JOURNAL OF MATERIALS SCIENCE **39** (2004) 4849–4854

Combustion forging of FeAl (40 at.% Al)

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

J. RODRIGUEZ
Procter & Gamble, P.O. Box 400, Cape Girardeau, MO 63701, USA

This paper presents work on combined combustion synthesis and forging of FeAl from elemental powders. The process can be used to simultaneously form and shape intermetallics with reduced external energy inputs. The effect of varying processing parameters including forging temperature and forging strain on the developed microstructure and hardness is presented. Results confirm the feasibility of this new approach in forging Fe-40 at.% iron aluminide.

1. Introduction

Iron aluminide intermetallics have recently attracted consideration for a large number applications including dies for superplastic forming, heating elements, exhaust manifolds and support hangers [1] which often necessitate the use of thermomechanical processing. It is well known that sintered powder metallurgy parts are often forged in order to improve their mechanical properties. This is believed to be an unnecessary and extra processing step that can be eliminated in the case of materials that can be combustion synthesized from elemental powders. The combustion synthesis process utilizes chemical energy generated during a reaction to process materials with minimal external energy inputs and in very short processing times. Recently it has been applied to iron aluminides as a low-energy alternative to their processing [2, 3]. In combustion synthesis of iron aluminides in the thermal explosion mode of ignition, elemental powders of iron (Fe) and Aluminum (Al) are mixed in the desired composition, compacted and then heated to initiate the exothermic reaction that forms the intermetallic (e.g., FeAl). Despite its energy-saving advantage this method often results in porous microstructures (and in some cases in-homogeneities) that require further high temperature consolidation [4]. Since during the reaction, the product is raised to very high temperatures, a high-temperature transient window of short duration exists in which these materials can be malleable and most suitable for processing. Deformation during this period should shape the product while achieving a higher degree of consolidation compared to just simultaneously deforming and reacting the elemental powder compact [5]. In essence the approach can be used to forge aluminides at operating temperatures $\sim$400°C lower than would be required for the forging of powder metallurgy intermetallic parts (which would also need to be sintered prior to forging). Combustion forging also avoids the extra processing step of heating the *already reacted* and cooled compact again to an elevated temperature (often in excess of $\sim$1000°C [6, 7]) and then applying deformation. While the application of pressure during the reaction has been extensively studied in the form of hot pressing, hot isostatic pressing and pseudo-hot isostatic pressing for aluminides [8–10], systematic work on bulk deformation and shaping during combustion synthesis has not been covered as much for intermetallics. Recently a number of interesting studies were conducted on what the authors termed reactive forging of interpenetrating phase composites [11, 12] and Ti_3SiC_2 based ceramics [13] with the aim of producing highly dense final products. In these studies green powder compacts were placed between two heated rams (also used to heat the compact) and a predetermined pressure (load control) was applied immediately after the onset of thermal explosion. The studies however did not systematically consider the effect of forging at different *product* temperatures while the specimen was cooling or forging to different predetermined strains, all of which can have an effect on the developed microstructures. LaSalvia *et al.* [14, 15] studied the combustion synthesis/impact forging of TiC and TiC-Ni based cermets using forging speeds 10–15 m/s. In these experiments a gas driven high-speed forging machine was used, and the specimen was placed on a heated anvil and the reaction initiated. After a certain time delay, the forging stroke was applied. This method uses SHS (self-propagating high temperature synthesis) in which the compact is reacted at one end and a combustion wave travels through the compact. Therefore, at the onset of forging the temperature distribution within the specimen would not be uniform. In the present work we are concerned with thermal explosion or volume combustion synthesis (VCS) [16] where the whole volume of the compact is simultaneously

*Author to whom all correspondence should be addressed.

