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The synthesis of bulk material through explosive compaction for making intermetallic compound Ti_5Si_3 and its composites

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The possibility of synthesizing Ti_5Si_3 from mixed elemental powders and the fabrication of its composites by explosive compaction is discussed. A new technique using underwater shock waves was developed and it was found to exercise better control over the influencing parameters. Two processes were employed viz., (1) direct shock-induced reaction and (2) explosive compaction followed by heat treatment. The methodology to produce bulk material by the above two processes are reported. Ti_5Si_3 intermetallic synthesized by the two processes reveals high hardness than commercially available Ti_5Si_3.

1. Introduction

The use of explosive energy and the processing techniques for various industrial materials such as explosively welded clad [1], and shock synthesized diamond [2] have been reported earlier. The possibility of using explosive or shock compaction for the synthesis of intermetallics have been suggested by many researchers [3–7]. Two methods using explosive compaction of powders for synthesizing bulk body of intermetallics have also been reported. One method is to initiate shock-induced reaction as to synthesize intermetallics directly, and the other is to synthesize intermetallics by heat-treatment of the explosively compressed and activated sample. As both processes are expected to produce superior properties resulting from microstructural refinement, the production of bulk material using such unique processes is of high research and commercial interest. Fabrication of intermetallics using SHS (Self-propagation High-temperature Synthesis) [8, 9] is not discussed, since the process deviates from explosive compaction.

Many difficulties are often encountered during the production of crack-free bulk material intermetallics. The problems are as follows, (1) propagation and interaction of strong shock waves which induce cracks, (2) excessive exothermic heat release induced by reaction, and (3) volumetric change due to reaction. The present investigation intends to establish a process for synthesizing various bulk bodies of intermetallics and their composites, without cracks, through explosive compaction techniques. The authors employed a method different from Chiba's method [10] of using underwater shock waves. Based on a series of experiments the way to optimize the parameters and the technique of making large-sized sample using explosive are discussed.

2. Assembly design and the experimental procedure

The schematic illustration of the assembly for explosive compaction is shown in Fig. 1. The water container has a straight cylindrical shape, unlike the converging design proposed by Chiba *et al.* [10]. In Chiba's experiments a converging high pressure is used for compacting difficult-to-consolidate powders, but such converging high pressure is not required for the present investigation as low pressure is adequate to consolidate mixed element powders or igniting shock-induced reaction. The advantage of the present method is that the control of the applied pressure is simple and is effected by changing the thickness of the water column t_w. The present investigation employed an SEP explosive which has stable detonation velocity independent of the size.

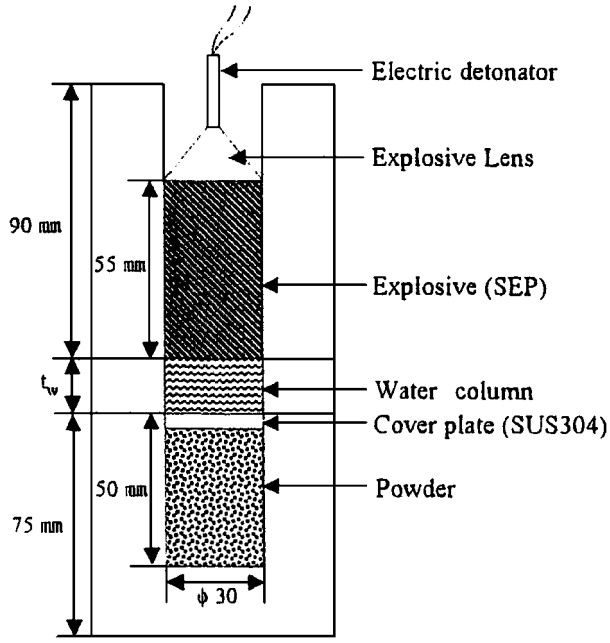

Figure 1 Experimental apparatus.

