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### Chemical reaction in Ti-Si mixture under controlled high-strain-rate plastic deformation

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Ti-Si powder mixtures were subjected to high strain, high-strain rate plastic deformation (global values:  $\epsilon_r \approx 0.25$  ;  $\dot{\epsilon} = 4 \times 10^4 \text{ s}^{-1}$  ) in a thick-walled cylinder geometry subjected to radial collapse. Low-detonation velocity explosives were the source of energy. Two experimental configurations were used: (a) the plastic deformation was applied to pre-densified powder; (b) plastic deformation was applied to the loose powder (  $\rho \sim 0.65\rho_{th}$  ) and localization of plastic deformation was induced by using a metallic glass layer. Shear localization was the preponderant deformation mechanism in the two cases, with local values of shear strain of up to 100 and shear strain rates of  $\sim 10^7 \text{ s}^{-1}$ . Chemical reactions were initiated at these bands and propagated throughout the entire specimen; only small isolated unreacted regions were left, close to the copper cylinder walls. These results confirm the importance of shear localization in exothermic chemical reactions between powders.

#### 1. INTRODUCTION

Chemical reactions under the action of dynamic loading have been the object of considerable study, and the books by Graham [1], Batsanov [2], Horie and Sawaoka [3], and the review by Thadhani [4] present extended treatments on the subject. Bridgman [5] was the first to point on the importance of shear (under quasi-static loading) on chemical reactions and this was reemphasized by Vereshchagin and co-workers [6] and Teller [7]. Under dynamic/shock conditions, Yu et al. [8], Potter and Ahrens [9], and Nesterenko et al. [10] demonstrated the importance of shear strains on reaction. Recent experiments on Nb-Si powder mixtures under controlled and prescribed shear strains have yielded direct evidence of shear-induced reactions [10].

The objective of the research whose results are reported here was to determine whether initiation of chemical reaction within shear localization regions can result in global, generalized reaction. The enthalpies of reactions for the Ti-Si and Nb-Si systems are:



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In the Nb-Si system only partial reaction was observed, in isolated regions within shear bands. The enthalpy of reaction for the Ti-Si system is four times larger, suggesting that under the same deformation conditions, reaction would be much more generalized.

## 2. EXPERIMENTAL PROCEDURES

The experimental procedure is described in detail by Nesterenko et al. [10, 11] and will only be presented here in succinct form. The Ti and Si powders were purchased from CERAC and had sizes of -325 mesh ( $<44\text{ }\mu\text{m}$ ). The thick-walled cylinder method, developed by Nesterenko and coworkers [12, 13], was used to impart the plastic deformation at high strain rates to the powder mixture and, in one of the configurations (Conf. 1), to densify it. Figure 1 shows the two configurations used to generate controlled and prescribed plastic strain in the

