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The collaborative program with Russian scientists was successfully initiated and has included visits, joint experiments and the preparation of a number of manuscripts. The principal result result of the research effort is that plastic shear can initiate exothermic reactions between Nb and Si powders. Regions of intense shear localization were generated in porous and densified mixtures by explosive loading. The research is being continued under an ARO contract.



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# A FUNDAMENTAL STUDY OF SHOCK-INDUCED CHEMICAL REACTIONS FOR RAPID ENERGY RELEASE APPLICATIONS

CONTRACT No DAAH04-93-G-0261

FINAL REPORT



SUBMITTED TO

THE MATERIALS SCIENCE DIVISION
U. S. ARMY RESEARCH OFFICE

BY

MARC ANDRÉ MEYERS

SEPTEMBER 8,1994

## 1. INTRODUCTION

The objectives of the program were met and, in some areas, exceeded. The primary goal of the program was to establish collaborative research programs with outstanding Russian scientists, in the area of shock compression. The first task of the program was to establish whether shock-induced chemical reactions could be aided by superimposed plastic strain. Controlled experiments established that chemical reactions between Nb and Si powders could be initiated within intense plastic shear regions (shear bands). Visits by S. S. Batsanov and V. F. Nesterenko initiated a successful research collaboration which is being continued under a three-year program that is expected to be funded by the U. S. Army Research Office. This research has led to the preparation of several manuscripts that are in the process of being submitted to scientific journals. A very productive workshop was held at Georgia Institute of Technology in May 1994 ("Shock Synthesis of Materials") with the participation of all principal scientists involved (US, Russian, Japan) in reactions under shock compression. In this workshop the main theories were presented and discussed in depth, and compared with the evidence of experimental results.

## 2. ACCOMPLISHMENTS

### 2.1 General Remarks

A number of collaborative experiments were planned and carried out; explosive experiments were conducted at the Center for High Dynamic Pressures, Mendeleevo (Moscow Region), and at the Institute of Hydrodynamics, Novosibirsk (Siberia) on materials obtained from US sources. The samples were then returned to UCSD for complete characterization by X-ray diffractometry, scanning and transmission electron microscopy, and other related characterization.

The collaborative research will be presented next.

## 2.2 Professor S. S. Batsanov

Professor Batsanov visited UCSD twice, for approximately two months. A longer absence from Mendeleevo, Moscow Region, would have disrupted his laboratory. Professor Batsanov reported to us the difficult situation of his co-workers, and the various shortages and salary delays. The members of his group being well versed in explosive handling and processing; they receive periodically offers to work on "fundamental problems" in N. Korea, Iraq, and other countries. Through small additional funds, from overseas, it is possible to retain the scientists at their present institute, until the crisis situation is overcome. It is hoped that, through the current project, some support will be made available to Prof. Batsanov's laboratory.

Prof. Batsanov brought with him boron nitride specimens which had been prepared by a method developed in his laboratory, named "Dynamic-Static Compaction". This experimental technique subjects the specimens to a high shock pressure followed by a static pressurization stage, that is enabled by an ingenious technique. The capsules containing the specimens are placed in a special chamber filled with a material that undergoes a shock-induced plastic transition. Upon shock pressure release, the reverse transformation in the chamber produces a pressure, that acts on the capsule. The use of this technique has yielded much higher transformed fractions and reduction in cracking, compared to simple shock compression. The specimens prepared by Prof. Batsanov and co-workers are being characterized at UCSD.

