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Dependence of burning velocity on the sample size in the nonactivated and mechanically activated Ni + Al systems

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The burning velocities of Ni + Al films 270–360 μm thick are greater than those of the samples 8–12 mm in diameter and 15 mm in height pressed from Ni + Al powder by a factor of 4–20. The data are explained in terms of a convective–conductive model of combustion wave propagation.

The effect of preliminary mechanical activation (MA)[1–8] on the combustion of powders has been intensely studied. This influence on solid-phase reactions consists in the removal of oxide and adsorbed layers from the surface of particles, in an increase in the area of contact between reagents, and in intense plastic deformation of material leading to a high concentration of crystal structure defects and internal tensions. The samples pressed from mechanically activated powders ignite at lower temperatures than those pressed from initial powders and the combustion velocity of MA samples can be markedly higher. In addition, there are examples of a transition from the reaction with the participation of liquid to a solid-phase mode of combustion after preliminary MA of the same mixtures.[6] Particular attention is given to the influence of MA on Ni + Al[4–8] powder combustion. It was established[9,10] that the combustion of Ni + Al powder without MA is described by the convective-conductive model of combustion.[11] According to the model, convective heat transfer during the propagation of a combustion front is caused by a current of a liquid layer of a fusible reagent under capillary forces and a pressure difference of impurity gases before and behind a fusion layer. Thus, the average velocity of fusion penetration into initial powder is the velocity of combustion wave propagation. It was suggested[4] that combustion in the Ni + Al system after MA is described by a microheterogeneous model,[12] according to which mixed powder is represented by a set of identical reaction cells; each cell contains fuel and oxidizer in the same proportions as in the initial mixture; temperature and other parameters are the same for a cell, and combustion extends due to heat exchange between cells. However, both in the quasi-homogeneous[13–15] and in the microheterogeneous theory, the convective transfer of heat and reagents related to fusion movement in the direction of the combustion front propagtion is not considered, as well as the effect of impurity gas emission. Thus, it was interesting to reveal whether there is a change of the combustion mechanism in Ni + Al system after MA. Convective–conductive and microheterogeneous combustion models qualitatively predict the dependence of the combustion velocities of a sample on its diameter. In the convective–conductive model, the combustion velocity can decrease with the diameter of a sample,[11,16] whereas in the microheterogeneous model, it increases with the diameter up to a constant value. Therefore, the dependence of combustion velocity on sample diameter (thickness) was chosen as a characteristic of the mechanism of combustion wave propagation.

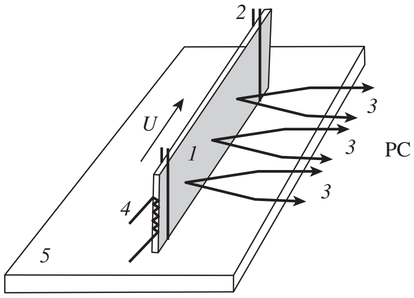

Figure 1 Measurement of the burning velocity of films: (*1*) film, (*2*) film holder, (*3*) thermocouples, (*4*) initiating spiral and (*5*) wafer.

In this work the burning velocity of the films[†] was determined according to the scheme shown in Figure 1. To prevent film deformation, the film was fixed in a holder, which was set vertically on a lateral edge on a horizontal surface. Combustion was initiated by a tungsten spiral. The 100 μm thermocouples were placed at the distance L from each other in contact with a lateral surface of a film. Signals from the thermocouples were stored in a computer memory. The burning velocity of a film U was calculated as $U = L/t$, where t is the time between the signals

