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Movie Mode Dynamic Transmission Electron Microscope: Revealing Material Processes at Nanometer and Nanosecond Scales with Multi-frame Acquisition

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The transient states that result in a material driven to far-from equilibrium conditions, evolve on short time scales, ranging from the femtosecond-scale processes occurring from non-equilibrated electron states to microsecond transient events of phase transformations and deformation processes. Typically, we are confined to conduct experiments near equilibrium or observe material post process due to the resolution limitations of conventional analytical techniques. Though insight about a material's behavior gained from these observations, attempts to understand the coupled and convoluted events in complex processes have been hampered by the difficulty of capturing the events in detail as they unfold on nanosecond and microsecond scales, requiring a technique with improved temporal resolution. To meet this need, the dynamic transmission electron microscope (DTEM) at Lawrence Livermore National Laboratory was developed which can capture a complete diffraction pattern or image of a fast-evolving material process in a single 15 ns exposure.

Prior DTEM hardware only allowed single-pump/single-probe operation. Building up a process's typical time history required repeating an experiment with varying time delays at different sample locations. It has been used successfully to reveal complex, coupled behavior involving chemical reactions, phase transformations, convective mass transport, and micro/nanostructural evolution [1-4]. Movie Mode DTEM upgrade now enables single-pump/multi-probe operation. These technical improvements provide the ability to track the creation, motion, and interaction of individual defects, phase fronts, and chemical reaction fronts, providing invaluable information of the chemical, microstructural and atomic level features that influence the dynamics and kinetics of rapid material processes. For example, we have used the new Movie Mode capability to study reaction dynamics in Ti-B based reactive multi-layer foils (RMLF). An example data set is shown in FIG. 1 collected as the reaction front passes through the field of view. The granulated topography of TiB_2 product phase is strongly contrasted in the image, providing an easily distinction between the reacted and unreacted zones and position of the propagating front. The reaction front velocity can be measured precisely from the sequence of images, which for 2Ti-3B composition, is 9.90±0.06 m/s. The stoichiometric Ti-2B RMLF have higher front velocity, 13.37±0.03 m/s (See FIG. 2), which is expected due to the higher reaction heat and enhanced diffusion kinetics.

To understand the phase evolution behind the reaction front of 2Ti-3B RMFLs in more detail, we obtained a sequence of diffraction patterns as the front passed through a 1.75 μm diameter field of view (shown in the FIG. 3). At the reaction front (0ns), the diffraction pattern exhibits three broad peaks with peak positions and intensities that cannot be attributed to nanocrystalline beta-Ti or boron structure. The peaks in the experimental data are much boarder and shifted to higher scattering vector indicative of an amorphous phase rather a crystalline, suggesting a liquid phase exists at the front. A significant fraction of TiB_2 phase forms within 250ns and the foil completely transforms within 750ns. At a front velocity of ~10 m/s, a transient zone existing for 750ns corresponds to distance of ~7.5μm that is illustrated by the scaled real space image adjacent to the diffraction spectra. At times

less than 250ns, there many of irregular shape dark regions that can be attributed liquid and diffraction spectra suggest that there is large fraction liquid behind the reaction front.

While a single pump-probe snapshot provides statistical data about these factors, movie mode DTEM allows all of the factors of material processes to be identified and its evolution to be explored in detail. Movie Mode enables entirely unprecedented kinds of measurements, finally allowing us to directly observe fast, complex material processes that have until now only been dimly glimpsed. This presentation will discuss the technical aspects of the Movie Mode DTEM in the context of recent material science studies using the novel *in situ* TEM capability.

References

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[5] This work was performed under the auspices of the U.S. Department of Energy by Lawrence Sciences, Division of Materials Science and Engineering of the U.S. Department of Energy under Contract No. DE-AC52-07NA27344.

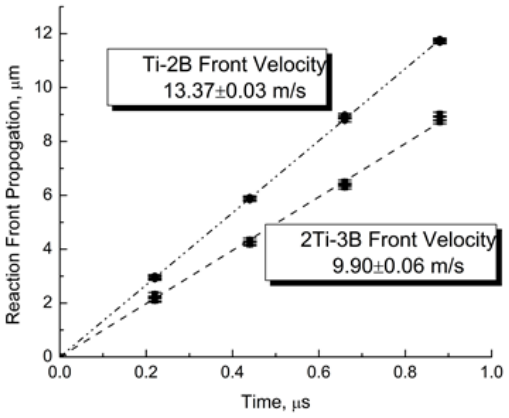

FIG. 2. Plot of the measured reaction front propagation as a function of time for Ti-2B and 2Ti-3B RMLF

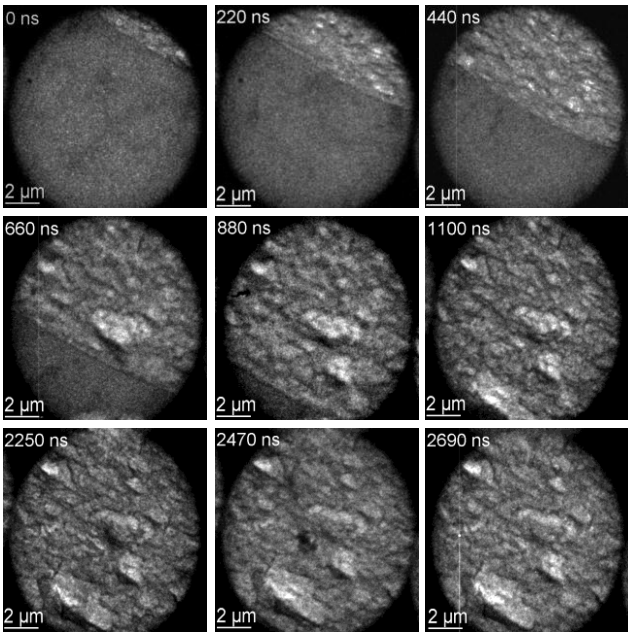

FIG. 1. An example of Movie Mode DTEM measurements A 9 image sequence of propagating front in a reacting 2Ti-3B RMFL. The exposure time of each image is 17 ns. Note the thin dark line at the reaction front which we infer from enhanced thermal diffuse scattering and dark contrast to be a liquid phase.

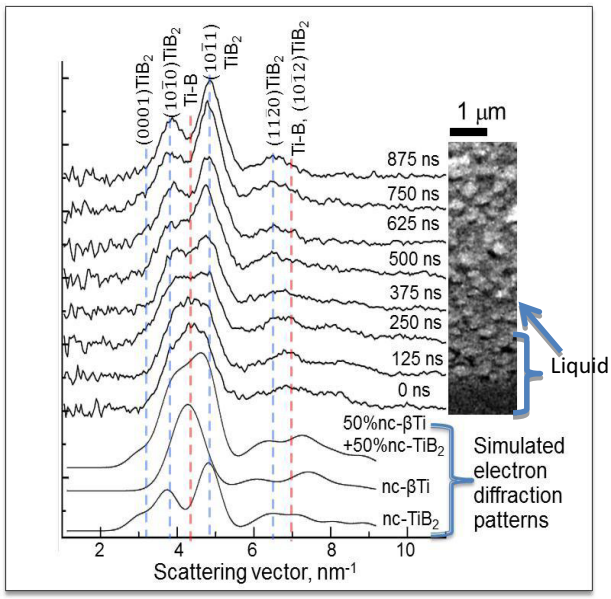

FIG. 3. A sequence of diffraction spectra collected with Movie-mode DTEM as a reaction passes through a 1.75µm diameter field of view, showing the evolution from an amorphous Ti-B structure to the TiB_2 hexagonal crystal structure.

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