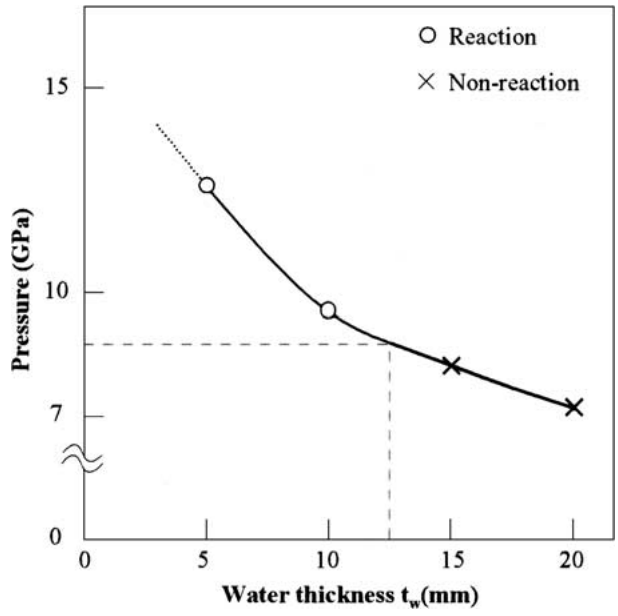

Figure 3 Change in maximum underwater shock pressure with water column thickness t_w.

The main explosive, SEP, was detonated using plane-wave generator composed of two explosives, SEP and HABW. The explosives SEP and HABW are produced by Asahi Chemical Industry Co., Ltd., The detonation velocity and the density are 6.97 km/s and 4.75 km/s, and 1310 kg/m^3 and 2200 kg/m^3 respectively. Without using the plane-wave generator, only a minor difference in the time of affecting the shock pressure to the powders resulted in the curved cross-sectional area due to the spherical propagation of shock front from the ignited position. The wall of the powder container was well lubricated as the friction between the wall and the compacted powders induces macroscopic shear cracks in the cross-sectional area.

An example of the underwater pressure profile just above the powder part and the maximum value of water column thickness t_w are shown in Figs 2 and 3 respectively. These figures were constructed based on a finite-difference analysis developed by Itoh *et al.* [11]. The underwater pressure is about 7.1 GPa at $t_w = 20$ mm and increased with decreasing t_w. The assembly used in this investigation is easy to scale up in diameter, due to the one-dimensional compression system. Increase in the thickness is limited due to the decrease in the shock pressure toward the lower position, but the material from 10 to 30 mm-thick is available choosing the moderate condition for compaction.

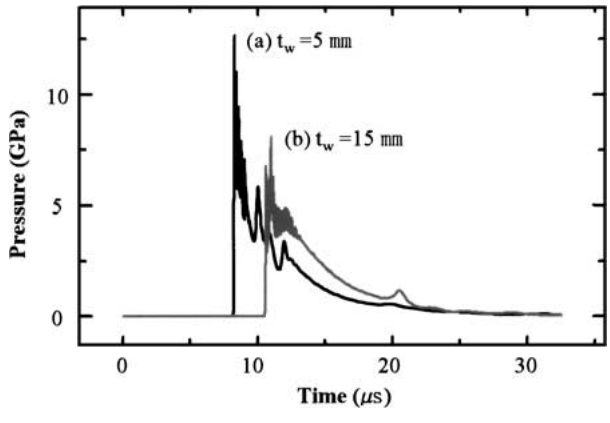

Figure 2 Underwater pressure profile numerically simulated.

