

Conference Paper

Express-analysis of Phase Composition of SHS Products

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Abstract

The first results of express identification of phase formation in the course of self-propagating high-temperature synthesis are presented. The proposed method does not replace the metallographic analysis. In the basis of the express method, the hypothesis about the integral picture of combustion formed during the reaction is an individual "image" of the process inherent in a particular system. The formation of the "image" is affected by the physical and chemical processes that are taking place. The results of the correlation analysis of the surface images of synthesized materials (FeO-Al-TiC, Al-C-Cr₂O₃-CrO₃, Nb₂O₅-Ti) and photographs of their sections are presented. The method can become an additional tool for analyzing the relationship between the combustion regime and the properties of the resulting material.

Keywords: SHS, correlation coefficient, phase formation

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1. INTRODUCTION

Often a correlation method is used to identify existing dependencies between the investigated variables. Successful application of the correlation analysis for navigation systems, control of technological processes, research of natural resources of the environment, etc. confirms the wide possibilities of the method. The popularity of the method is determined by two factors: correlation coefficients are relatively simple in counting, and their application does not require special mathematical preparation. Correlation methods are capable of assessing the mutual similarity of images that differ greatly in brightness characteristics if these images contain stable objects that make a significant contribution to the energy of the spectrum. Correlation methods work well in the absence of mutual rotation and scaling of images, and not too significant brightness distortions.

Carrying out experiments on self-propagating high-temperature synthesis, it was noted that the observed patterns of combustion of the same systems have a significant degree of similarity. This is explained by a certain sequence of reaction components

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of the batch, due to their physico-chemical properties. The burning of each system in the same modes is unique, as can be seen from Figure 1.

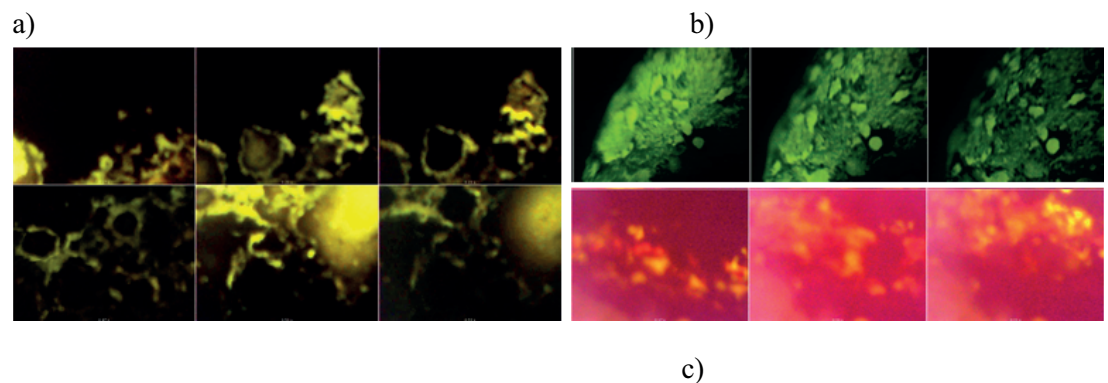


Figure 1: Frames of the process of SHS system a) FeO-Al-TiC; b) Nb₂O₅-Ti; c) Al-C-Cr₂O₃-CrO₃.

Having created a database containing such templates, it is possible to identify combustion modes in in-situ mode. Thus, the task is reduced to creating a sample (template), which will be searched. It is known that the thermophysical parameters of the reaction strongly affect the structural and phase composition of SHS products. With the help of a metallographic microscope, both qualitative and quantitative microstructural analysis can be performed. A photo of an etched section can become a generalized pattern of the structure of the product obtained. Thus, to achieve a global goal, two tasks must be solved: to learn how to identify the required phase in the combustion of SHS and to establish a relationship between the burning regime and the properties of the final product. At the first stage, it is necessary to check whether the identification can be realized by the correlation method. The purpose of this paper is to test the possibility of identifying the presence of a phase in the stage of its formation during the combustion of SHS by the correlation method.