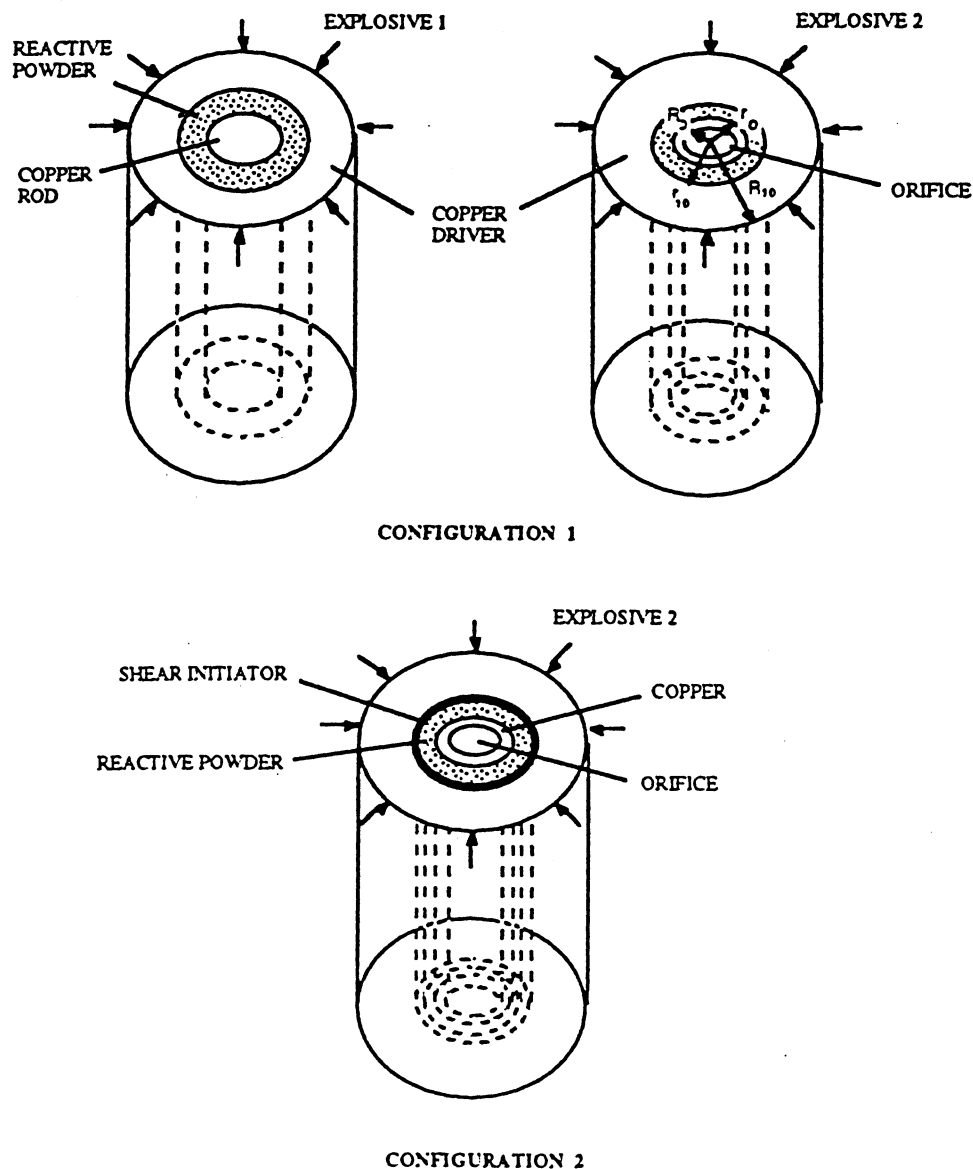


Figure 1. Experimental configurations used to obtain controlled and prescribed plastic deformation in Ti-Si powder mixture.

reactive mixture. In configuration 1, the powder was placed between a copper rod and a copper tube. Explosive was placed around the cylinder and detonation was initiated at the top. A low detonation-velocity explosive was used (2.5 km/s) and the powder was fully consolidated after this stage. The overall plastic deformation was sufficiently small to ensure homogeneous plastic deformation: no shear localization or chemical reaction were observed after this stage (see Section 3). An orifice was then drilled along the axis of the cylinder, and the explosive procedure was repeated, using an explosive with higher detonation velocity (4.1 km/s) to ensure full collapse of the central hole. This second explosive event produced global radial strain of approximately 0.37, along the external radius; The global strain rate was approximately  $4 \times 10^4 \text{ s}^{-1}$ . Under these conditions deformation in copper is homogeneous. Experiments with Nb-Si mixtures [10, 11] revealed, at this stage, profuse shear localization, as was the case in the Ti-Si mixture. In the second configuration (Conf. 2 in Fig. 1), densification and plastic deformation were carried out simultaneously. The central mandrel had a longitudinal hole in it (see Fig. 1(b)) and only one explosive event was carried out. A layer of metallic glass, which is known to deform primarily by shear localization [14], was used along the outer wall of the powder container. The function of this layer was to provide initiation sites for shear localization. This metallic glass layer was obtained by rolling a few layers of amorphous foil along the cylinder walls. Nesterenko et al. [12, 13] observed profuse localization in the Nb-Si mixture after this stage.

