Instability of the Ni-Al combustion wave in the Zeldovich-Barenblatt parameters

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Abstract. The study of the thermal structure of the combustion wave of self-propagating high-temperature synthesis using differential chronoscopy (DCS), using the example of the Ni-Al system, demonstrates the possibility of estimating the criterion of thermal diffusion instability of the front in accordance with the Zeldovich-Barenblatt model. The application of standard image recognition methods, in the form of a high-speed Trace-transform algorithm, to a 2D chronogram of the interframe difference in the coordinates of the combustion front, calculated from high-speed video recording of the propagation of a SHS wave as a DCS-map, demonstrates the ability to control the sustainability mode of the burning wave in situ.

1. Introduction

Self-propagating high-temperature synthesis (SHS) is the basis for the development of additive technologies for the production of 3-D materials from metal-ceramics [1, 2]. In order to form functional properties, inert additives in the form of low-melting salts [3] or complex oxides [4] are introduced into the composition of the main exothermic mixture of Ni and Al powders. The SHS burning wave is visualized and the stability of the process parameters is monitored using high-speed nanosecond video cameras [5, 6]. The temperature and the synthesis rate are measured by high-speed brightness micropyrometry [8-10]. At the same time, the question of the experimental possibilities of visualizing the characteristic features of theoretically unstable combustion regimes based on the results of high-speed video and micropyrometry [11–13] has not been studied. A special feature of the high-speed process registration methods used in this work is that first in each video frame, the combustion front is highlighted, and then the interframe difference of the combustion front coordinates along the propagation direction of the SHS wave is calculated [9]. As a result, an instantaneous displacement map of the combustion sites can be obtained, which visualizes the topological and temporal coherence of the combustion wave during the decay into thermal cells [10]. This technique eliminates the masking effect of the random thermal structure of the combustion wave and provides a high degree of compression of the video data stream of high-speed video [15]. The purpose of the study is to determine the visual signs of instability of the combustion front of a SHS, when it decays into cellular thermal heat sources, to enable the subsequent comparison of the spatial parameters of the thermal structure of the combustion wave with the theoretical values of the instability criterion of the combustion wave.

2. Theoretical model and experimental procedure

The theoretical basis for the quantitative analysis of the stability of combustion regimes was laid in the article by Zeldovich - Barenblatt [7], where the appearance of a visually observable phenomenon of the
heat diffusion front decay into small thermal structures was predicted, and all the main characteristic scales were presented as a small perturbation model:

\[ x = f(y, t) = \varepsilon \exp(iky + i\omega t + \varphi t) - \varepsilon \exp(\varphi t) \sin(ky), \]  

where \( f(y, t) \) is the surface of the combustion front; \( \varepsilon \) is a small value; \( k \) is the wave number of the transverse perturbation; \( \omega \) is the circular frequency for the thermochemical induction time; \( \varphi \) is the decrement of the temperature attenuation of the Michelson heating zone (see Figure 1).

\[ \text{Figure 1. Visual signs of instability of a flat combustion front } f_0(t) \]

The change in the visually observed speed of combustion of the sample \( V^* \) and the temperature \( T_r \) of the SHS reaction in a perturbed thermal front must satisfy the conditions:

\[ V^* = \langle V_x \rangle = \frac{E(D - \alpha)}{2RT_{ad}} \times \frac{\partial^2 f(y, t)}{\partial y^2}, \]

\[ T_r = T_{ad} + (T_{ad} - T_0)h \times e^{iky + i\omega t + \varphi t} \]

where, \( \langle V_x \rangle \) is the propagation velocity of the unperturbed plane burning front \( f_0(t) \) along the OX axis of the sample; \( E \) is the activation energy; \( T_{ad} \) is the adiabatic burning temperature; \( D \) is the diffusion coefficient of the component of the fuel mixture; \( \alpha \) - coefficient of thermal diffusivity of a mixture of powders; \( h \) is the dimensionless coefficient of efficiency of heat and mass transfer in the reaction zone.

It is convenient to apply differential chronoscopy (DCS) methods [8] for recognition of non-stationary phenomena that arise as a result of loss of thermal stability of SHS, in the form of a reaction front breakdown at several high-temperature foci, followed by the analysis of the thermal structure of the combustion wave using Fast Fourier Transform (FFT), Trace Transformations (TT) or Hough Algorithms (HA) [9]. As it is shown in [10], image analysis of DCS with the help of TP is invariant with
respect to spatial frequencies, and hence the magnification scale at high-speed video, which determines the choice of TP as the main analysis tool.

The images of 2D chronograms of the thermal structure of the combustion wave obtained by DCS are presented below.