2. EXPERIMENTAL SETUP

The solution of the problem of phase identification in the SHS process requires an integrated approach. Bright brightness micropyrometry methods, metallographic analysis, phase diagrams were taken into account, high-speed video recording and correlation analysis were carried out. To obtain samples of the FeO-Al-TiC system, self-propagating high-temperature synthesis used aluminum powder PA4 with a dispersion up to 50 μm , titanium carbide with a dispersion up to 100 μm , and iron oxide with a dispersion up to 300 μm . The powders were mixed in percent by weight: FeO-80% by weight; Al-20% by weight, which constituted the termite mixture. TiC was added to

the thermite mixture. 2-5 experiments were performed by the addition of TiC-10 wt.% And 20 wt. The charge was placed in a quartz reactor. The whole process of synthesis was filmed on a video camera. The photograph of the experimental setup is shown in Fig. 2.

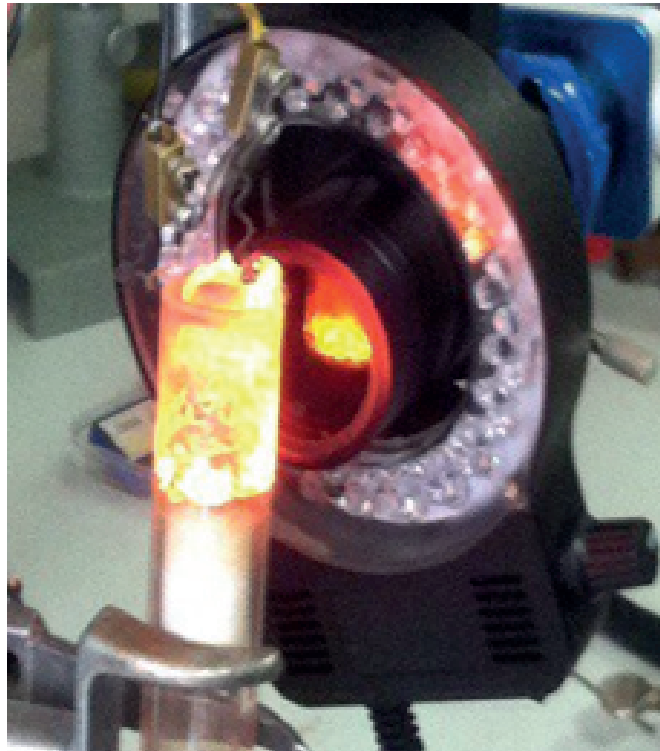


Figure 2: Experimental setup.

One of the conditions that significantly simplifies the analysis of results is the video shooting at the same scale as the microstructure template. Therefore, the micro-scale process was monitored with a USB5M Microscope camera and a FYSCOPE lens with a magnification of up to 500X. The microscope was directly connected to the computer, the video recording of SHS was conducted at a frequency of 30 frames per second, the working distance was 50 mm. The camera is calibrated using a reference temperature lamp. The temperature of the field of interest was measured by the method of brightness micro-pyrometry. The image of the selected active region $150 \times 150 \mu\text{m}^2$, is formed by pixels of $6.25 \times 6.25 \mu\text{m}^2$, in each of which the local temperature is determined. Comparing the experimentally obtained combustion thermograms of the selected region, the statistics of the local temperature observed in each pixel of the recorded frames, the thermal imaging image of the same region and the phase diagrams of the state, one can clearly visualize the processes occurring in SHS.

By comparing the current local temperature with the temperatures of the phase diagram of states, regions corresponding to the temperature of the phase transformations

are found [1]. It follows from the phase diagrams that the phase formation of carbides passes at high temperatures. On the images of the SHS process, these areas will be the lightest. In addition to measuring the average temperatures of localized regions located along the perimeter of the pores, micropyrometry was carried out in the active zones of the melt in the cavities. At some point, the melt in the cavity underwent structural and phase changes. At the same time, the camera fixed the increase in brightness and movement of the substance. In this case, the brightness temperature was averaged over the region $50 \times 50 \mu\text{m}$, since the pore sizes are sufficiently large. The video files were saved in uncompressed avi format. The analysis of the video files of the SHS process, obtained during video shooting, was carried out in the freely distributed ImageJ program.