The powder container was filled with the mixed element powders at a fixed theoretical density of 0.6. A 2 mm-thick stainless steel cover plate was placed on the powder part and a Ti powder layer was packed beneath the mixed elemental powder layer for successful recovery. In the present investigation, a titanium silicide (Ti_5Si_3) was focussed as the material to be synthesized. Intermetallic Ti_5Si_3 compound shows high melting temperature (2403 K) and can be used for heat resistant material and other novel applications requiring high hardness, high oxidation-resistance and high thermal conductivity [12, 13]. Ti and Si powders were mixed at the atomic ratio 5 : 3. Some of the powders were manually mixed in argon atmosphere while the others were mechanically mixed using a conventional ball mill for 48 h. In few experiments, another inert powder were also admixed with the element powders. The inert powder controls the excessive exothermic reaction heat. Bulk Ti/Ti_5Si_3 and TiAl/Ti_5Si_3 composites were attempted. The later combination has been reported using mechanical alloying (MA) and hot isostatic pressing (HIP) process, and is useful for aerospace materials requiring high temperature strength and toughness [14, 15]. Table I shows the experimental conditions in detail and Table II display the list of the powders used. The materials recovered were characterized using microscope, micro-Vickers hardness test and X-ray diffraction analysis.

3. Results and discussion

3.1. Explosive compaction of Ti and Si powders

Ti and Si powders were compacted at various pressure conditions by changing the thickness of water column t_w. Reaction is induced at high pressure condition over $t_w = 12.5$ mm as listed in Table I. Depending upon the steady detonation velocity of the explosive used, the critical condition of reaction is well defined

TABLE I Experimental conditions and results

Experiment number	Powders (atomic ratio)	Water thickness (mm)	Recovered condition
#T5	Ti : Si (5 : 3)	5	Porous, fully reacted
#T10	Ti : Si (5 : 3)	10	Porous, fully reacted
#T15	Ti : Si (5 : 3)	15	Good compaction, non-reacted
#T20	Ti : Si (5 : 3)	20	Good compaction, non-reacted
#TT1	Ti : Si (5 : 3) (50 mass%) + Ti (50%)	5	Good compaction, fully reacted
#TA1	Ti : Si (5 : 3) (60 mass%) + TiAl (40%)	5	Porous, fully reacted
#TA2	Ti : Si (5 : 3) (50 mass%) + TiAl (50%)	5	Good compaction, partially reacted
#TA3	Ti : Si (5 : 3) (40 mass%) + TiAl (60%)	5	Good compaction, partially reacted
#TA4	Ti : Si (5 : 3) (30 mass%) + TiAl (70%)	5	Good compaction, non-reacted

TABLE II Powders employed

Powders	Source	Powder size
Ti	Sumitomo Sitix Corp.	$-45\,\mu$m
Si	CERAC	-325 mesh
TiAl	Sumitomo Sitix Corp.	$-150\,\mu$m

for the mixed powders. The maximum water pressure applied to the powders was estimated as 8.7 GPa.

The cross-section of the recovered sample are shown in Fig. 4a for reacted and b non-reacted states. The microstructure of these samples is shown in Fig. 5. As shown in Figs 4b and 5b, the powders are observed to be successfully compacted to high density form without reaction. The theoretical density of the compacts is 0.97 using Archimedes' method. Such high density is hard to achieve by conventional presses. In the reacted sample, most of the reacted part is not visible (Fig. 4a) and the recovered part shows porous structure (Fig. 5a). The reacted part appears to be molten due to the excessive reaction heat released. The same porous structure has been confirmed in other reports for synthesizing various intermetallics from mixed elemental powders [16]. However, the recovery of the sample becomes increasingly difficult when the cooling time becomes shorter than the pressurization time. Hence, with careful consideration (Fig. 3), the size of the reaction layer was chosen in the order smaller than 150 μm. The experimental results employing such method is discussed in the following section.

The X-ray diffraction patterns of the reacted sample (#T5) is shown in Fig. 6. Only the peaks for suggesting the existence of Ti_5Si_3 are confirmed, and other phases are not confirmed. The micro-Vickers hardness measurement at the reacted solid part without pores under load of 0.25 N shows 10.2 GPa which is higher than the hardness of commercially available Ti_5Si_3. Such high hardness is also reported by shock synthesized (shock compressed and heat treated) Ti_5Si_3 [12, 17]. The result suggests that the process employing explosive can refine the grain size showing superior mechanical property.

