

## Simultaneous combustion synthesis (thermal explosion mode) and extrusion of nickel aluminides

K. MORSI\*

Department of Mechanical Engineering, San Diego State University, 5500 Campanile Drive, San Diego, CA 92182, USA  
E-mail: kmorsi@mail.sdsu.edu

S. O. MOUSSA

Department of Chemistry, Ain Shams University, Cairo, Egypt

JAMES J. WALL

Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN 37996, USA

Combustion synthesis (CS) is a low-energy processing approach for a wide range of materials (e.g. ceramics, intermetallics and composites), whereby reactants are ignited to spontaneously transform to products in an exothermic reaction. The process can be subdivided into two modes of ignition. The first is termed *self-propagating high temperature synthesis* (SHS) in which a reactant compact is typically ignited at one end using a heated tungsten coil, forcing a local reaction which then travels in the form of a wave consuming the reactants and forming the products. This type of ignition can generate a temperature distribution along the length of the hot product, since the material reacted first will also start to cool first. The second mode of ignition is termed *thermal explosion*, in which the whole volume of the compact is heated to the ignition temperature at which all reactant powder spontaneously convert to product(s). Both modes of ignition have previously been used in the fabrication of one of the most promising intermetallic materials today (nickel aluminides) [1, 2]. A general problem with CS is the generation of unwanted porosity in the final product. A number of uniaxial and isostatic pressure-assisted reactive processes emerged to combat this [3–6]. However, less attention has been given to the application of the more traditional thermomechanical processes such as extrusion *during* the high temperatures achieved in combustion synthesis of intermetallics. Morsi *et al.* [7–10] previously reported work on hot extrusion reaction synthesis (HERS), in which the idea was to use the heat generated during the extrusion of elemental powder compacts at billet temperatures 300–500 °C (i.e. below the ignition temperature of ~640 °C) to simultaneously form and consolidate intermetallics. The work resulted in the formation of intermetallics, however the reaction always occurred after the material had exited the die, thus bypassing the consolidation stage in extrusion, resulting in porous products. Also recently Minay *et al.* [11] reported some

interesting work on HERS of Ni<sub>3</sub>Al, in which the temperature was increased to 600 °C. By varying the steady-state billet temperature, billet hold time at temperature prior to extrusion (2–12 min) and extrusion speed, relatively long extrusions were produced, however the product was not completely reacted, and a mixture of phases resulted. In the present study we instead heat the compact to the ignition temperature (640 °C) and allow the exothermic reaction to proceed to completion inside the extrusion chamber thus ultimately heating the compact to the maximum combustion temperature (maintained for seconds). Then extrusion is conducted at pre-selected temperatures while the fully reacted product cools inside the extrusion chamber. This approach should offer a number of advantages including, the production of single phase alloy (assuming prior optimization of green density, particle size, etc.), consolidation, and grain size refinement through recrystallization. SHS extrusion of refractory materials such as TiC reinforced with a metallic binder (e.g. Ni) (to facilitate high-temperature deformation) was previously carried out in Russia (using SHS type ignition) [12, 13]. The present study instead deals with the reactive extrusion of nickel aluminide (Ni<sub>3</sub>Al) intermetallic alloys using the *thermal explosion* mode of ignition thus allowing the whole compact to simultaneously react and promote a more uniform temperature distribution prior to extrusion. An experimental setup for this reactive extrusion approach was recently reported by Morsi *et al.* [14] which allows for the variation and monitoring of processing parameters (e.g. extrusion speed, compact temperature and strain). In that work however, only the experimental verification of the process was presented, with hardly any variation in processing parameters. In the present paper, the effect of *compact* extrusion temperature on the Ni<sub>3</sub>Al product quality and microstructure is discussed.

Elemental powders of aluminum (gas-atomized 99.99% pure, <45 μm) from Atlantic Equipment

\*Author to whom all correspondence should be addressed.

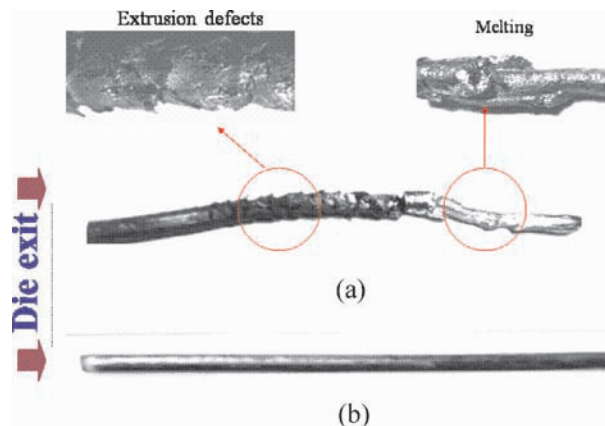


Figure 1 Reactively extruded Ni<sub>3</sub>Al at 5.5 s<sup>-1</sup> strain rate, 10:1 extrusion ratio and extrusion temperature of: (a) 1250 °C and (b) 1050 °C.

