experimental data on the SHS process is associated with the study of the macrokinetics of this phenomenon [2]. Studies of the discreteness of the propagation of a combustion wave made it possible to explain the appearance of layered structures in the products of synthesis [1–3]. However, to study the thermodynamics of microheterogeneous processes, registration of high-resolution SHS is required when the inhomogeneity of the combustion wave front becomes comparable to the characteristic size of the field of observation [4]. In this case, there is an uncertainty in the choice of the direction of the measuring cross-section in the images of thermal imaging, which requires the development of new approaches to the analysis of the recorded data. The authors of the article previously proposed an original method of chrono-topographical representation of the results of thermal imaging studies for joint analysis of microheterogeneous processes and macrokinetics of the phenomenon [5].

The purpose of this work was to detect patterns in the thermodynamics of reaction foci in a SHS wave using chrono-topographic analysis (CTA) of high-resolution thermal imaging data.

2. Principles of chrono-topographic analysis

Modern models of the stationary combustion mode link two factors in the equation of state — the depth of chemical transformation of the reaction cells and their temperature [6]. Consequently, a given temperature level can serve as a criterion for determining at the stage of ignition cells with the same depth of response. Moreover, the corresponding elements of the thermal image of the reaction cells, the signal level of which for the first time reached the set value, should be the wave front of the SHS with the appropriate depth of chemical transformation. By determining the coordinates of the front elements in all frames of a thermal imaging survey, you can construct a chronographic map that reflects the space-time distribution of the reaction cells with the same chemical conversion depth in the combustion wave front.

The chronographic map in figure 1 is based on high-resolution thermal imaging: 5.8 µm and 1 ms (figure 2). Her analysis reveals the following properties.

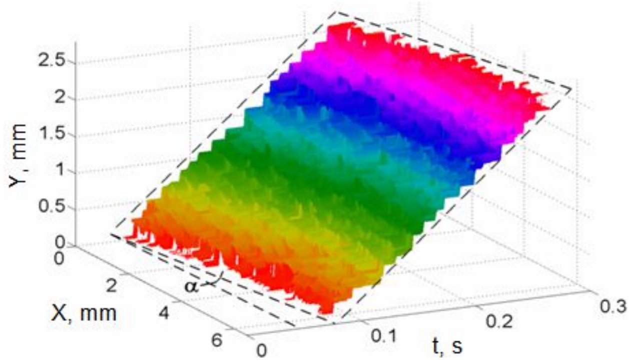

Figure 1. Integral chronographic map of the SHS process.

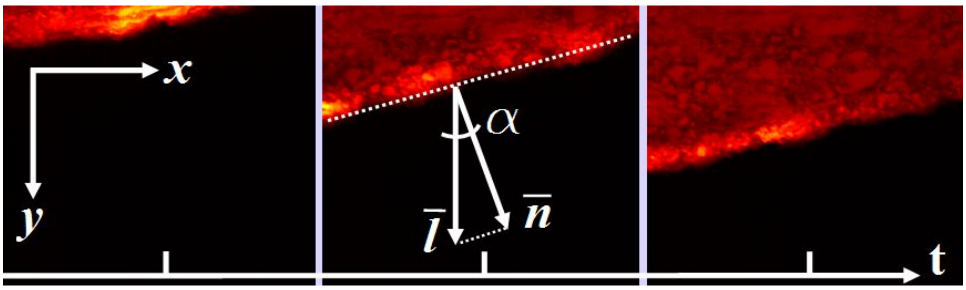

Figure 2. The coordinate system of thermal imaging shooting SHS process.

The experimental data on the position of the SHS wave front $y = f(x,t)$ is very well modeled by the plane. In relation to the CBC process, this means that the estimates of the mathematical expectation of the front speed are equal both at different points in time and at different points in space. From this fact, it follows that the SHS process in the wave mode can be considered not only stationary but also ergodic. Thus, when determining the statistical characteristics of the SHS process, data from different cross-sections of the chronographic map can be combined into a common sample. For modern high-speed cameras, this allows a 3–4 order increase in the sample size and a 1–2 order reduction in the confidence interval of the estimated parameters of the macrokinetics of the process.