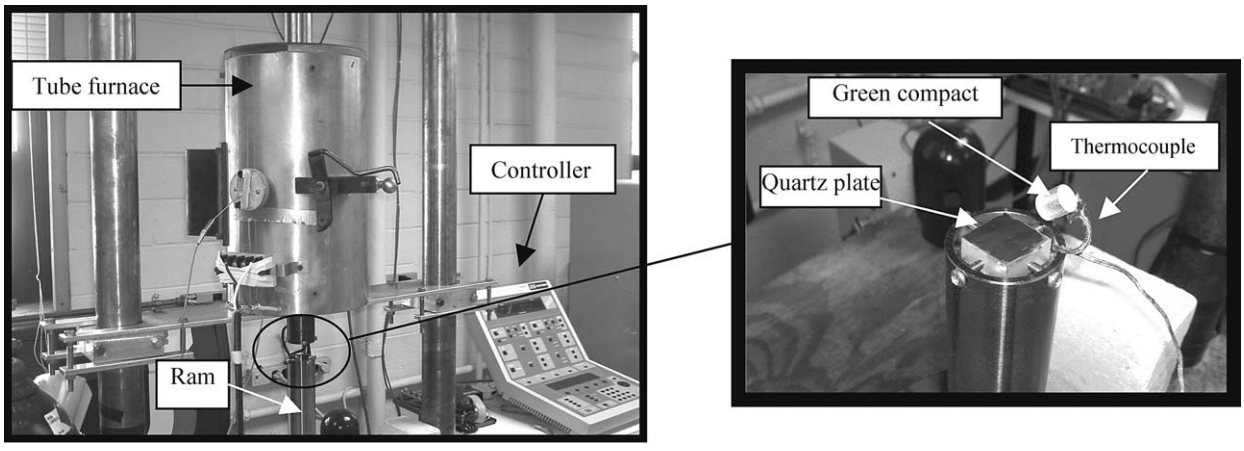

Figure 1 Experimental setup for combustion forging experiments.

heated to the reaction temperature. Also some materials such as iron aluminides (which are very sensitive to strain rate during forming [17]), may not be suitable for impact forging. In these experiments as mentioned, a time delay was specified after the reaction was initiated to allow for the reaction to go to completion. A time delay of 20 s for example was found to result in cracking [14]. Although time delay is significant in the initial stages of the reaction where reaction kinetics play a role in converting the reactants to product(s) and microstructural development, the temperature (which is an important parameter in conventional forging) as the specimen cools from the combustion temperature would be a more suitable parameter to use in combustion forging as opposed to time delay. Ideally, one would apply the forging stroke at specified temperatures during the cooling stage, and hence a higher temperature would dictate a lower flow stress and higher ductility . . . .etc. Although the above mentioned studies provide important contributions, a controlled systematic study of simultaneous combustion synthesis and forging of aluminides covering the conventional forging parameters is needed. An experimental setup for combustion forging was recently reported by Morsi *et al.* [18] which allows for the variation and monitoring of forging processing parameters. In that work however, only the experimental verification of the combustion forging process was presented, with hardly any variation in processing parameters. The present work therefore provides an insight into the effect of combustion forging processing parameters on the developed microstructure and hardness of FeAl (40 at.% Al—one of the FeAl compositions considered for structural use [19]).