Engineers, NJ 07621, USA, and nickel (99.99% pure, INCO123, 3–7 μm in diameter, INCO Selective Surfaces Inc., NJ 07481, USA) were turbula mixed (Gienmills Inc., Clifton, NJ 07012, USA) together with 10 vol.% pre-alloyed Ni<sub>3</sub>Al powders (99.0% pure, <45 μm, Alfa Aesar, 30 Bond Street, Ward Hill, MA 01835-8099, USA). All powders were isostatically pressed to ~78% theoretical green density and then vacuum degassed at 300 °C for 12 hr. The compacts were then heated inside the extrusion chamber (15 mm inner diameter) maintained at a temperature of ~700 °C. A combination of glass and graphite was used for lubrication. The compact temperature was monitored and recorded using a K-type thermocouple inserted into the bottom of the specimen through the die exit, which is then simply pushed out on extrusion. Extrusion was conducted at a strain rate of 5.5 s<sup>-1</sup>, extrusion ratio (area reduction) of 10:1 and at varying compact temperatures of 1250, 1050 and 950 °C while the specimen was cooling down from the combustion temperature. Following extrusion, the samples were ground to half thickness along the extrusion axis, then polished and etched for microstructural examination. Etching was carried out using an etchant of composition 30 ml H<sub>2</sub>O, 10 ml HCl and 10 ml HNO<sub>3</sub>, and the average grain size of as-prepared Ni<sub>3</sub>Al was determined using the areal method according to ASTM standard [15]. X-ray diffraction (XRD) was also used to determine product phases. Initial preliminary experiments indicated that isostatic pressing was needed to enable uniform reaction characteristics and homogenous microstructures in the extruded product. Also dies with a conical die angle of 130° resulted in lower extrusion pressures compared to flat dies. XRD

scans on the extruded product also confirmed that the elemental powders transformed to the intended intermetallic (Ni<sub>3</sub>Al) with no un-reacted nickel or aluminum remaining. Fig. 1 and Table I summarize the results of extrusion at different compact temperatures.

At a compact temperature of 950 °C the extrusion load needed to extrude the material exceeded the load limit on the press, and therefore extrusion was not possible due to the high flow stress of the material at this temperature. However extrusion at 1050 °C (flat die) and 1000 °C (conical die) resulted in a successful extrusion 4.5 mm in diameter and ~10 cm long. Fig. 2 shows micrographs of the latter extrudate microstructure at different locations centrally along the extrusion axis. It is clear that there is a sudden increase in grain size as the material exits the confinement of the extrusion chamber, and therefore grain growth is no longer suppressed when the material exits the die. The grain size was also found to slightly increase with distance away from the die exit. Similar results were previously reported for the extrusion of pre-alloyed NiAl intermetallic [16]. Fig. 2 also shows the presence of oxide stringers, resulting possibly from pre-existing oxides on the initial elemental powders. At a billet temperature of 1250 °C the extruded material showed a clear transition from semi-molten to solid structure (Fig. 1). Both this and the increase in grain size away from the die exit suggest that the extrudate exit temperature continuously decreased with ram displacement during extrusion. It is also worth noting that cracks appear on the 1250 °C extrudate just before the material transitioned to a solid piece. These cracks are believed to be caused by the presence of still a small amount of molten phase at the point where the extrusion exited the die, leading to what is commonly known as hot shortness [17]. It is interesting to point out that similar defects were also previously reported by Morsi *et al.* [7, 8] when extruding elemental powder compacts at temperatures *below* the ignition temperature in HERS, where then the presence of a transient molten phase was suggested to be the cause.