Taking into account the ergodicity of the process, it can be argued that the angle between the direction of the columns of the photomatrix $\vec{l}$ and the normal to the front of the combustion wave $\vec{n}$ (figure 2) is equal to the angle α shown in figure 1. If the camera is oriented incorrectly during the

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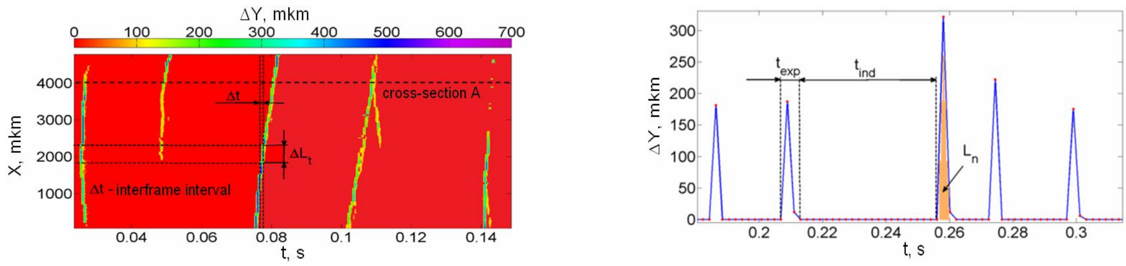
registration of the SHS process, the burning front will appear in one of the corners of the thermal image earlier than in the other. With a constant front propagation velocity, the value of the time delay will depend linearly on the distance between cross-sections along the axis *X*, and the direction of the normal to the front of the combustion wave can be determined from the average time delay of the occurrence of foci in different cross-sections of thermal imaging images, even in the case of large magnifications of the optical system. Chronographic maps of the SHS process allow us to determine the direction of the normal to the front of the combustion wave, combining two methods: analysis of the spatial data of thermal images; control of the delay time of appearance of the reaction front in different normal cross-sections.

The differential form of the chronographic map (figure 3) makes it possible to identify individual foci in the reaction wave, to study their thermodynamics and interaction. A joint analysis of thermal imaging data and a differential chronographic map allows us to construct a topographic map of the SHS process, which displays the areas where individual combustion centers developed during the whole process of registration (figure 4). The topographic map makes it possible to assess how the combustion depression during the SHS process contributes to the structure of the final product, and to determine the characteristic dimensions of the sample areas with a high depth of response of the initial components. The method of measuring the statistical parameters of the SHS process using chronographic and topographic maps is presented in [7].



(a) (b)

Figure 3. Differential form (a) and cross section A (b) of the chronographic map of the SHS process (V_t is the velocity of the source propagation along the SHS wave front; t_{exp} is the lifetime of a separate combustion center; t_{ind} is the induction time in the measuring section of a new combustion center; and L_n is the characteristic size of the combustion foci in the direction normal to the reaction front).

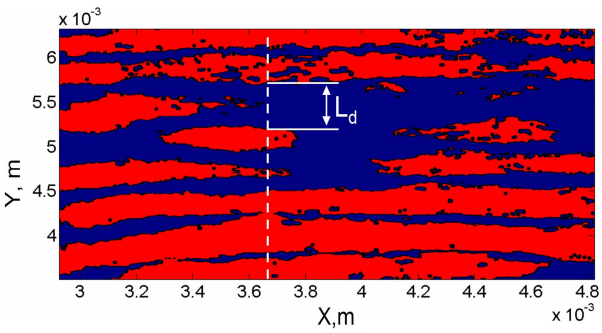


Figure 4. Topographic map of the SHS process (L_d is size of zone of the combustion depression between the foci).

Experimental estimation of the error of measuring the velocity of propagation of a SHS wave by the CTA method using high-resolution thermal imaging data with a sensor size of 1 MPixels was 0.05%. With the use of sensors of higher dimensionality or the approach of tracking thermal imaging control, an error level of $5 \cdot 10^{-4}$% can be achieved.

3. Thermal imaging studies

3.1. Development of foci of combustion in the SHS wave

In the present work, an experimental study of the evolution of individual foci in a SHS wave was carried out on the basis of the Ni–Al system. To control the thermal effect of the reaction in the charge, the mass fraction of Ni was varied from Ni_3Al to NiAl stoichiometry with a step of 0.5%. The density of the mixture at room temperature was chosen so that combustion with the formation of Ni_3Al was critical. The high-temperature synthesis was controlled by the original thermal imaging system with a spatial and temporal resolution of 5.8 µm and 1 ms, respectively [7].

The temperature images of figure 5 show the development of a burning center along the SHS wave front — the X-axis, while the front of the Ni_3Al synthesis reaction itself moved in the Y-axis direction. The size of the observation area in X was 7.3 mm; in Y, 1.87 mm.