2. Experimental procedure

Elemental powders of iron (<45 micron, 99.9% pure)[1] and aluminum (<45 micron, 99.9% pure)[2] were turbula mixed for 30 min in the composition of FeAl (40 at.% Al). The mixed powders were then compacted into cylinders approximately 11.3 mm in diameter and 11.3 mm high with green densities approximately 67, 77 and 84% of theoretical green density (TGD). All compacts were vacuum degassed at 300°C for 12 h in order to remove any low boiling point species adsorbed on the powder surface that may evolve during the reaction and cause unwanted porosity. Each pellet was placed between two quartz plates connected to two vertical rams, and the whole arrangement surrounded by a vertical tube furnace and heated (at a rate of ∼10°C/min) to the ignition temperature under an argon atmosphere. A K-type (Chromel-Alumel) thermocouple was imbedded into the specimen at half height. The experimental arrangement is shown in Fig. 1. All specimens were combustion synthesized using the same setup without applying any deformation in order to select a suitable compact green density that yields a high combustion temperature (maximum temperature during the reaction) and suitable temperature-time profiles. Repeat experiments verified that the combustion temperatures were reproducible to within ±25°C. For the combustion forging experiments, a computer-controlled setup with rapid response sensors was used to allow the control of the onset of deformation at a specified temperature along the temperature-time profile. In our current study, for the 77% TGD specimens, the forging was allowed to commence at temperatures from 800 to 1100°C as the specimen was cooling down from the maximum combustion temperature and using an average strain rate of 0.5 s^{-1} to an engineering strain of 0.5. Also combustion forging to a range of engineering strains (0.1–0.7) (i.e., 10–70% height reduction) was conducted at an average strain rate of 0.5 s^{-1} (by varying the ram speed) and 1000°C during cooling. Graphite was used as the lubricant. All specimens were sectioned centrally along the forging axis for microstructural characterization. The effective consolidation (porosity content) was measured by conducting image analysis on the central region of the cross-section (five areas were investigated for each specimen). Microstructural and compositional analyses were conducted using SEM/EDS (AMRAY-600). Phase analysis was carried out using X-ray diffraction (XRD) (Scintag PADV diffractometer). Hardness was measured using the A-scale on a Rockwell hardness indenter (Buehler Ltd.) (ten measurements for each region were taken and the average calculated and reported). To reveal the microstructure,

[1,2]Atlantic Equipment Engineers (AEE), NJ 07621, USA.

specimens were etched using an etching agent of composition (5% HNO_3, 5% HF, 75% glycerol, 15% water).

3. Results and discussion

3.1. Combustion synthesis

The formation of the FeAl compound from iron and aluminum powders can be preceded with the formation of transitional phases during the heating of elemental powders. However, the complete conversion to FeAl would have already occurred as the specimen cools from the combustion temperature [20, 19]. Therefore it is a logical conclusion that the forging should be performed as the specimen is cooling and not before reaching the peak combustion temperature. This assumes prior optimization of processing parameters such as green density and elemental particle size so that the elemental powder would transform to ∼100% intermetallic prior to the forging stroke. In our initial experiments we varied the green density from 67 to 84% TGD. Each pellet was then combustion synthesized using the same arrangement used for reaction forging but without actually forging.

The ignition for all specimens was found to initiate at temperatures close to the melting point of aluminum and slightly above the eutectic temperature of 652°C, and the heat of reaction heated the compact to the peak combustion temperature. Some studies previously reported onset of ignition close to the eutectic temperature of Fe-Al [3]. Also recently dilatometer studies showed that two exothermic peaks where observed during the heating of FeAl. One attributed to the formation of Fe_2Al_5 at temperatures as low as 560°C and is accompanied by an expansion of the green compact and the other at ∼655°C close to both the eutectic temperature and melting point of Al [19]. We did not notice any significant exothermic peak at 560°C. Our ignition temperature is however very close to the melting point of Al and therefore the melting of Al is very much an intricate part of the reaction. The combustion temperatures for the 84% TGD, 77% TGD and 67% TGD specimens were 1072, 1174 and 797°C respectively. As indicated, the ∼77% TGD specimen yielded the highest combustion temperature of ∼1174°C and therefore is expect to show the highest intermetallic yield i.e., $Fe + Al \rightarrow FeAl$. This value is also slightly lower than a reported value of 1220°C for Fe-50 at.% Al [21], due to a slightly greater enthalpy of formation for Fe-50 at.% [22] in addition to higher heat losses to the tooling and flowing argon for our material/system setup. XRD revealed that the 67% TGD reacted specimen contained a large number of phases including FeAl, Fe_3Al, $FeAl_2$, Fe_2Al_5 and un-reacted iron. The combustion synthesis process in aluminides is often facilitated by the formation of a transient liquid phase (eutectic/aluminum), this is also the case for the Ni-Al system, where a eutectic has been shown to trigger the reaction [23]. The process could be classified as a reactive transient liquid phase sintering process [24]. The transient liquid phase then spreads by capillary action throughout the compact encapsulating the iron particles [24]. The spreading of the aluminum phase is essential in ensuring proper Al/Fe interaction leading to a complete reaction and relative consolidation via capillary forces. At high porosity levels (as observed for the 67% TGD specimen), the number of contact points between elemental powders is low and in addition, the capillary forces are also low due to the excessive porosity. This appears to have resulted in an incomplete reaction, with a lower heat generation rate after ignition signified by a lower slope on the temperature–time profile compared to the 77% TGD specimens (Fig. 2) and lower combustion temperature. Also it shows that at the lower green density of 67% TGD the combustion temperature is reached after a longer time following ignition. The decrease in combustion temperatures due to decrease in green densities has also previously been reported for the reaction synthesis of both iron and nickel aluminides [3, 25].