The main stages of working with images include: separation of RGB channels and pre-processing, segmentation, description and analysis of the image. At the stage of preliminary processing, noise removal (smoothing, filtering), improving contrast, distortion correction and image binarization. At the third stage, segmentation algorithms are implemented. There is a division of the image into its component parts: objects, their fragments or characteristic features.

After passing the wave of combustion and cooling, the samples were prepared for metallographic studies. The most laborious stage of sample preparation is polishing of the surface of synthesized samples. The acid treatment of the surface of the thin sections makes it possible to observe the various phases of the material obtained. Metallographic studies were carried out using a microscope

and Axiovert»-200MAT, ProGres registration system and "VideoTest-Structure 5" television image processing system. In addition, photographs of the microscope polished sections were analyzed in the ImageJ program. The main phases were identified in the images of the surface of the sections. The images clearly show two phases-black and white. The sizes of the isolated phases were measured. The black phase on metallographic images is carbides. With the help of the fast Fourier transform, we tried to reveal the repeated regularities in the images. The statistical data of measurements of the dimensions of the phases are shown in Fig. 3.

The microstructure of the sample with the addition of 10 mass% of titanium carbide consists of a large lamellar perlite with large inclusions of carbides. The carbides in this sample are large, almost regular in shape (Figure 4b), there are also carbides in the form of small oblong bands.

The obtained samples were examined on a DR-01 "Radian" diffractometer, EVO 50XVP scanning electron microscope (Carl Zeiss) with an attachment for X-ray micro

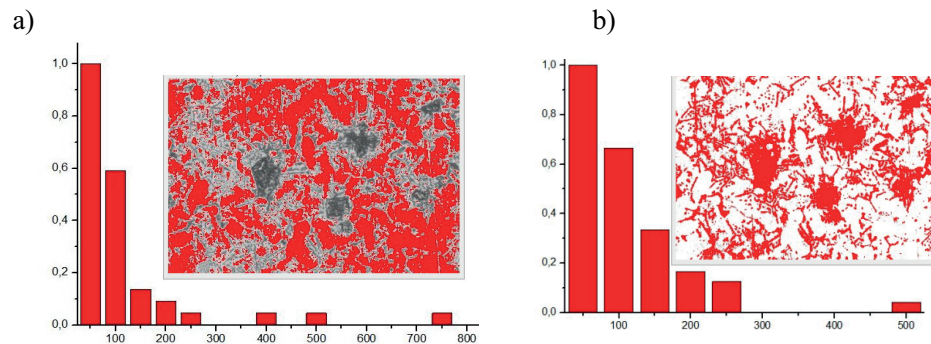


Figure 3: Statistical data on the sizes of the white phase (a) and black (b).

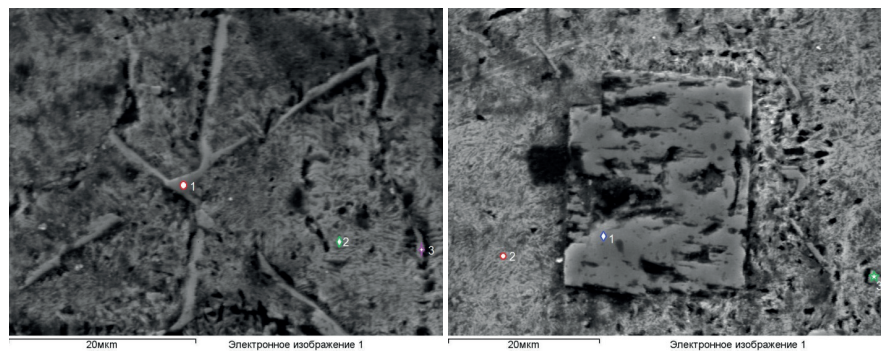


Figure 4: SEM micrographs of the surface of the combustion products of the FeO-Al-TiC system.