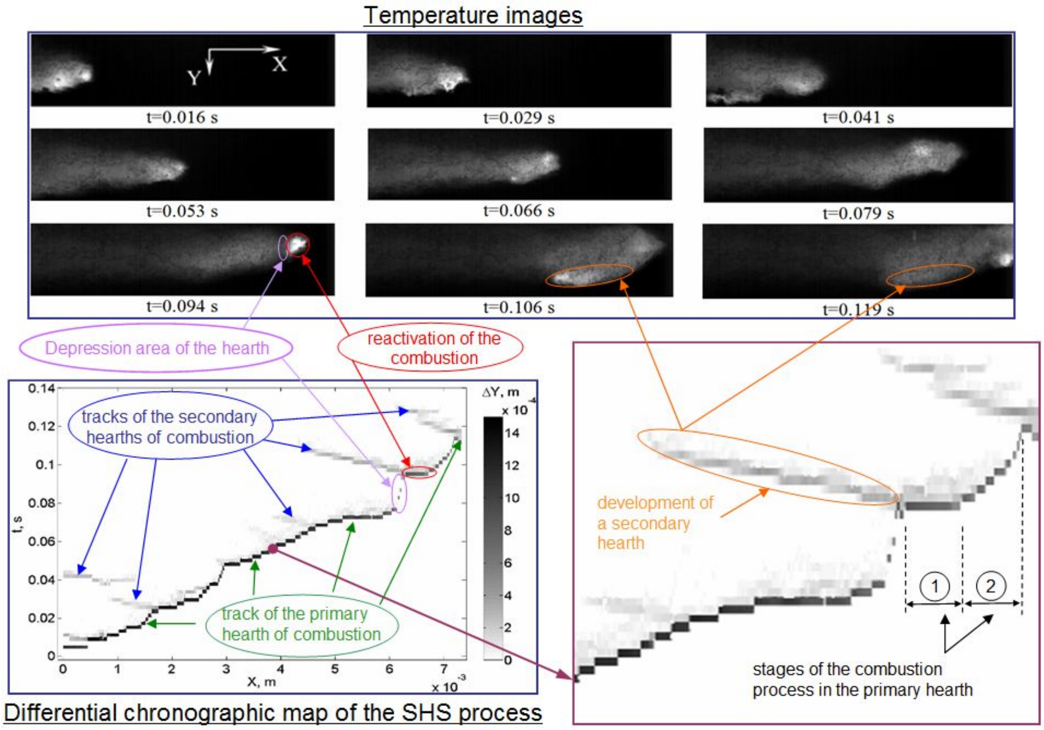


Figure 5. Development of combustion foci along the Ni_3Al SHS wave front.

The differential chronographic map in figure 5 clearly shows that the development of foci along the wave front is similar to the propagation of the reaction front itself - there is a stage of rapid displacement of the boundary of the center in the direction of the X axis with a sharp rise in temperature (flash stage), and a depression stage of the reaction with decreasing temperature. As an example, in figure 5 one of the areas of rapid displacement of the boundary of the source along the SHS wave front and one of the areas of depression of the combustion reaction in the focy are underlined.

A chronographic map of the SHS process also shows that the transition from the depression stage of the reaction to the flash stage is accompanied by the appearance of a secondary focus, which spreads along the wave front of the SHS, but in the opposite direction of the primary focy. An example of the emergence and development of a secondary focus, reflected on the chronographic map, is associated with micro thermal imaging shots that were recorded at time points: 0.094, 0.106 and 0.119 s (figure 5).

In general, based on the analysis of the differential chronographic map of the SHS process, several conclusions can be drawn. First, the velocity of propagation of the boundary of the foci along the wave front of the SHS is finite and can be measured by high-resolution thermal imaging with a small thermal effect of the reaction, which is close to critical. With an increase in the thermal effect, the velocity of propagation of foci along the front increases, but it becomes very difficult to identify the movement of the boundary of the focy. Increasing the temporal resolution of the thermal imaging camera leads to a decrease in the level of the detected signal, up to its loss against the background noise. With a small time resolution, the boundary shifts so quickly that it is not possible to register the phases of this movement in a combustion focy. Reducing the spatial resolution of a thermal imaging system contributes to an increase in the signal level, but leads to a “loss” of the source due to the discreteness of the matrix photo sensor. Secondly, there are at least two types of foci with different propagation dynamics in the front of the SHS wave. In the development of primary foci, two stages can be noted: coherent combustion (flash region) and inhibition of reaction propagation (figure 5). At the stage of inhibition, the dependence of the coordinate of the boundary of the primary burning focy of X_{bp} on time is similar to a logarithmic law. The coordinate of the boundary of the secondary foci of X_{bs}, up to the time of the termination of combustion, linearly depends on time. Thirdly, the flashes observed in the thermal imaging of the SHS wave are most likely not separate foci of combustion, but represent only the development stage of the primary focus, which, like the SHS wave front, also has a discrete propagation pattern. Actually, the developing primary foci of combustion form the wave front of the reaction. Fourth, secondary foci, spreading in the opposite direction in relation to the primary, burn out areas that become an obstacle to the development of the primary focus. This barrier can limit the area of distribution of the primary focus and lead to the fragmentation of the combustion wave front into separate sectors. This hypothesis is confirmed by a topographic map of the SHS process (figure 4). Thus, each product layer can be formed by several primary foci simultaneously.

With a small thermal effect of the reaction in the Ni–Al system, almost all secondary foci go out. With an increase in the magnitude of the thermal effect, there were cases when the secondary focus turned into the primary, i.e. the direction of propagation of the primary and secondary foci was reversed.