3.2. The fabrication of Ti_5Si_3 composites

For making a shock-synthesized bulk body of intermetallics through explosive compaction, one possible

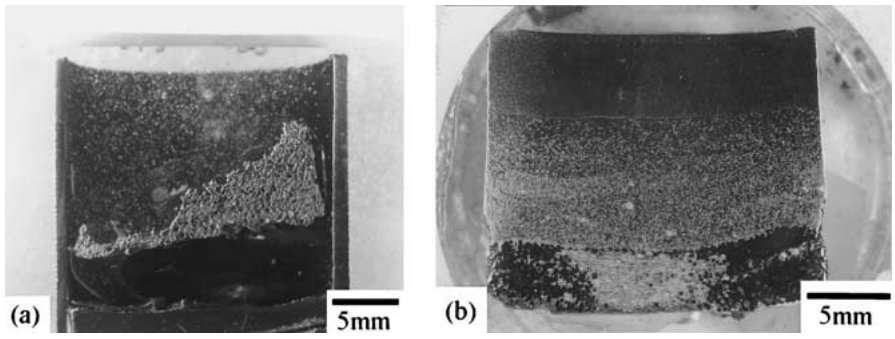

Figure 4 Cross-section of recovered samples. (a) non-reacted (#T20) and (b) reacted (#T5) samples.

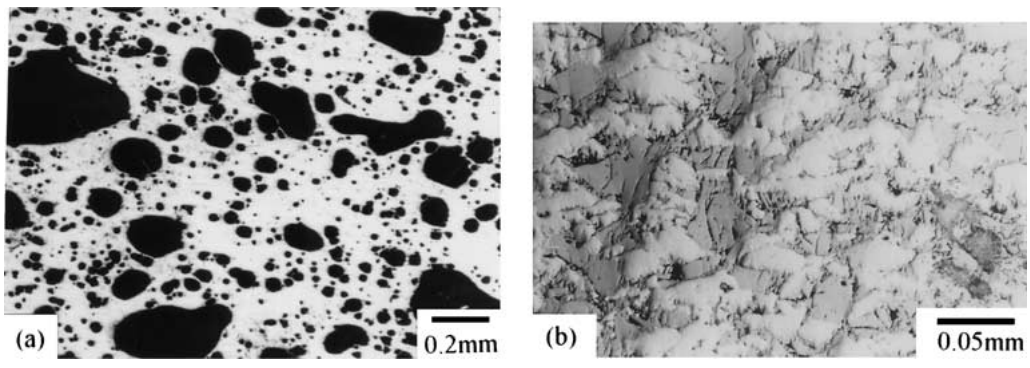

Figure 5 Microstructure of recovered samples. (a) non-reacted (#T20) and (b) reacted (#T5) samples.

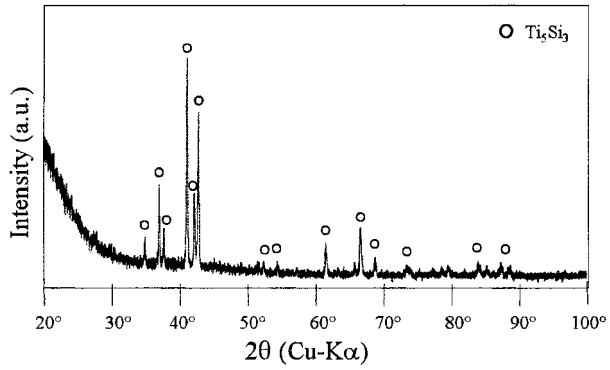

Figure 6 X-ray diffraction pattern for reacted Ti + Si powders mixture through explosive compaction (#T5).

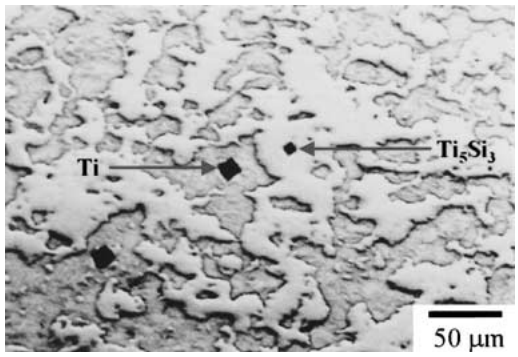

Figure 7 Microstructure of Ti/Ti_5Si_3 composite shock synthesized (#TT1).