It should be noted that the global (shock) energy imparted by the explosive to the powder is considerably below the threshold energy calculated by Meyers et al. [15]:  $\sim 100 \text{ J/g}$ .

### 3. RESULTS AND DISCUSSION

Figure 2 shows the Ti-Si mixture subjected the densification stage (final density: 89% of theoretical value) in Configuration 1. Virtually no reaction was observed. Figure 3 shows the calculated reaction initiation pressure vs. initial density curves for Nb-Si, Mo-Si, and Ti-Si mixtures, assuming that shock energies of 700 J/g (for  $\text{MoSi}_2$  and  $\text{NbSi}_2$ ) and 100 J/g (for  $\text{Ti}_5\text{Si}_3$ ) are required for initiation of reaction [16]. The pressure for the Ti-Si system is 2 GPa for the initial porosity of 35% and  $\sim 4 \text{ GPa}$  for the porosity of 11% (end of Stage 1). In stage 2,

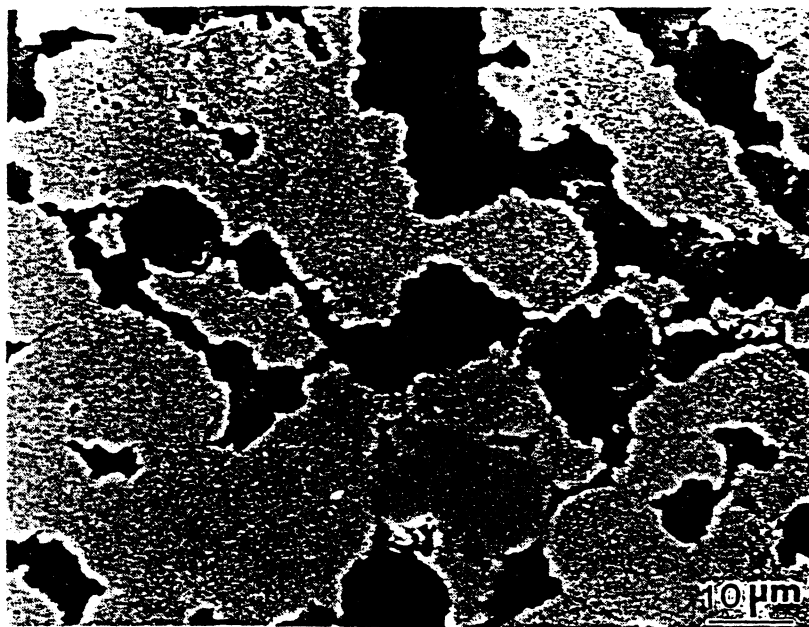


Figure 2. Scanning electron micrograph of Ti-Si mixture subjected to dynamic densification (stage 1) in configuration 1.

the detonation velocity of 4.1 km/s generates, at most, a pressure of 2 GPa. The same can be said for Configuration 2. Thus, the full reaction of the specimens, obtained after stage 2 of configuration 1 and in Configuration 2, and seen in Figure 4, has to be attributed to initiation at shear localization sites. It was possible, only from the steps in the copper container to establish the pattern of shear localization. Figure 5 shows typical shear bands for the Ti-Si and Nb-Si systems. The displacements,  $\Delta$ , are approximately equal. The pattern of shear bands and these spacing (~1-2 mm apart) is similar to those in the Nb-Si system. It can also be seen that, with the exception of localized regions close to the copper, the Ti-Si system is fully reacted. The unreacted regions are marked with arrows. The entire Ti-Si specimen is filled with voids, due to solidification shrinkage and/or gas evolution after reaction. For the Nb-Si system (Fig. 5(b)) the shear band is seen as a clear line (marked by arrow) linking the two steps in copper. The displacement,  $\Delta$ , divided by the thickness of the band provides the shear strain. Shear strains as high as 100 were observed for Nb-Si, and the same values are expected for Ti-Si.