3.2. Dynamics of combustion in the reaction cells

The CTA method has been used to study the effect of combustion sources located in the SHS wave front on the thermodynamics of the reaction cells entering the previously formed combustion centers. For this purpose, in the analyzed section X_0, the dynamics of the maximum temperature of individual foci identified in the thermal imaging frames (figure 6) were monitored. Typical results of these studies are presented in figure 7.

It has been established that in modes close to unstable combustion, the thermograms of the reaction cells have several characteristic peaks. Moreover, the occurrence of each peak is very well synchronized in time with the moments of the appearance of new foci of combustion in the X_0 section. Based on these data, it can be assumed that the growth of the product layer between the fuel and oxidizer particles does not occur continuously as a result of reaction diffusion, but is discrete, i.e. the process of diffusion and mixing of the initial components is separated from the combustion process, which is experiencing stages of depression and flash.

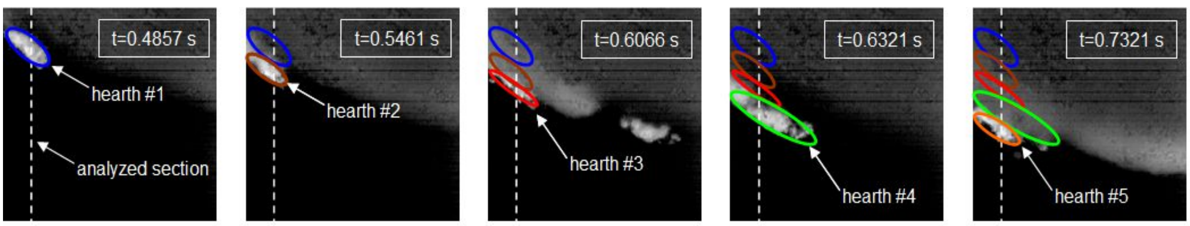


Figure 6. The principle of analysis of the thermodynamics of the reaction cells in temperature images.

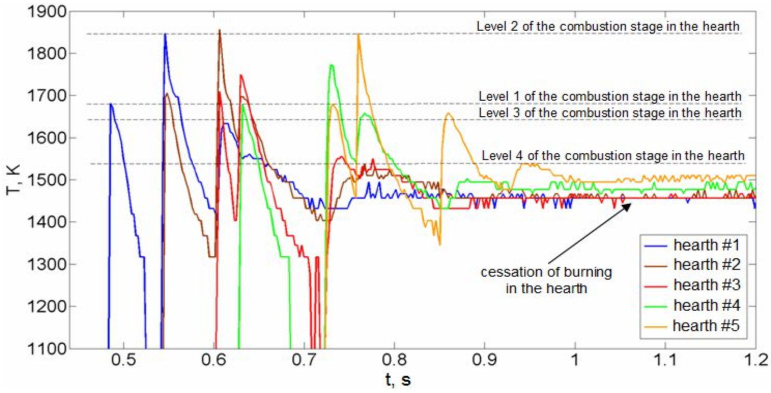


Figure 7. The dynamics of the maximum temperature in the analyzed section of foci.

After the occurrence of a layer of products burning in the cell stops. In accordance with the theory of reaction diffusion, combustion must resume after an interval over which the fuel (and/or oxidizer) overcomes the layer of products. However, the experiment shows that combustion in the cell resumes only with the formation of a new focy in the X_0 section. A similar scenario is likely when the processes in the reaction cell are strongly influenced by both internal and external heat sink. Then the diffusion mixing of the fuel and oxidant in the reaction cell does not lead to burning. The formation of a new source of combustion in the wave front, at a minimum, blocks, from the side of the initial mixture, the external heat sink of the cell entering the region of the previously formed source, and creates in it conditions for the resumption of the combustion reaction, which can occur in the thermal explosion mode.

In the considered case, by changing the perimeter of the sample, it is possible to control the time of diffusion mixing of the fuel and oxidant in the reaction cell. The presence of stages of extinction and explosion in it should lead to product stratification. Therefore, by varying the mixing time, the dispersion of the product can be controlled.

4. Conclusions

The discrete nature of the SHS process is not only manifested when the wave front propagates. In the microheterogeneous mode of combustion, it is detected both with the development of foci forming the front of the synthesis wave, and on the scale of the reaction cells. Based on the knowledge of product stratification in the high-temperature synthesis wave and the results of these experimental studies, it can be concluded that an increase in the heat deficit in microheterogeneous combustion regimes leads to an increase in the dispersion of the resulting product.

Acknowledgements

The work was supported by the Russian Foundation for Basic Research in research projects no. 18-08-01475, 18-08-01152, as well as grant no. 13-01-20 / 28 "Leading Scientific School of Ugra State University".

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