X-ray diffraction carried out on the 77 and 84% TGD specimens confirmed full conversion of elemental powders to FeAl. Since the FeAl diffraction peaks are close to those of iron XRD data were also confirmed using SEM (back scattered mode) and EDS analysis. The reduction in combustion temperature for the 84% green

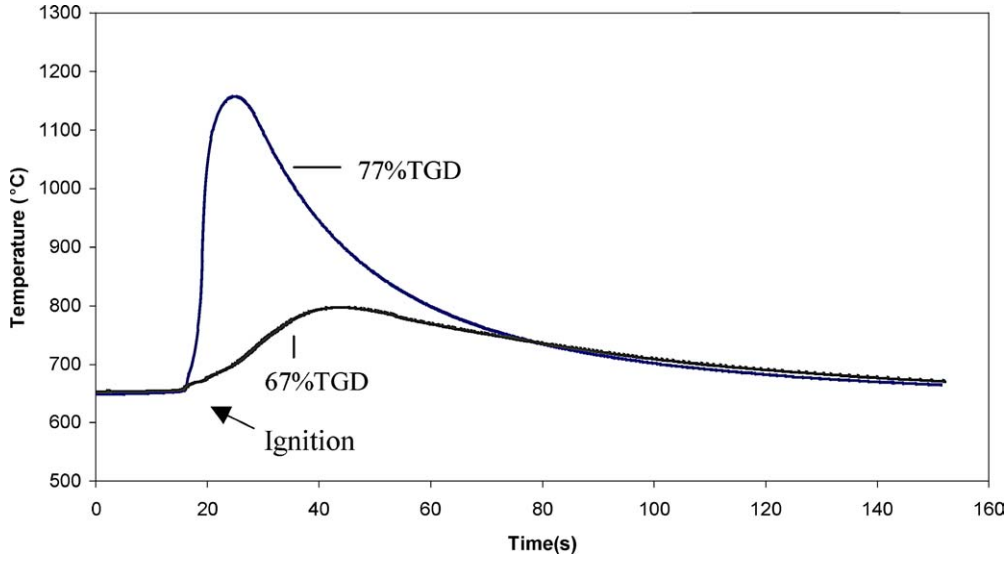

Figure 2 Synchronized temperature profiles for combustion synthesized FeAl (actual data).