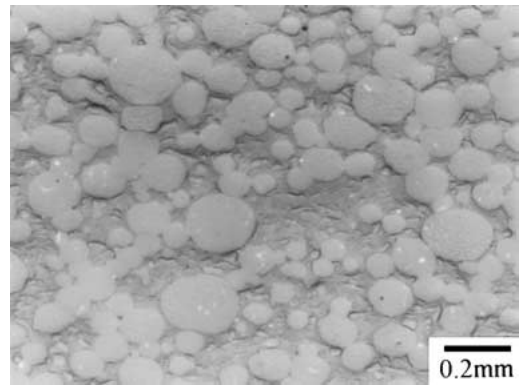

Figure 8 Microstructure of $TiAl/Ti_5Si_3$ composite shock synthesized (#TA3).

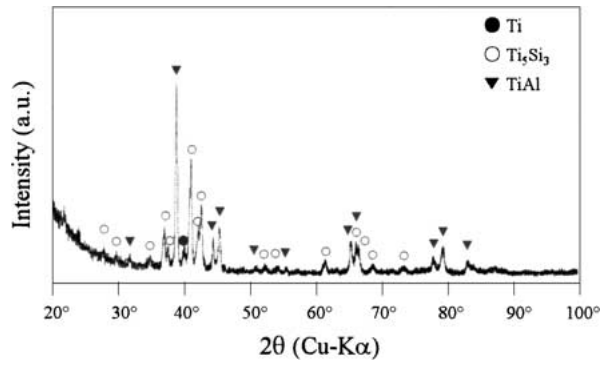

Figure 9 X-ray diffraction pattern of $TiAl/Ti_5Si_3$ composite synthesized (#TA3).

method is to add the other powders as mentioned in the previous section. Ti and TiAl powders were mixed to accommodate the excessive exothermic heat of reaction. Since the size of the elemental powders and the inert powder are restricted, Ti and Si powder mixture is mechanically milled to refine the condition of mixing. The powders are not mechanically alloyed but mixed for a fine size in the order of μm or less.

Some of the experiments were conducted by changing the ratio of mixed reactive powders and inert powders. The ratio of reactive powder was high enough to induce reaction. Otherwise, the reaction is not induced due to the lack of energy (Table I). In other experiments, the mixed reactive powder is packed over the layer to be a composite to enhance reaction in the composite part.

A microstructure of Ti/Ti_5Si_3 composite is shown in Fig. 7. It is evident that an uniform microstructure is obtained after compaction. The white area in Fig. 7 is Ti_5Si_3 synthesized and the traces of Vickers penetrator under load 0.25 N suggest the formation of high hardness material. The average Vickers hardness measured was 10.2 GPa thus indicative of superior mechanical property.

The microstructure of the $TiAl/Ti_5Si_3$ is shown in Fig. 8. The reactive mixed powder part is reacted making Ti_5Si_3 but a weak peak of unreacted Ti is confirmed through X-ray diffraction as shown in Fig. 9. One possibility is that the mixing condition is not uniform everywhere. Although the reaction is not fully terminated, the hardness in the reacted area shows higher values ranging from 9 to 11 GPa.

A bulk body of Ti_5Si_3 using Ti_5Si_3 powders (with Ti and Si powder mixture) through the present method is feasible. However the authors did not make any experimental attempt as the properties of commercially available Ti_5Si_3powders are unsatisfactory.