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[†] In the Ni powder (PNE-1 brand, 99.5 wt% Ni), particles with a size less than 43 μm made up 50%, and particles with a size less than 51 μm made up 90% of the total number of particles. In the Al powder (ASD-4, 99.2% Al), the particles with a size less than 11 μm made up 50%, and particles with a size less than 16 μm made up 90% of the total number of particles. The fractional structure and particle-size distribution were determined on MicroSizer 201. The MA of equimolar Ni + Al powders was performed for 50 s at 90*g* (*g* is the acceleration of a free fall) in an AGO-2 mechanoactivator in an Ar atmosphere.[4] The weight ratio between spheres and powder was 20 : 1. After MA, the composite particles formed were eliminated. Activator drums, with layers of the mixture, stuck to its walls and the surface of spheres, were filled with 40 ml of distilled water, and then additional activation for 3 min was carried out. Activation in liquid caused the flaking of stuck layers and their dispersion with the formation of ultrafine particles of a scaly shape. Samples of 8, 10 and 12 mm in diameter and 10 – 18.5 mm in height were pressed both from the initial mixture and the particles activated in water. The initial sample weight was 1.4–6.43 g, and the relative density was 0.53–0.65. The combustion of samples was performed in Ar at 1 bar. Combustion was initiated from the top end face of a sample by means of a tungsten spiral. For initiation on all surface of an end face of a sample an intermediate tablet from fast-burning Ti + 2B composition was placed between it and the spiral.

The combustion was recorded with a video camera. Combustion velocity was determined from the record of combustion of the central part of a sample to exclude the influence of an initiating tablet and a substrate. The error of velocity measurement was no higher than 10%.

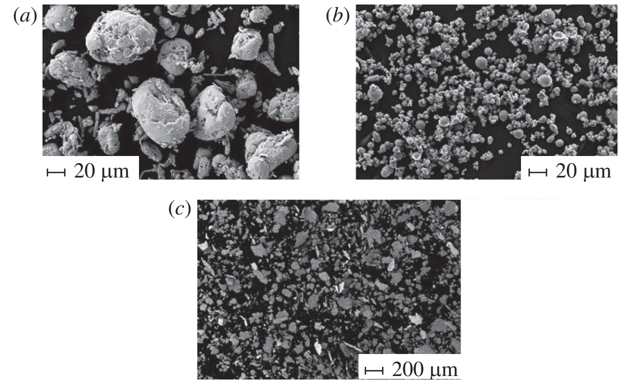

Figure 2 Images of (*a*) Ni particles, (*b*) Al particles and (*c*) mixed Ni + Al particles after MA for 50 s and activation with the addition of water for 3 min.

of thermocouples. The X-ray phase analysis of the burned-down samples showed that combustion products do not contain oxides.

Figure 2 shows the morphology of the initial Ni and Al powders and Ni + Al particles after MA for 50 s and activation with water addition (dispersion) for 3 min. Initial particles of Ni and Al have a spherical shape with a smooth surface; however, Ni particles were considerably greater than Al particles. During activation, composite particles consisting of the alternating layers of initial components[4] are formed. In the course of further activation in water, the formed composite particles are stratified to form flat scaly particles. Note that the X-ray phase analysis of activated mixtures demonstrated that their phase structure is identical to the phase composition of the initial mixture.

The dependence of the burning velocity on the diameter of the initial and activated samples is presented in Figure 3. The burning velocity of the initial mixture exceeded that of the mixture subjected to MA. Although the samples pressed from MA mixtures elongated in the course of combustion, and the samples pressed from the initial mixture did not change the length, the dependence of the burning velocity on sample diameter for both samples were nonmonotonic and had a maximum at a sample diameter of 10 mm. Note that it is impossible to obtain a uniform sample density for small samples. On the other hand, the samples of large diameters crack in the course of combustion due to impurity gas emission, which influences the combustion velocity. However, in the range of diameters from 8 to 10 mm, heat losses are a main factor affecting the dependence of burning velocity on diameter.

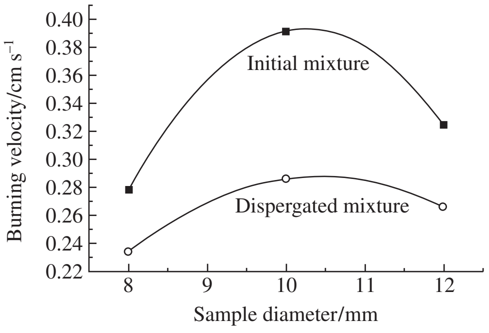

Figure 3 Dependence of the burning velocity on the diameter of a sample for the initial and activated mixtures.