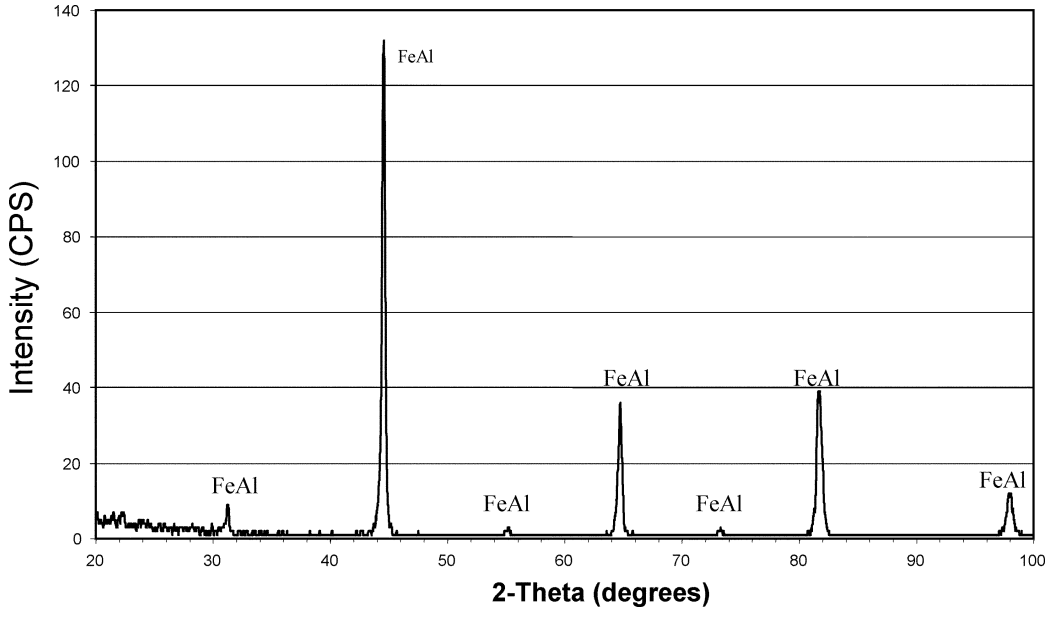

Figure 3 XRD plot of FeAl produced by combustion forging at 1000°C to 0.5 strain and at 0.5 s^{-1} average strain rate.

density is therefore believed to be due to higher heat losses from the compact (a possible density/compact thermal conductivity effect). Combustion forging experiments were subsequently conducted using green pellets of 77% TGD specimens. The temperature-time profile (Fig. 2) shows that it takes ∼25 s for the temperature of the compact to decrease from the peak temperature to a temperature of 800°C, facilitating the investigation of effect of forging temperature. For larger compacts it is expected that this forging regime could run into minutes due to a lower surface area/volume ratio and therefore lower heat losses.

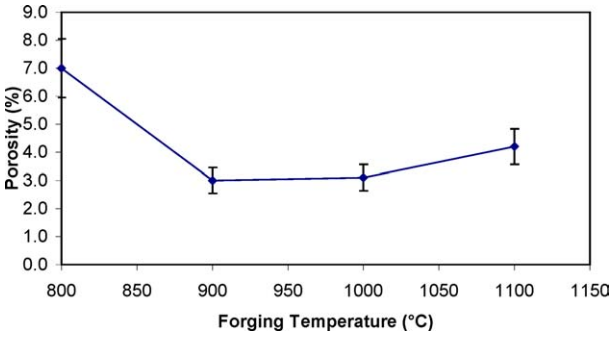

Figure 4 Effect of forging temperature on porosity (0.5 strain rate and 0.5 strain).

3.2. Combustion forging

Fig. 3 is an XRD plot of a combustion forged FeAl specimen (forged at 1000°C to 0.5 strain and at 0.5 s^{-1} strain rate). EDS analysis also revealed a bulk composition of Fe-40.45 Al (±2) at.%.

By simultaneously heating the whole compact to the ignition temperature (VCS) as opposed to locally igniting the compact mixture (SHS), the maximum temperature obtained during the reaction (the combustion temperature) is increased, since the heat of reaction is used mainly in heating the product as opposed to being partly consumed in heating the reactants to the ignition temperature in SHS. For our system, this facilitates the examination of a larger temperature range. The effect of forging temperature (during cooling from the combustion temperature) on the % residual porosity of specimens reaction forged to 0.5 strains at 0.5 strain rate is presented in Fig. 4. It can be seen that consolidation was more effective at forging temperatures of 900°C and above, possibly a result of the increased ductility at these temperatures. Although there appears to be a slight increase in porosity at 1100°C, the error bars overlap data at lower temperatures (900 and 1000°C) and therefore such conclusions can not be decisively drawn. It has previously been recommended that hot working of iron aluminides at strain rates similar to ours be conducted at temperatures between 950–1150°C for "good" (crack free) processing [26]. It is believed that increased ductility at temperatures of 900°C and above facilitates pore closure.