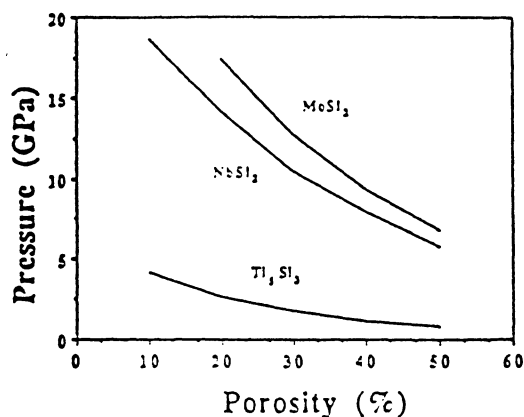
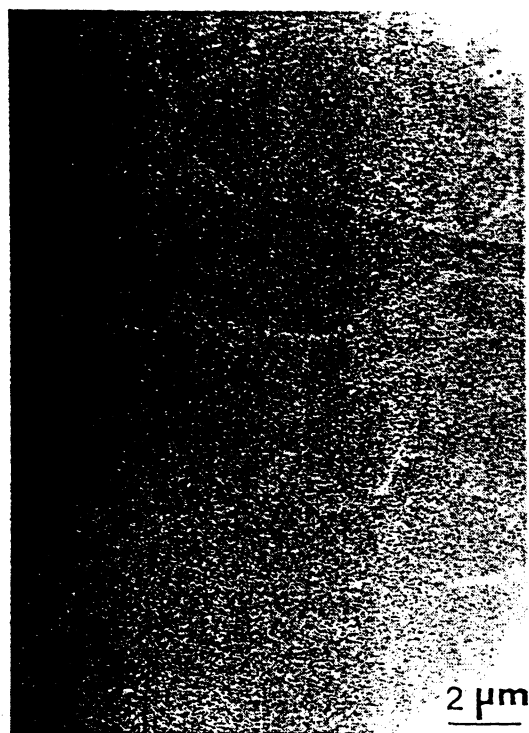


Figure 3. Predicted pressure to initiate reaction as a function of porosity for initiation of reaction in Nb-Si ( $\text{NbSi}_2$ ), Mo-Si ( $\text{MoSi}_2$ ), and Ti-Si ( $\text{Ti}_3\text{Si}_2$ ) systems (from L.H. Yu [16], p.152).



(a)

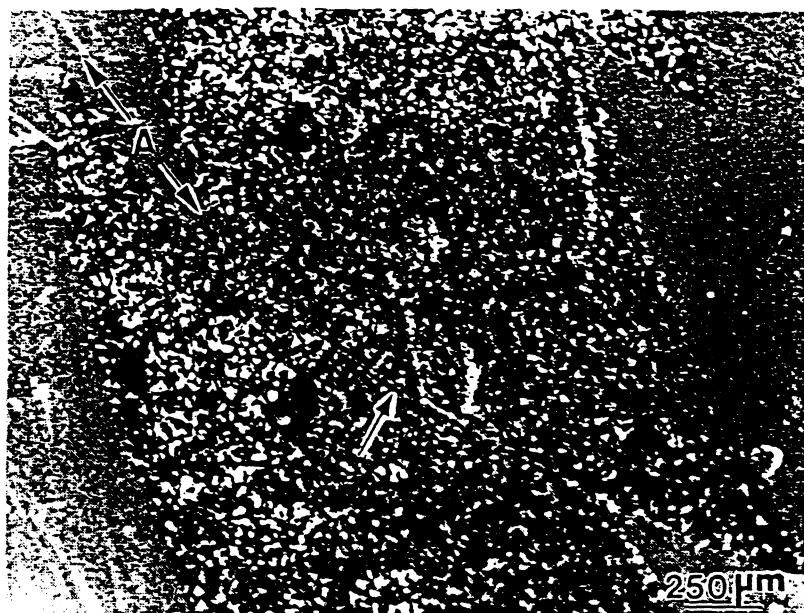


(b)

Figure 4. Fully reacted regions (SEM) in (a) Configuration 1 (after stage 2) and (b) Configuration 2.