While Prof. Batsanov was at UCSD on his first visit, he wrote an article on shock-induced chemical reactions. This article required extensive editing and is presently being prepared for submission to Materials Science and Engineering [1]. Prof. Batsanov provided great assistance in the preparation of a proposal submitted to the U. S. Army Research Office. A number of experiments were planned during Prof. Batsanov's first visit. Prof. Batsanov carried out these experiments and brought the shocked capsules in his second trip. These capsules were characterized and yielded very interesting results, essentially confirming the mechanism proposed by Meyers, Vecchio, and Yu in two recent Acta Metallurgica et Materialia articles [Vol. 42, 1994, pp. 701-714, pp. 715-729]. Two configurations were used by Prof. Batsanov in the cylindrical geometry: (a) a symmetric geometry, in which the convergence of shock waves towards center created a Mach stem with essentially two pressure regions; (b) an asymmetrical geometry, which produced plastic deformation in addition to consolidation. Within the Mach stem of the symmetric geometry Nb and Si (and Mo and Si) fully reacted, whereas outside of this region no reaction occurred. Thus, the threshold pressure for shock-induced reactions was confirmed. Within the asymmetrically loaded specimens, shear localization regions were identified, within which chemical reaction was observed. Thus, it was shown that chemical reaction could be triggered at pressures below the Krueger-Vreeland threshold, if additional energy is provided to system as, for instance, plastic deformation [2]. Based on these positive results, a series of experiments, utilizing an entire range of powder mixtures, was planned:

Fe+Si
Cu+Si
W+Si
Ti+Si
Ta+Si
C+Si
Cr+Si

These experiments were carried out by Prof. Batsanov and the capsules are being sent to the U. S.. The characterization of these specimens will be carried out in collaboration with Pro. K. S. Vecchio and Dr. A. Strutt. The generality of the mechanism for shock-induced chemical reaction will be verified.

Concomitantly, Prof. Batsanov is attempting to synthesize carbonitrides by shock compression processing. The specimens will be characterized at UCSD.

**2.3 Professor V. F. Nesterenko**

Professor V. F. Nesterenko arrived in March 1994, and it will be possible to extend his stay until June 1995. Funding was requested and is being obtained from the Office of Naval Research ( Dr. Judah Goldwasser, Program Manager), from the Institute for Mechanics and Materials (Prof. R. J. Skalak, Director), and from the U. S. Army Research Office (Dr. E. Chen, Program Manager). With the help of Dr. R. Graham, Sandia National Laboratories, Professor V. F. Nesterenko is being offered a contract to write a book entitled "Dynamics of Heterogeneous Materials" (Springer) based on a book published a few years ago in Russia. This book will contain a comprehensive and up-to-date review of important FSU contributions to the field, as well as an extensive treatment of Professor V. F. Nesterenko's contributions.

Collaborative experiments had been discussed and planned with Professor V. F. Nesterenko as early as August 1993, at the APS Shock Compression Conference, in Colorado, Springs. Professor V. F. Nesterenko developed the TWC (Thick Walled Cylinder) method, which enables the attainment of high strains at very high strain rates ($\sim 10^5 s^{-1}$). A cylinder with a cavity along its longitudinal axis is explosively loaded along the external lateral surface; consequently, the central cavity is collapsed. By appropriate choice of explosives and experimental configurations, wave reflection effects are minimized. The TWC method was applied to the tubular cavity within the copper container, which on its turn, contained the axial orifice. High-strain rate deformation of the Nb-Si mixture led to the formation of numerous shear localization areas. These shear localization areas were subjected to prescribed and controlled strains at controlled strain rates. Very interesting original results were obtained in this work, and a novel phenomenon was observed: vortex formation within the shear localization region. Evidence of reaction on the scale up to 7μm within the shear band with thickness 10~20 μm was also obtained. The basic importance of these results resides in the fact that the small thickness of shear bands ensures that reaction took place on the same time scale as high-strain-rate deformation (~$10^5$ $s^{-1}$). The results were published in an IMM Report [3] and submitted for publication.

The formation of a regular and periodic array of shear bands in the tubular Nb-Si specimens and discussions held with Prof. R. J. Clifton, Brown University, were the motivation for the application of the TWC method to granular alumina. This is a fundamental problem of great practical importance, because, during ballistic impact of ceramic armor, the shock wave ahead of the projectile comminutes the ceramic. This pulverized ceramic has to be displaced for penetration to proceed. The deformation of the pulverized ceramic is still poorly understood. The TWC method can be used to subject pulverized ceramic to high strain and high-strain-rate deformation. Two alumina powders were used, in consultation with Prof. R. J. Clifton: 0.5 and 5 μm particle sizes. These powders are also being used by Prof. Clifton in inclined plate impact experiments. The alumina powders were first consolidated in the tubular TWC set-up. A second explosive event imparted considerable plastic strain to the densified/ granular alumina. Upon "plastic" deformation, this alumina underwent shear localization with clearly defined shear bands having a regular spacing. Their structure was qualitatively different for materials with different particle sizes. The results of this work are currently being prepared for publication [4]. A combined account of the Nb-Si and $Al_2O_3$ research will be presented at the October AIME symposium "Dynamic Behavior of Materials", in Chicago; a manuscript will be submitted to Metallurgical and Materials Transactions [5].