The grain size of the reacted but unforged material was 13.2 μm. The microstructures of the combustion forged materials were however very difficult to reveal through etching. The etched microstructure of the combustion forged specimens contained some regions which appeared to be preferentially over-etched, while other regions of the microstructure were easier to reveal. It is possible that the over-etched regions may be un-recrystallized areas with high dislocation density, however further work needs to be conducted in this respect. The grain size of the well etched regions was measured in order to gain a qualitative evaluation of recrystallization (no less than fifty grains were counted for each region). The specimen combustion forged at 800°C contained equiaxed grains with an average grain size of 3.9 μm, a marked decrease compared to the un-forged specimen, suggesting recrystallization. In a recent study by Zhao *et al.* [27], forging FeAl (40 at.% Al) at 800°C yielded dynamically recrystallized grains 3–5 μm in size within the recrystallized regions, comparing well with our findings. Higher combustion forging temperatures prompted an increase in grain size due to grain growth subsequent to recrystallization such that forging temperatures of

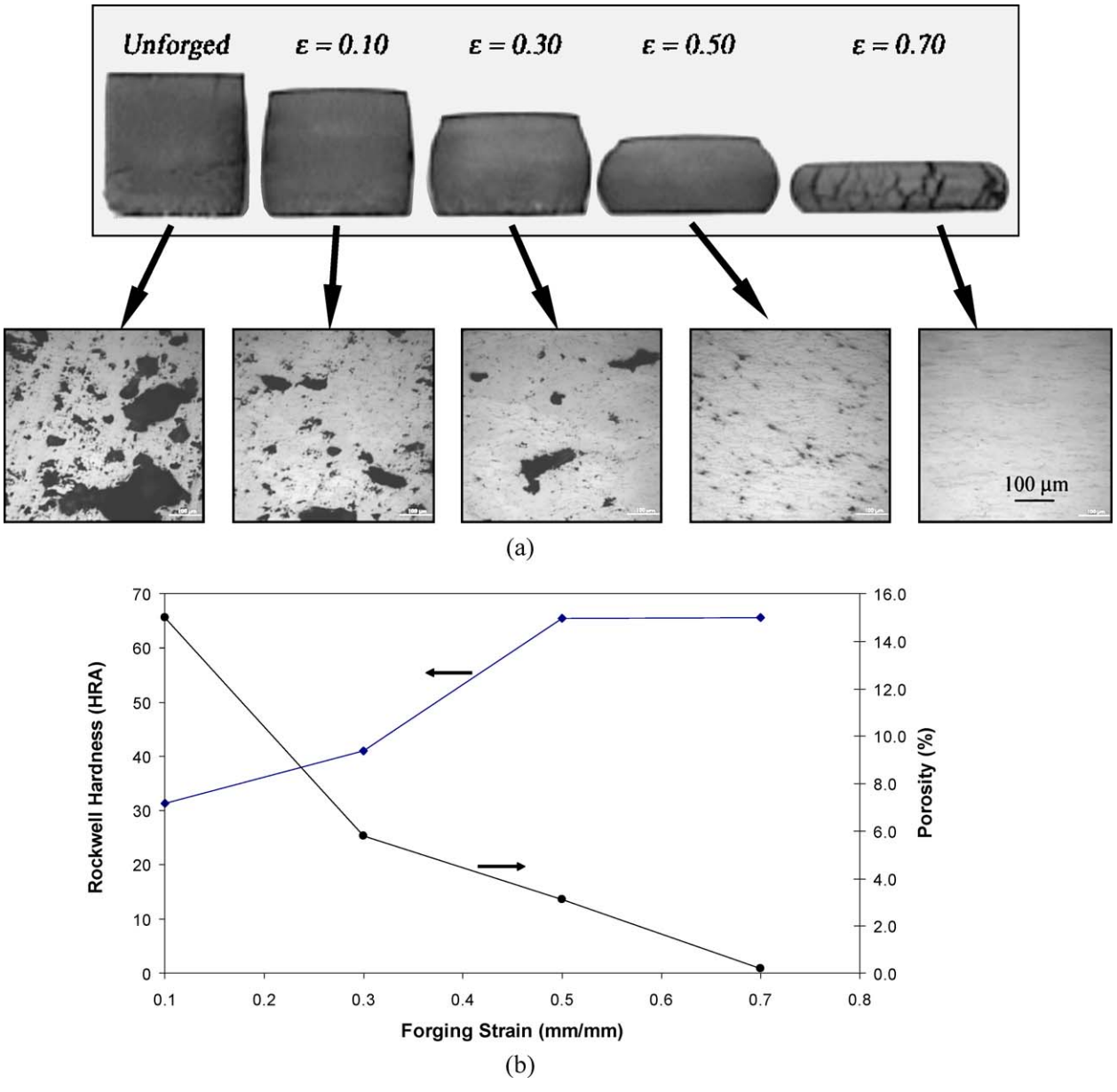

Figure 5 (a) Combustion synthesized and combustion forged specimens (forging strain (ε) 0.1 to 0.7) also showing microstructures with corresponding porosity reduction with strain and (b) effect of forging strain on Rockwell hardness and % area porosity.

1000°C and 1100°C resulted in grain sizes of 6.5 and 11.7 μm respectively. The development of subgrain structure was recently reported for FeAl alloy that had undergone conventional forging to 45% height reduction, at 1000°C and 1 s^{-1} strain rate [27], subgrain structures were also observed in our specimen forged at 1000°C to 50% height reduction. The development of subgrain structure and dynamic recrystallization above 750°C for FeAl (40 at.% Al content) has been recently attributed to the change in deformation mechanism from a ⟨111⟩-type superdislocations to a ⟨100⟩-type perfect dislocations giving rise to considerable refinement of the microstructure through an increased ability for dislocation climb [28].