3.3. Synthesis of intermetallic compound and composite for shock-activated sample

There is another route to synthesize intermetallics using a explosive compaction followed by the heat treatment. Thadhani *et al.* [17] reported the advantage of the this method and synthesized high-hardness Ti_5Si_3 through this technique. Some of the non-reacted Ti + Si samples were heat treated but the reacted microstructure shows porous structure as shown in Fig. 10 due to diffusion and

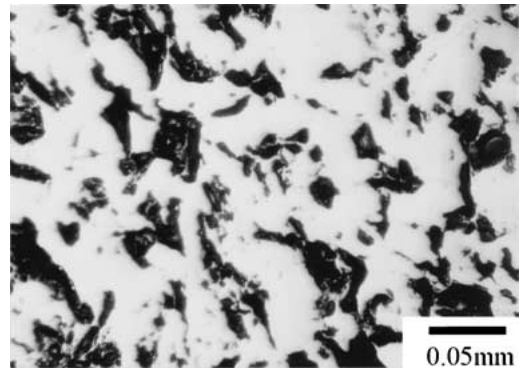

Figure 10 Porous structure found in the explosive compacted and heat treated (at 1273 K for 4 h) sample (#T20).

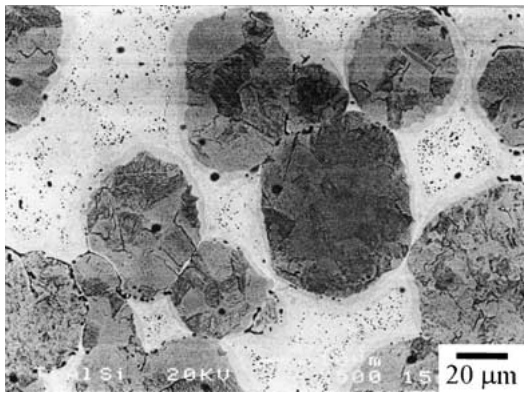

Figure 11 Microstructure of explosive compacted and heat treated (at 1273 K for 1 h) TiAl/Ti_5Si_3 composite (#TA4).

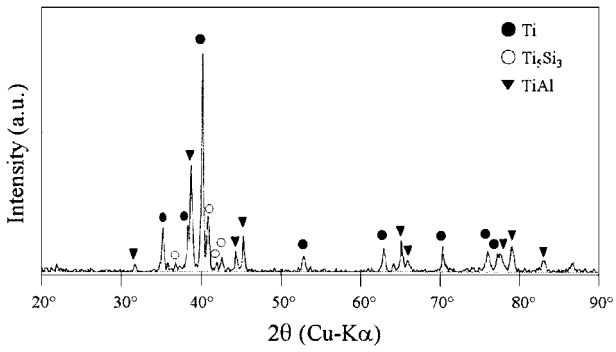

Figure 12 X-ray diffraction pattern of explosive compacted and heat treated (at 1273 K for 1 h) TiAl/Ti_5Si_3 composite (#TA4).

the volumetric change. The use of mechanical milling or alloying is one of the method to solve the problem and Thadhani *et al.* [13] suggested the possibility of making bulk material without porosity using mechanically alloyed powder.

The reaction of the powders is generated above 1173 K. The temperature is lower than the reaction temperature for synthesizing Ti_5Si_3 (1373 K) [17]. Thadhani *et al.* [17] reported the reaction temperature for shock compacted and activated sample and stated that a higher temperature 1573 K is required. The present investigation shows slightly different result of a lower reaction temperature, but this may be due to the different experimental conditions.

TiAl/Ti_5Si_3 composite was also fabricated through explosive compaction and heat treatment. After a heat treatment for a sample (#TA4) at 1273 K for 1 h, Ti and Si was fully reacted for generating Ti_5Si_3. The microstructure is shown in Fig. 11 and the X-ray diffraction pattern is shown in Fig. 12. As seen in the microstructure, the reaction or diffusion between the interface of TiAl and Ti_5Si_3 is confirmed. The X-ray diffraction shows the existence of TiAl (α' and γ phases) and Ti_5Si_3 (Fig. 12). Through a measurement of compositional change, the interfacial area is considered to be composed of some coating layers (Fig. 11). A significant quantity, though less, of Al in TiAl has diffused into Ti_5Si_3 and transformed to a few μm thick $Ti_5(Si,Al)_3$ layer. The decrease in the Al content in TiAl powder layer forms a layer of Ti_2Al (α' phase) and the center position is composed of TiAl (γ phase). The micro-Vickers hardness also confirm the layers formed. It is interesting to note that the thin ternary intermetallic layer $Ti_5(Si,Al)_3$ shows higher hardness than the Ti_5Si_3. The Ti_5Si_3 synthesized after heat treatment also displays very high Vickers hardness above 11 GPa which is expected to have superior mechanical properties than the commercially available ones. Also, $Ti_5(Si,Al)_3$ layer reveals higher hardness (greater than 12 GPa).