The reduction of the burning velocity at an increase in the sample diameter from 10 to 12 mm is explicable on the basis of a convective–conductive combustion model[11] if we assume that the dominating factor is impurity gas emission ahead of the reaction zone. With increasing the sample diameter, the pressure of impurity gases grows to cause a decrease in the burning velocity according to the convective–conductive combustion model. However, the other possible explanation is associated with a reduction of heat conductivity of a sample in a warming up zone due to loosening effect of impurity gases.[12] To reduce the influence of impurity gas emission, the films obtained by rolling were used. If the burning velocity of films was greater than that of the samples pressed from the same particles, this would be evidence for the occurrence of a convective–conductive combustion mechanism.[11] In a microheterogeneous model, the burning velocity increases with the sample diameter. Therefore, if the microheterogeneous mechanism of combustion occurs, the velocity of combustion front has to decrease, or combustion has to be missing because of a strong increase in heat transfer coefficient. Note that, upon a transition from cylindrical samples to films, the specific surface of S/V (S is the lateral surface of a sample, and V is its volume), characterizing the influence of heat losses on combustion in the thermal theory[15] of flame propagation increases by a factor of 10. Figure 4 shows that the burning velocities of films obtained from initial powders and from MA increase by a factor of 4 and 20, respectively.

Such a significant acceleration of the films burning can not be explained by an increase in the heat conductivity coefficient. Indeed, according to the quasi-homogeneous theory of the combustion of heterogeneous systems,[15] the velocity of a reaction front is proportional to the square root of a heat conductivity coefficient. Therefore, the observed increase in the burning velocity (by a factor of 4 or 20) requires raising the heat conductivity coefficient by a factor of 16 or 400, respectively. However, data on the change of a heat conductivity coefficient of Zr + Al powder mixture (which is close to thermal and physical characteristics of Ni + Al mixture) show that it changes only twice over a porosity range from 35 to 50%.[18] Therefore, the observed increase in the burning velocity cannot be explained in terms of the quasi-homogeneous combustion theory, and it suggests the occurrence of a convective–conductive combustion mechanism.[11]

Thus, the experimental data obtained in this work can be explained in terms of a convective–conductive combustion model, which suggests an essential role of fusion and impurity gas emission in the process of combustion wave propagation.

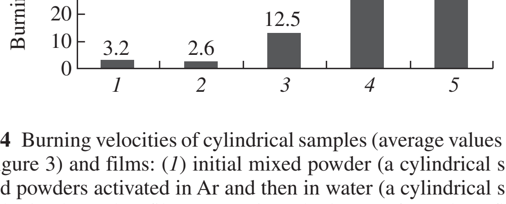

Figure 4 Burning velocities of cylindrical samples (average values according to Figure 3) and films: (*1*) initial mixed powder (a cylindrical sample); (*2*) mixed powders activated in Ar and then in water (a cylindrical sample); (*3*) initial mixed powder (film); (*4*) activated mixture of powders (film) and (*5*) mixed powders activated in Ar and then in water (film).

To exclude the influence of impurity gases on the velocity of a reaction front, the burning velocity of the films 270–360 μm thick obtained by rolling from initial mechanically activated Ni + Al mixed powders was determined. The preparation of the films was described elsewhere.[17] The thickness of the films prepared from initial mixture was 270 μm; from mechanically activated and dispersed mixture, 360 μm, the width of the films was 10–11 mm, and the relative density varied within a range of 0.66–0.74.

The structure of the initial and dispersed powders was investigated by a scanning electron microscopy (SEM) using an Ultra Plus microscope (Carl Zeiss). The initial powders and films before combustion were photographed with a Levenhuk D320L microscope.

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