analysis. X-ray phase analysis showed the presence of phases: Fe_2C , Fe_3C , FeC , Al_2O_3 , TiC , as well as the presence of pure iron phase. Comparing the x-ray microanalysis and the phase diagrams of titanium-carbon and iron-titanium-carbon, it can be concluded that in region 1 and 2, TiC is formed in spectrum 1. In the remaining spectra of selected sites, perlite with cementite is formed.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The scale of the images of the metallographic microscope and video files was 5 times different. In spite of this, a trial correlation analysis of images of the combustion process and images obtained with a metallographic microscope was carried out. The correlation coefficient is a quantitative measure of the degree of kinship of two quantities. Correlation analysis was carried out in the ImageJ program. The plugin generates a scatter plot consisting of the pixel values Image1 (array1) and Image2 (array2). As a reference image, a photograph was taken of one of the phases (titanium carbide) obtained on a metallographic microscope. The resulting statistics contain: the correlation coefficient between Image1 and Image2 (R); slope of the regression line; The

constant C (the intersection with the y axis). Correlation can be performed on the basis of single pixels or local areas. The size of the local area can be predefined. The average value is calculated for each local area and is used to calculate the correlation coefficient.

A cross-correlation coefficient (Pearson correlation coefficient, r) is estimated according to [2]:

$$r = \frac{\sum_{i=1}^m \sum_{j=1}^n (f(m_i, n_j) - \bar{f})(g(m_i, n_j) - \bar{g})}{(mn - 1)\sigma_f \sigma_g},$$

$$\sigma = \left(\frac{1}{mn - 1} \sum_{i=1}^m \sum_{j=1}^n (z(m_i, n_j) - \bar{z})^2 \right)^{1/2},$$

$$\bar{z} = \left(\frac{1}{mn} \sum_{i=1}^m \sum_{j=1}^n z(m_i, n_j) \right).$$

where m and n are the number of local regions in the x and y -directions respectively, $f(m_i, n_j)$ and $g(m_i, n_j)$ are the local values of the chosen statistic at the local position (m_i, n_j) , \bar{f} and \bar{g} are the mean of the given statistic of the whole image and σ_f and σ_g are the standard deviations from the statistic on the left and right images respectively. Several statistics are implemented that may be used for correlating two pairs of images. The carried out statistics secure, complete assessment of the pattern in the images, including the mean greylevel, standard deviation, skewness and kurtosis of the greylevel (basis weight) distribution. In addition, the correlation may also be carried taking into account the local orientation of a given pattern. The orientation is derived from the mean resultant vector (Curry 1956), as implemented by Chinga et al. (2007). The Image CorrelationJ plugin also generates a correlation map. This is a valuable tool as the local correlation at each local position (m_i, n_j) can be evaluated. Figure 5 shows the results of a comparison of a series of SHS frames and a metallographic image of the phase of titanium carbide.

In the program, it is possible to perform a correlation analysis of images on the stack. The stack contains the frames of the combustion process and the reference image. The reference is the metallographic image of the selected phase - the "source". The analysis is carried out with the active slide containing the image of the "source". Searching for the similarity between the "source" and the slides in the "Goal" stack is performed. If you choose two stacks, but you want only to analyze the current slice, select this option channel 1 with its corresponding slice in channel 2. Based on the measurements, a correlation map is generated showing the difference between the local areas. A correlation graph "source" - "goal" is constructed.