The exsistence of Ti phase through reaction can be explained by phase diagram. No α' phase is found, hence the phase should be non-equilibrium phase. Since the heat treatment is performed like a conventional diffusion couple technique, the phase generated is concluded to be α-Ti phase.

4. Conslusions

Shock synthesis by the detonation of explosives is one of the attractive techniques to produce new or difficult-to-get materials with relatively ease and not requiring expensive equipment. The present investigation suggests that a material of superior properties is expected to be obtained due to a very high-velocity phenomena during the process. The main difficulty for making a large-sized sample for industrial use is the safety issue and the precise control of experimental conditions. Recent developments in the measurement and simulation techniques should enlighten the goal to make a real material for industrial application of shock synthesized intermetallics.

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References

1. B. CROSSLAND, "Explosive Welding and its Application" (Oxford University Press, Oxford, London, 1982).
2. P. S. DECARLI and J. C. JAMIESON, *Science* **133** (1961) 821.
3. K. SIVA KUMAR, T. BALAKRISHNA BHAT and P. RAMAKRISHNAN, *J. Mater. Processing Technol.* **85** (1999) 125.
4. N. N. THADHANI, A. H. MUTZ and T. VREELAND JR., *Acta Metall.* **37**(3) (1989) 897.
5. M. A. MEYERS and S. L. WANG, *Acta Metall.* **36**(4) (1988) 925.
6. K. HOKAMOTO, M. FUJITA, S. TANAKA, K. KODAMA and Y. UJIMOTO, *Scripta Metall.* **39**(10) (1998) 1383.
7. T. AKASHI and A. B. SAWAOKA, *J. Mater. Sci.* **22** (1987) 3276.
8. R. TOMOSHIGE, T. GOTO, T. MATSUSHITA, K. IMAMURA, A. CHIBA and M. FUJITA, *J. Mater. Processing Technol.* **85** (1999) 101.
9. R. TOMOSHIGE, Y. KAKOKI, K. IMAMURA and A. CHIBA, *ibid.* (1999) 105.
10. A. CHIBA, M. FUJITA, M. NISHIDA, K. IMAMURA and R. TOMOSHIGE, in "Shock-Wave and High-Strain-Rate Phenomena in Materials," edited by M. A. Meyers, L. E. Murr and K. P. Staudhammer (Marcel Dekker, New York, 1992) p. 415.
11. A. A. AMSDEN, H. M. RUPPEL and C. W. HIRT, SALE, A Simplified ALE computer program for fluid flow at all speeds, LA-8095, UC-32, 1980.

12. T. YAMASAKI, Y. OGINO, K. MORISHITA, K. FUKUOKA, T. ATOU and Y. SYONO, *Mater. Sci. Eng.* A **179/180** (1994) 220.
13. P. J. COUNIHAN, A. CRAWFORD and N. N. THADHANI, *Mater. Sci. Eng.* A **267** (1999) 26.
14. K. P. RAO and Y. J. DU, *ibid.* A **277** (2000) 46.
15. O. N. SENKOV, M. CAVUSOGLU and F. H. FROES, *Mater. Sci. Eng.* A **300** (2001) 85.
16. L. H. YU and M. A. MEYERS, *J. Mater. Sci.* **26** (1991) 601.
17. N. N. THADHANI, S. NAMJOSHI, P. J. COUNIHAN and A. CRAWFORD, *J. Mater. Processing Technol.* **85** (1999) 74.

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