It is well known that the exit temperature of the extruding material is in general higher than the initial billet temperature. Both the transition from semi-molten phase to solid phase (at billet temperature 1250 °C) and the increase in grain size away from the die exit (1050 °C) can be explained by taking into consideration the significant heat interchanges experienced by a material during extrusion. Heat is generated due to plastic deformation, internal shear and friction between the deforming material and the tooling, heat is also transferred within the billet and between the billet and the tooling [18]. Hence competing processes exist that would

TABLE I Summary of reactive extrusion results

| Specimen | Extrusion temperature (°C) | Die shape | Peak extrusion pressure (GPa) | Remarks                         |
|----------|----------------------------|-----------|-------------------------------|---------------------------------|
| A        | 1250                       | Flat      | 1.11                          | Partial melting + cracking      |
| B        | 1050                       | Flat      | 1.49                          | Successful                      |
| C        | 950                        | Flat      | 1.63                          | Reached upper load—no extrusion |
| D        | 1000                       | Conical   | 1.30                          | Successful                      |

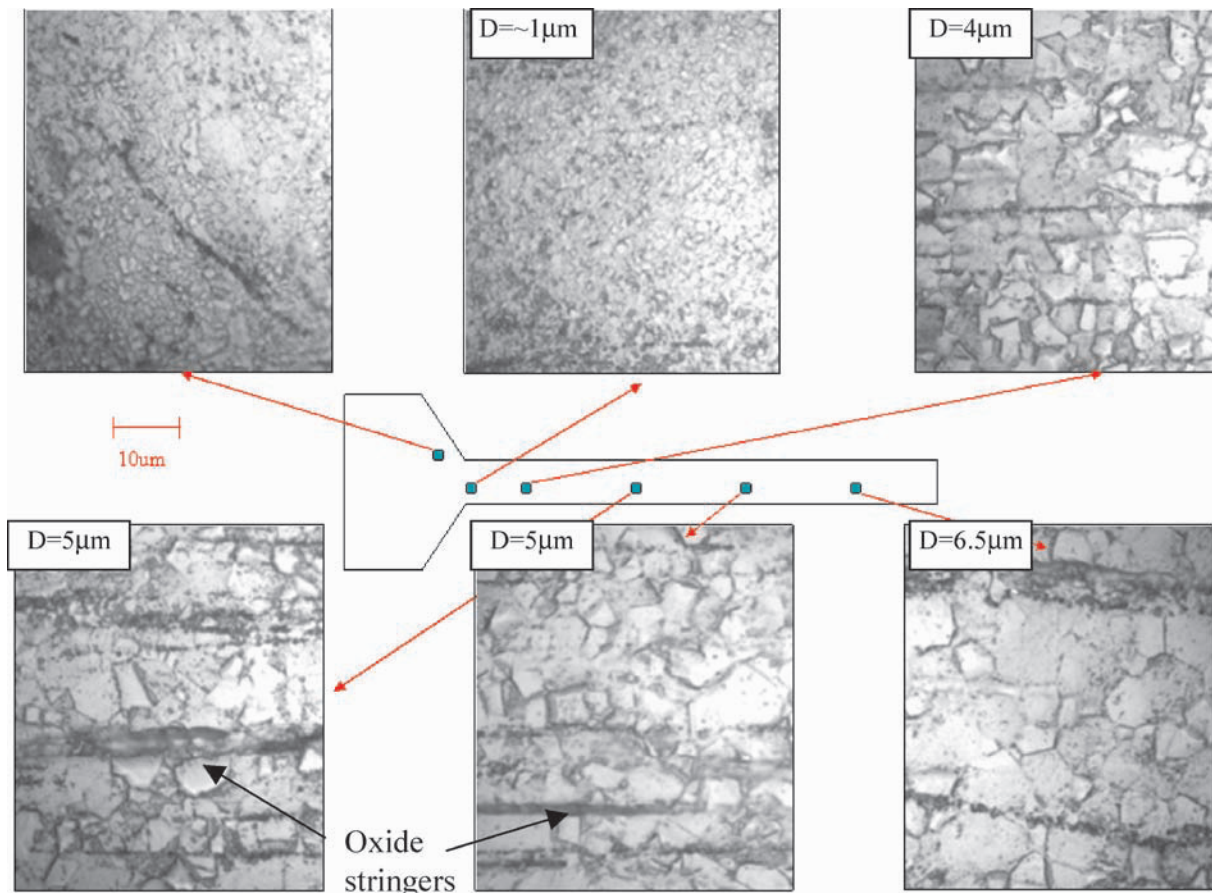


Figure 2 Microstructure along extrusion direction of  $\text{Ni}_3\text{Al}$  reactively extruded using conical die at  $1000^\circ\text{C}$ ,  $5.5\text{ s}^{-1}$  and extrusion ratio 10:1 (10  $\mu\text{m}$  scale bar refers to microstructure).

