

Activation effects on the deflagration speeds in Ni-Al

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1 Introduction

Combustion synthesis or self-propagating high-temperature synthesis (SHS) has received considerable attention in various metallurgical and pyrotechnical applications. In exothermic metal alloying processes, two metals are ground into a fine powdered mixture and are ignited to allow a reaction front to be established which propagates through the mixture, converting the metal reactants into an alloy product. To characterize the SHS process it is important to understand the physical and chemical mechanisms involved in the reaction along with the effects of various process controlling parameters such as thermal and physical parameters. Physical parameters include density, particle size, mixture ratio and powder compact geometry while thermal parameters refer to the initial temperature and thermal conductivity of the powder compact. Consequently, provided that the necessary thermodynamic conditions for self-propagation are met, control over SHS propagation for diffusional transformations, depends mostly on thermal properties such as diffusivity and conductivity, which are closely related to compact porosity and particle sizes. Another important parameter to SHS reactions is the energy supplied to the solid-solid system prior to the reaction initiation, which combines heating and mechanical work. As it was mentioned by Benderskii et al. (1989) [3], potential elastic energy stored in a reactive system reduces the apparent activation energy barrier to the reaction. In this study, activation refers to the potential energies stored in the form of residual elastic deformation during powder compaction or in the form of non-equilibrium defects created by mechanical alloying. To assess the changes in activation energy, deflagration speeds of mechanically activated powders are recorded using high-speed imagery and compared to non-activated powders for various compaction densities.

2 Experimental setup

Consider a mixture of reactive Ni-Al powders mixed in a stoichiometric proportion and pressed into a shape of a disk. Different densities of powder compact are achieved by changing the die compaction pressure and thickness of the disk. Activation of some of the powders was accomplished with a mechanical mill. These powders were milled inside a high-energy planetary ball-mill with a ball to powder weight ratio of 20:1 for periods between 30 to 60 minutes and by changing rotation direction every two minutes. The milling atmosphere was argon and the milling process was monitored using

temperature and pressure sensors inside the milling vials. In order to maximize the milling dose, arrested-milling was performed until the powders reacted in one of the vials, such that the second vial contained the arrested milled (ARM) powders. Ignition of the powders was done using a butane torch for low density compacts and with a propane-oxygen torch for higher densities. The amount of heat or preheating required to ignite a powder compact was appreciated using the time of heat supply until reaction and the type of torch used. Another apparatus used to analyze the activation effects on deflagration speed is a 5x5x100mm steel channel where powders are compacted and later ignited using a trail of loose powders at one end.

3 Results and discussion

If we take a closer look at the compacted mix of un-burned and un-milled powders structure (Figure 1), they seem to be thoroughly mixed and the average particles size is close to $8\mu\text{m}$. Nickel appears as light circles and aluminum as darker circles. By increasing the density of the powder compact the contact surface area of the reagents increases and the porosity of the compact decreases. This augmentation in contact surface area will lower the amount of preheating required prior to combustion and will allow higher conversion temperature to be reached closer to the adiabatic temperature of the transformation. As explained by Zeldovich-Frank-Kamenetskii model [4], an increase in conversion temperature will also increase the burning velocity of the SHS reaction.

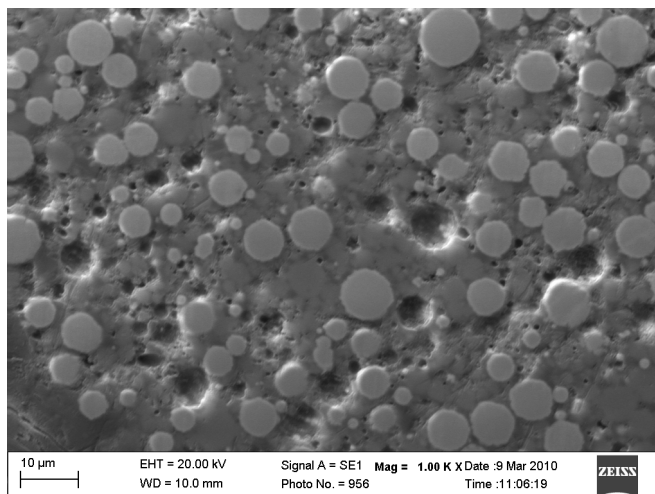


Figure 1. SEM image of nickel-aluminum (1:1) powder prior to combustion.

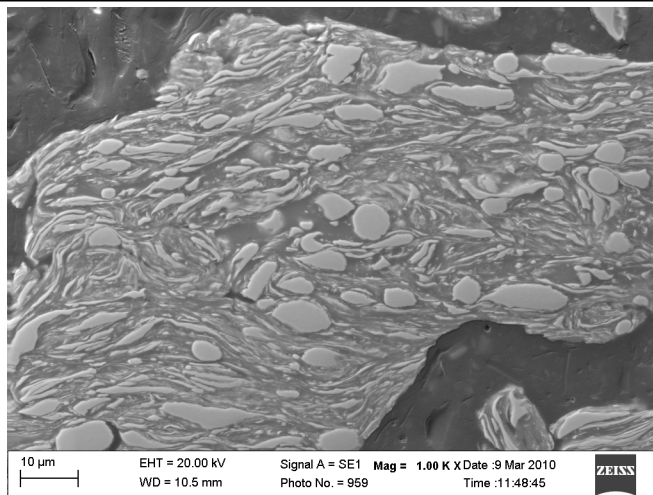


Figure 2. SEM image of mechanically activated nickel-aluminum (1:1) powder prior to combustion.