To investigate the effect of forging strain, combustion forging was conducted at the same forging strain rate of 0.5 s^{-1} (achieved by varying the corresponding ram speed) and temperature of 1000°C to different strains (height reductions) (Fig. 5).

The forgings were generally sound and crack-free with relatively good surface finish. Severe cracking was however observed at 0.7 engineering strain, showing a strain limit at the current forging conditions (Fig. 5a). It was also clear that forging was effective in consolidation, porosity decreasing with increase in forging strain (Fig. 5b). Hardness is also seen to increase with forging strain as a result of densification. An important outcome of our work and experimental approach is that in addition to consolidation, Fe-40 at.% Al iron aluminides can be successfully forged during the high temperatures achieved in combustion synthesis. This experimental approach should be applicable to other aluminides and intermetallics with future work addressing the closed-die combustion forging of intermetallics into more complex shapes.

4. Summary and conclusions

Forging was successfully applied during combustion synthesis to produce Fe-40 at.% Al aluminide. An initial green density of 77% TGD provided the highest combustion temperature and good forging

characteristics. Porosity was found to decrease with increase in forging strain, leading to an increase in hardness, with severe cracking occurring at a forging strain of 0.7 (signifying a strain limit at the specified forging conditions). Consolidation was more effective at forging temperatures of 900°C and above. Recrystallization in combustion forged microstructures was evident allowing grain size refinement compared to just combustion synthesis without forging.

Acknowledgement

The authors would like to thank Mr. S.O. Moussa for the Rockwell hardness measurements.