The work on mesomechanic phenomena in powder densification is under development in collaboration with Professors V. F. Nesterenko and D. Benson; results will be published in [6]. In collaboration with an existing NSF grant on shear localization and with the U S. Army Research Office University Research Initiative Program, experiments on titanium and tantalum specimens were carried out with the TWC method. In titanium, a regular array of shear bands, having a spiral morphology, were produced. The TWC method yields itself very well to the study of the collective behavior of shear bands. Fundamental questions that need to be addressed are: what determines the amount of spacing between shear bands? What microstructural factors determine the initiation of shear bands? What are the structural characteristics of the shear band itself? The tantalum specimens were subjected to a high strain and high-strain-rate regime and the microstructural evolution is being characterized.

## 3. CONCLUSIONS AND FUTURE WORK

This was a highly successful program; the enthusiasm displayed by Profs. Batsanov and Nesterenko was a source of inspiration to the UCSD participants. The synergism of the interactions is clear in the novel experimental results and important advances are being made in the dynamic mechanical response of granular ceramics. The recent award of a three-year contract from the U. S. Army Research Office will enable the continuation of this research program. The research program is well balanced, and takes advantage of a unique expertise in Mendeleevo and Novosibirsk of shock-compression experimental set-ups which produce prescribed and controlled shock pulses and plastic deformation; this is combined with the analysis and characterization capability at UCSD. It would be desirable to fund Prof. Batsanov's and Prof. Nesterenko's laboratories at a rate of U$ 10,000 per year. This will accomplish two objectives: (a) ensure the stability of these laboratories during this critical period; (b) enable the continuation of the collaborative research.

In the future, we plan to implement some of the Russian experimental techniques in the U. S.. Plans are being made to carry out TWC (thick wall cylinder) experiments at New Mexico Tech. The possibility of using additional diagnostics is being discussed.

## 4. FORTHCOMING PUBLICATIONS

1. M. A. Meyers, S. S. Batsanov, S. M. Gavrilkin, H. C. Chen, J. C. La Salvia, and F. Marquis, "Effect of Shock Pressure and Plastic Strain on Chemical Reaction in Nb-Si and Mo-Si Systems", in preparation, to be submitted to Mat. Sci. Eng., (1994)
2. S. S. Batsanov, S. M. Gavrilkin, V. A. Lukashava, I. I. Maksimov, M. A. Meyers, R. Prummer, E. E. Serko, V. A. Tumanov, "Phase Transition h-BN -> w-BN under Dynamic-Static Compression and Properties of Product" in preparation.
3. V. F. Nesterenko, M. A. Meyers, H. C. Chen and J. C. La Salvia, "Controlled High-rate Localized Shear in Porous Reactive Media", Appl. Phys. Lett., Submitted (12 July, 1994), and IMM/UCSD Technical Report No. 94-11, August (1994)
4. V. F. Nesterenko, M. A. Meyers, H. C. Chen, "Dependence of Shear Localization on Particle size in Densified Granular Alumina", Acta Met. et Mat., in preparation, to be submitted (1994)
5. V. F. Nesterenko, M. A. Meyers, H. C. Chen and J. C. La Salvia, "High-rate Shear Localization in Heterogeneous Porous Materials", Met. Trans., in preparation, to be submitted (1994)
6. M. A. Meyers, D. Benson, and V. F. Nesterenko, "Shock Consolidation: Microstructures, Mechanisms, and Limitations", Materials Reports, in preparation (invited overview article), to be submitted (1994).