Concerning the milled powders (Figure 2), we can observe a fine lamellar structure which resulted from high plastic deformations during milling. Due to the distortions of the reactants phase crystal lattice, energy is stored in the form of potential energy because of non-equilibrium defects (dislocations, grain boundaries, point defects) and residual elastic deformation. The stored deformation energy alone cannot explain the apparent decrease in the activation energy without correlating the effects on the thermal parameters, but the long term action of intense periodic plastic deformations will decrease the barrier to the nucleation of a new phase by lowering the concentration gradient at the interface boundary of the two species [2].

The recorded velocities of combustion with respect to the density of the powder compact for milled and un-milled powders are shown in Figure 3 and Figure 4 respectively. For each type of reaction, a distinct front of propagation was observed and appeared to be constant throughout the combustion. As expected, an increase in density causes an increase in burning speed but required more preheating to initiate. In the case where powders were compacted inside the steel channel, initiation of high density SHS compacts could not be accomplished because of the heat sink effect of the channel and the higher thermal conductivity of the powder compact which is assumed to increase with decreasing porosity [2].

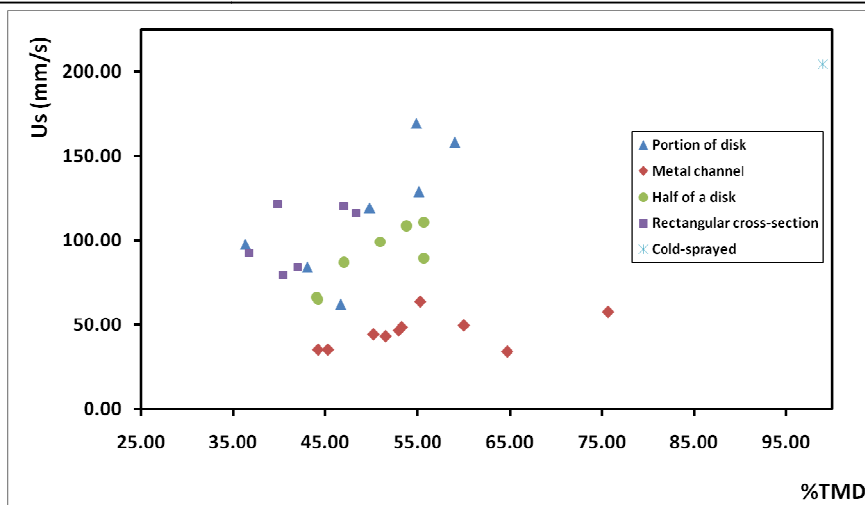


Figure 3. SHS burning speeds for milled powder compacts as a function of theoretical density.

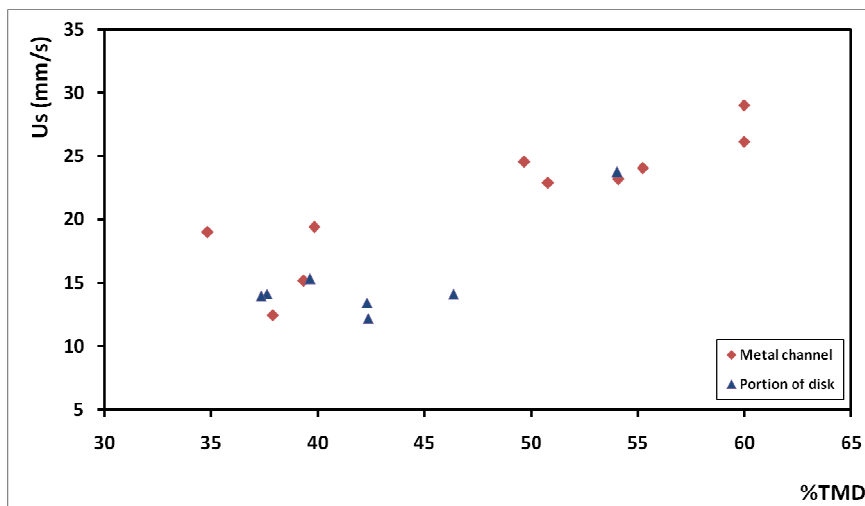


Figure 4. SHS burning speeds for un-milled powder compacts as a function of theoretical density.

If we now look at the recorded burning velocities of mechanically activated powders (Figure 3), they also appear to be dependent on compact density as for the un-milled ones, however noticeably higher. The increased contact interface area of the species and the increased thermal conductivity of the powder compact allow heat to be transferred quickly in front of the propagating wave, reducing the activation energy requirements. The latter phenomena coupled with the enhanced diffusion during mechanical activation allow for faster product phase nucleation and increases the conversion degree at lower temperatures [2]. This was observed experimentally by the significantly lower required preheating of the milled powder compact and the higher burning velocities.

4 Conclusion

The above experimental results demonstrated that for higher density powder compacts, the reaction propagation increases. This effect was observed for both types of powders: mechanically activated and as-received powders. The increase in contact area was observed by the presence of lamellar structure

after mechanical activation. Performing long cycles of mechanical alloying seems to reduce the concentration gradient at the interface boundaries of the species, allowing for faster nucleation of the product phase, as observed by the faster deflagration speeds. The effect of heat loss on the reaction speed was also appreciated when using various shapes of powder compacts and experimental apparatus (metal channel).

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