References

1. V. K. SIKKA and C. T. LIU, *Mater. Tech.* **9**(7/8) (1994) 159.
2. D. L. JOSLIN, D. S. EASTON, C. T. LIU and S. A. DAVID, *Mater. Sci. Eng.* A **A192/193** (1995) 544.
3. B. H. RABIN and R. N. WRIGHT, *Met. Trans.* A **22A** (1991) 277.
4. K. MORSI, *Mater. Sci. Eng.* A **A299** (2001) 1.
5. K. MORSI, H. B. MCSHANE and M. MCLEAN, *Metall. Mater. Trans.* A **31A** (2000) 1663.
6. R. BOWMAN, R. NOEBE and S. RAJ, *Met. Trans.* **23A** (1992) 1493.
7. V. K. SIKKA, in "Physical Metallurgy and Processing of Intermetallic Compounds," edited by N. S. Stoloff and V. K. Sikka (Chapman and Hall, NY, 1996) p. 588.
8. H. DOTY and R. ABBASCHIAN, *Mater. Sci. Eng.* A **A195** (1995) 101.
9. D. ALMAN and N. S. STOLOFF, *Int. J. Powd. Metall.* **27**(1) (1991) 29.
10. K. MATSUURA and M. KUDOH, *Mater. Sci. Eng.* A **A239/240** (1997) 625.
11. D. HORVITZ, I. GOTMAN, E. Y. GUTMANAS and N. CLAUSSEN, *J. Euro. Ceram. Soc.* **22** (2002) 947.
12. D. HORVITZ and I. GOTMAN, *Acta Mater.* **50** (2002) 1961.
13. Y. KHOPTIAR and I. GOTMAN, *J. Euro. Ceram. Soc.* **23** (2003) 47.
14. J. C. LASALVIA, D. K. KIM and M. A. MEYERS, *Mater.Sci. Eng.* A **206** (1) (1996) 71.
15. L. C. LASALVIA, M. A. MEYERS and D. K. KIM, *J. Mater. Synth. Proc.* **2**(4) (1994) 255.
16. A. VARMA and A. S. MUKASYAN, "Combustion Synthesis of Advanced Materials," in Powder Metal Technologies and Applications, ASM Handbook, (ASM International, 1998) Vol. 7, p. 523.
17. T. TESTANI, A. DI GIANFRANCESCO, O. TASSA and D. POCCI, in Proceedings of the International Symposium on Nickel and Iron Aluminides: Processing, Properties and Applications, edited by S. C. Devi, P. J. Maziasz, V. K. Sikka and R. W. Cahn, (Materials Park, OH, ASM International, 1996) p. 213.
18. K. MORSI, J. WALL, J. RODRIGUEZ and S. O. MOUSSA, *ASM J. Mater. Enging. Perform.* **12** (2003) 147.
19. S. GEDEVANISHVILI and S. C. DEEVI, *Mater. Sci. Eng.* A **325** (2002) 163.
20. C. R. KACHELMYER, A. VARMA, I. O. KHOMENKO, A. S. ROGACHEV and A. G. MERZHANOV, Thermodynamics of Phase Transformations, Materials Research Society Symposium Proceedings, (Materials Research Society, Pittsburgh, PA, USA, 1996) Vol. 398, p. 593.
21. B. H. RABIN and R. N. WRIGHT, *Met. Trans.* A **22A** (1991) 277.
22. R. M. GERMAN, in "Thermal Analysis in Metallurgy," edited by R. D. Shull and A. Joshi (The Minerals, Metals and Materials Society, Warrendale, PA, 1992) p. 205.
23. W. MISIOLEK and R. M. GERMAN, *Mater. Sci. Eng.* A **A144** (1991) 1.
24. RANDALL and M. GERMAN, "Liquid Phase Sintering," (Plenum Pub Corp, New York, 1985) p.164, 172.
25. J. J. MOORE and H. J. FENG, *Progr. Mater. Sci.* **39** (1995) 260.
26. Y. V. R. K. PRASAD, D. H. SASTRY and S. C. DEEVI, *Intermetallics* **8** (2000) 1074.
27. P. ZHAO, D. G. MORRIS and M. A. MORRIS-MUNOZ, *J. Mater. Res.* **14**(3) (1999) 715.
28. R. IMAYEV, E. EVANGELISTA, O. TASSA and J. STOBRAWA, *Mater. Sci. Eng.* A **A202** (1995) 128.

Received 25 August 2003
and accepted 21 April 2004