Figure 2. DCS maps of the Ni-Al system combustion wave at various degrees of dilution with an inert additive

The dark areas in the figures correspond to the “attenuation” of the motion of the combustion wave for the duration of thermochemical induction, which is necessary for heating the next cold layer to the ignition temperature. The interframe difference of the coordinates of the combustion front along the axis
OX, corresponding to small values of $\varepsilon$ in equation (1) and equal to the size of the thermal half-width of the arising local source of combustion, is plotted vertically in the form of light steps. Visually noticeable is the change in the wave number $k$ of the circular frequency $\omega$ of the instability $x=f(y, t)$, with an increase in the internal heat sink due to the introduction of an inert additive.

The detection of unstable SHS burning modes is performed in a standard way for computerized pattern recognition methods [10], procedures for converting DCS maps into image Trace-matrices of “typical” $T_3$, $T_4$ and $T_5$ functionals corresponding to different Radon harmonics for stationary combustion of the undiluted Ni-Al system (in Figure 3a) and unstable combustion of this system when diluted with 30% inert additive (in Figure 3b).

![Figure 3. Comparison of matrices of Trace-images at various degrees of dilution with inert additive](image)

3. Results and discussion
The experimental procedure consisted in the video recording of the combustion front of the SV-synthesis of an equimolar mixture of nickel powders (PNK-UT3) and aluminum (PA-4) with an inert addition of
powder from the final products of the \( \text{Ni}_1 \text{Al}_1 \) synthesis, the mass fraction \( \sigma \) of which changed from 0 to 30\% in steps of 2.5\%. The choice of the additive is due to the fact that the same heat capacity of the inert and synthesis products makes it possible to evaluate the efficiency of the internal heat sink, when the dispersion of the additive changes under the conditions of equal mass fractions of the powders. To register the propagation of the combustion wave of the SVS, an optoelectronic microspectrometry complex of nanosecond resolution was used on the basis of an ultrafast television camera “Video SprintNG” (ZAO NPK VideoScan, Russia) with an electron-optical photomultiplier converter on a microchannel plate and an optical gate “Nano-Gate” [5, 6].

To the limit of dilution in 15\% of the mass fraction of inert, the matrix of Trace images of DCS, the combustion waves were almost identical, as it is shown below.

**Figure 4.** Experimental confirmation of the Zeldovich-Barenblatt criterion for the stability limit of SHS when diluted to the theoretical limit of 23.9\%

In this case, the Zeldovich-Barenblatt criterion for the stability of the chemical reaction of the combustion of the \( \text{Ni}-\text{Al} \) system \((E \approx 128-149 \text{ kJ} / \text{mol}; T_{ad} \approx 1700-2100 \text{ K})\) experimentally confirmed for the processes of solid-flame burning at \( \sigma_{\text{max}} \approx 23.9\% \) that corresponds to the region of thermally stimulated diffusion, where \( D > \alpha \). The limiting value of an inert additive is limited by the region of thermal instability, where \( D < \alpha \) and the Lews number decreases with an increase in the proportion of inert additive, due to the increased heat transfer of the combustion wave to the cold inert [14].

As it can be seen from Figures 4 and 3b, in the matrices of Trace-images, signs of thermal instability of the wave are observed, which manifest themselves in the degeneration of the central elements of the matrix and the increase in the values in the peripheral columns of the TT-matrix. This confirms the earlier conclusion [15, 16] about the effectiveness of using the simplest criteria for detecting thermal instability of a combustion wave in amplitude and offset of the histogram of the values of the matrix elements of the Trace images of the DCS of the SHS wave, but with a substantial correction to the indicated sample areas to calculate the corresponding statistical criteria.

4. **Conclusion**

1. The study of the thermal structure of the combustion wave of SHS using DCS methods, using the example of the \( \text{Ni}-\text{Al} \) system, demonstrates the possibility of estimating the criterion of thermal diffusion instability of the front in accordance with the equation in the Zeldovich-Barenblatt model.

2. The application of standard image recognition methods, in the form of a high-speed Trace-transformation algorithm, to a 2D chronogram of the interframe difference in the coordinates of the combustion front, calculated from high-speed video recording of the propagation of a CBC wave as a DHS map, demonstrates the ability to control the stability of the in-situ burning mode.

3. By analogy with the Plancherel theorem on convolution of functions, in spatial-frequency Fourier analysis, for using Trace-analysis in SHS stability control systems, it is necessary to accumulate an experimental database of Trace- matrix at images DCS of the combustion wave in the obviously unstable modes and select the corresponding "masks transmittance" in the Trace-spectrum.
4. The simplest statistical criteria, in the form of evaluating changes in the average amplitude distribution of harmonics (elements) in the Trace-matrix, confirmed their effectiveness and good agreement, using the example of an experimental test of the Zeldovich-Barenblatt theoretical criterion, for the stability of the chemical reaction of the Ni-Al combustion system of equimolar composition.

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