tend to either continuously increase or decrease the extrudate exit temperature with ram displacement during extrusion. The deformation and frictional heating favor a continuous increase in the material exit temperature with ram displacement, however, the lower temperature of our container ( $\sim 700^\circ\text{C}$ ) compared to the hot billet ( $\geq 1050^\circ\text{C}$ ) will favor significant heat losses from the material to the tooling and therefore a decrease in exit temperature with ram displacement during extrusion. It appears that at both  $1250$  and  $1050^\circ\text{C}$  heat losses to the tooling were more significant than the heat generated leading to an overall decrease in exit temperature with ram displacement during extrusion. As compared to a compact that was just ignited inside the extrusion chamber without extruding, the porosity was found to decrease from  $\sim 18\%$  to  $< 0.5\%$  upon extrusion. The average grain size also decreased from  $28.4 \pm 1.7$  to  $4.5 \pm 2.2\ \mu\text{m}$  due to recrystallization. Both consolidation and grain size refinement after extrusion resulted in an increase in the Rockwell hardness (A-scale) from  $53 \pm 2.9$  HRA to  $77 \pm 3.4$  HRA for the reactively extruded material. This value is greater than a reported value of 52 HRA [19] for reaction synthesized  $\text{Ni}_3\text{Al}$ , due to the smaller grain size of the reactively extruded (consolidated) material. In conclusion, the compact/product temperature can have a significant effect on the quality of the reactively extruded  $\text{Ni}_3\text{Al}$  intermetallics, and it appears that there is a small temperature-processing window through which sound, crack free materials can be reactively extruded.

The product however is highly dense with a refined microstructure.

## References

1. J. -P. LEBRAT, A. VARMA and A. E. MILLER, *Met. Trans. A* **23A** (1992) 69.
2. K. A. PHILPOT, Z. A. MUNIR and J. B. HOLT, *J. Mater. Sci.* **22** (1987) 159.
3. D. ALMAN and N. S. STOLOFF, *Int. J. Powd. Metall.* **27**(1) (1991) 29.
4. T. CHENG and J. SUN, *Scripta Metall. Mater.* **30** (1994) 247.
5. C. S. MARCHI and A. MORTENSEN, *Met. Mater. Trans. A* **29A** (1998) 2819.
6. D. C. DUNAND, *Mater. Manuf. Process* **10**(3) (1995) 373.
7. K. MORSI, H. B. MCSHANE and M. MCLEAN, *Met. Mater. Trans. A: Phys. Metall. Mater. Sci.* **31**(6) (2000) 1663.
8. *Idem.*, *Mater. Sci. Eng. A* **290**(1) (2000) 39.
9. K. MORSI, H. MCSHANE and M. MCLEAN, *J. Mater. Sci. Letts.* **17**(19) (1998) 1621.
10. *Idem.*, *ibid.* **19** (2000) 331.
11. E. J. MINAY, H. B. MCSHANE and R. D. RAWLINGS, *Intermetall.* **12** (1) (2004) 75.
12. V. V. PODLESESOV, A. V. RADUGIN, A. M. STOLIN and A. G. MERZHANOV, *J. Eng. Phys. Thermophys.* (translated from Russian journal *Inzhenero-Fizicheskii Zhurnal*) **63** (5) (1992) 525.
13. A. MERZHANOV, A. M. STOLIN and V. V. PODLESOV, *J. Eur. Ceram. Soc.* **17** (1997) 447.
14. K. MORSI, J. WALL, J. RODRIGUEZ and S. O. MOUSSA, *ASM J. Mater. Eng. Perform.* **12** (2003) 147.

15. Annual Book of ASTM Standards, "Metals-Mechanical-Testing; Elevated and Low-Temperature Tests; Metallurgy" (ASTM 1916 Race Street, Philadelphia, PA, USA, 1995) vol. 3.01, p. 236.
16. J. BYSTRZYCKI and R. A. VARIN, *Intermetall* **6**(4) (1998) 277.
17. G. E. DIETER, "Mechanical Metallurgy," Metallurgy and Metallurgical Engineering Series (The Mapel Press Company, York, PA, USA, 1961) p. 514.
18. EXTRUSION, in ASM Specialty Handbook, Al and Al-Alloys (The Materials Information Society), edited by J. R. Davis and Associates (1993) p. 262.
19. W. MISIOLEK and R. G. GERMAN, *Mater. Sci. Eng.* **A144** (1991) 5.

*Received 3 May  
and accepted 2 July 2004*