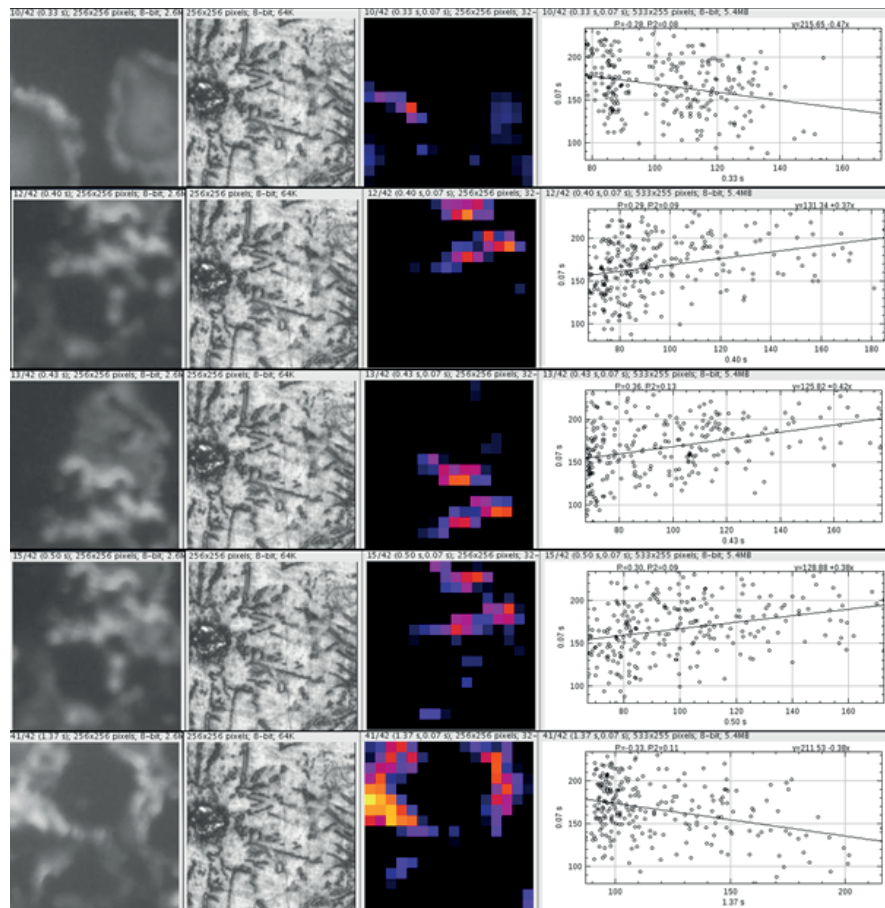


Figure 5: A series of frames (from top to bottom); from left to right: Target- Source- Correlation map- Scatter plot composed of the pixel values of Image1 (array1) and Image2 (array2).

In addition, several statistical data can be compared, including: mean, std, asymmetry, kurtosis and direction. Table 1 shows the statistical data: the correlation coefficient between the “source” and the “target” for standard deviation.

It is known [3] that: strong, or close at the correlation coefficient $r > 0.70$; average (at $0.50 < r < 0.69$); moderate (at $0.30 < r < 0.49$); weak (at $0.20 < r < 0.29$); very weak (at $r < 0.19$). This is the Pearson’s correlation coefficient. Zero-zero pixels are not included in this calculation. In many forms of correlation analysis the values for Pearson’s will range from 1 to -1. A value of 1 represents perfect correlation; Represents perfect exclusion and zero represents random localization. However, this is not the case for images. While perfect computation gives a value of 1, perfect exclusion does not give a value of -1. Low (close to zero) and negative values for Pearson’s correlation coefficient for images can be difficult to interpret. However, a value close to 1 does not show reliable correlation. Based on the correlation analysis data, there is a weak and moderate negative correlation between the dark areas of the metallographic image and the light areas of the video files. Thus, a negative correlation determines titanium

TABLE 1: Statistics of the correlation between Image1 and Image2 (std).

array1	array2	R	R2	Slope	C
Target	Target	1.00	1.00	1.00	0.00
Stack-1	Source	0.00	0.00	0.05	11.62
Stack-2	Source	0.63	0.40	3.41	6.31
Stack-3	Source	0.61	0.37	2.79	7.49
Stack-4	Source	0.63	0.40	1.49	8.88
Stack-5	Source	0.70	0.50	2.33	7.74
Stack-6	Source	0.65	0.43	3.09	6.73
Stack-7	Source	0.65	0.42	2.17	7.39
Stack-8	Source	0.61	0.37	2.21	7.34
Stack-9	Source	0.68	0.46	1.26	9.09

carbide. If we compare the metallographic image of the TiC phase and the CB synthesis image of the Nb₂O₅-Ti system, then the correlation coefficient does not exceed 0.15. This can be seen on the graphs (Figure 6).