(a)



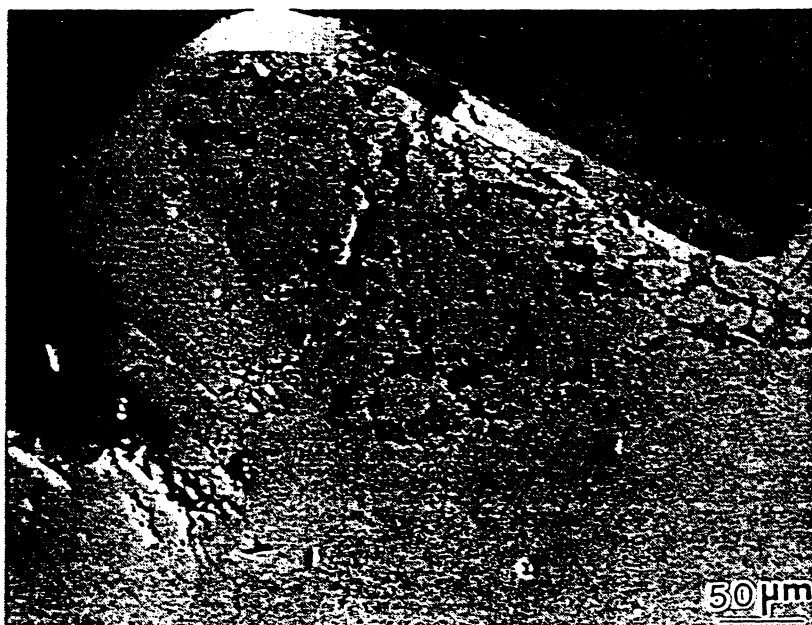
(b)

Figure 5. Overall configuration of shear bands for (a) Ti-Si system; (b) Nb-Si system.

Figure 6 shows the residual unreacted regions adjoining the copper walls for the Ti-Si in both Configuration 1 (Fig. 6(a)) and 2 (Fig. 6(b)). These regions remain unreacted because the copper, due to its high thermal conductivity, quenches the reaction. The triangular shape (wedge) created by the shear band creates an ideal configuration for heat extraction, since two converging heat sinks generate an increasingly effective reaction quencher as the apex is approached. The interface between the reacted and unreacted material (see arrow) seems to exhibit the similar features as the one observed by Yu and Meyers [17] and Vecchio et al. [18] for Nb-Si system: spherule formation at the Ti surface and its release into molten silicon. This mechanism is described in detail by Meyers et al. [15].



(a)



(b)

Figure 6. Residual unreacted Ti-Si in regions close to copper mandrels: (a) Configuration 1; (b) Configuration 2.

#### 4. CONCLUSIONS

1. High-strain-rate plastic deformation ( $\epsilon_{\text{p}} \approx 0.25$ ,  $\dot{\epsilon} \approx 10^4 \text{ s}^{-1}$ ) of predensified porous 5 Ti + 3 Si mixture (89% of theoretical density), was produced by explosive loading using low detonation velocity explosive. It resulted in shear-band pattern with typical spacing between shear bands of  $\sim 1\text{-}2$  mm.

2. Chemical reactions initiated inside shear bands and propagated throughout entire specimen. Small unreacted regions adjacent to copper driver tube remained unreacted. This is due to rapid cooling and arrest of reaction due to high thermal conductivity copper.
3. Heat of reaction of  $\text{Ti} + \text{Si}$  ( $\text{Ti}_3\text{Si}_2$ ) is 580 kJ / mole. This is four times the heat of reaction for  $\text{Nb} + \text{Si}$  ( $\text{NbSi}_2$ ) and explains drastic difference between behavior of two powder mixtures. In  $\text{Nb} + \text{Si}$  reaction was initiated within shear bands under the same conditions of strain and strain rate, but did not propagate throughout material volume.
4. High-strain-rate plastic deformation of  $\text{Ti} + \text{Si}$  mixture with larger initial porosity (46% of theoretical density) with amorphous layer, assisting shear localization, also results in complete chemical reaction throughout the material volume.

## ACKNOWLEDGMENT

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