The same result is obtained by studying the correlation between the image of the TiC phase and the image of the synthesis of Al-C-Cr₂O₃-CrO₃.

4. CONCLUSIONS

Results of correlation analysis of surface images of synthesized materials (steel, ceramic-metallic material, Nb₂O₅-Ti system) and metallographic sections:

- there is a moderate correlation between the formed phase and its presence in the photos of the section;
- using a correlation analysis to identify the phase during phase formation, a weak correlation should be eliminated;

To improve the results, it is necessary to select the same magnification of the metallographic microscope and eyepiece of the video camera. It is necessary to create generalized images of phases containing features inherent in the phase formation of a particular system. This will allow for a preliminary analysis of the phase composition of the reaction products. The accumulation of comparative statistics and the formation of a common base of the revealed features of combustion of various systems will make it possible to significantly refine the express method; can become an additional tool

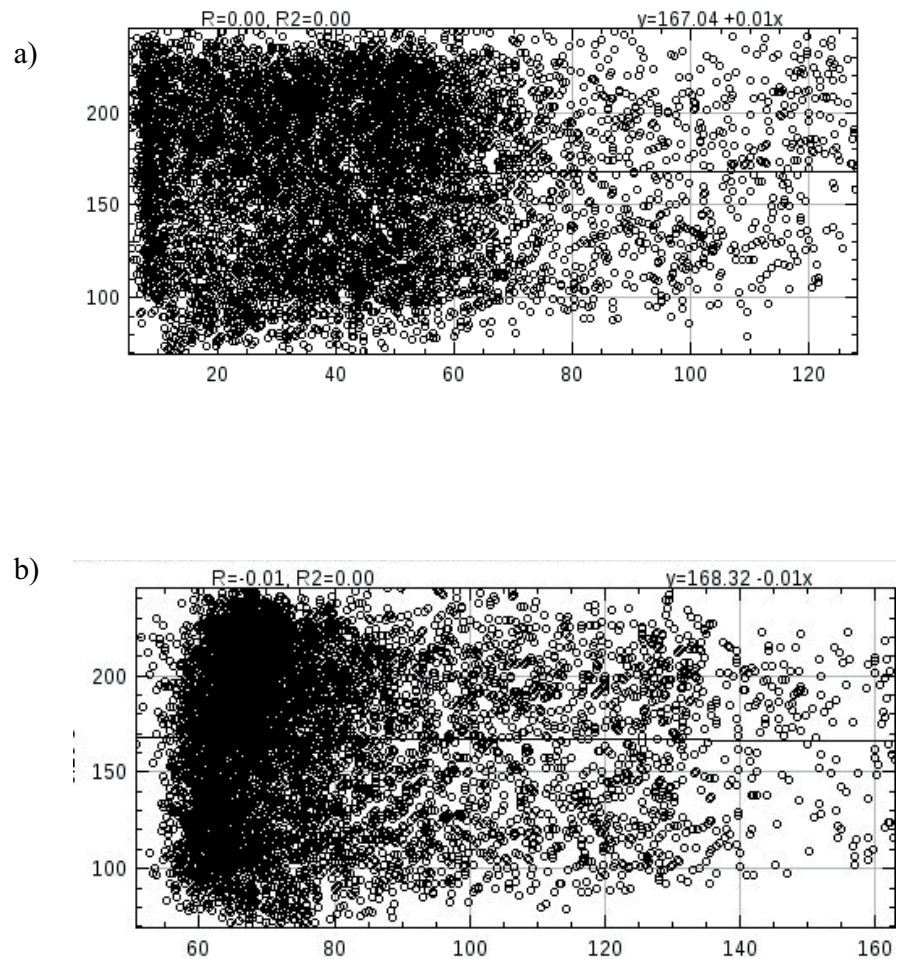


Figure 6: A scatter plot consisting of the pixel values of the image of the combustion surface of $\text{Nb}_2\text{O}_5\text{-Ti}$ (a) and $\text{Al-C-Cr}_2\text{O}_3\text{-CrO}_3$ (b) and photographs of the metallographic section.

for controlling the combustion regime; will help to identify unknown factors that affect the properties of the resulting material.

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