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Final Report on DTRA Basic Research Project #BRCALL08-Per3-C-2-0006

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Final Report on DTRA Basic Research Project #BRCALL08-Per3-C-2-0006

“High-Z Non-Equilibrium Physics and Bright X-ray Sources with New Laser Targets”

IACRO #10-4238I

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1. Goals of the Project

This project had two major goals, as follows.

- Final Goal: obtain spectrally resolved, absolutely calibrated x-ray emission data from uniquely uniform mm-scale near-critical-density high-Z plasmas not in local thermodynamic equilibrium (LTE) to benchmark modern detailed atomic physics models.
    - Scientific significance: advance understanding of non-LTE atomic physics
- Intermediate Goal: develop new nano-fabrication techniques to make suitable laser targets that form the required highly uniform non-LTE plasmas when illuminated by high-intensity laser light.
    - Scientific significance: advance understanding of nano-science
- Relation to DTRA C-WMD mission: The new knowledge will allow us to make x-ray sources that are bright at the photon energies of most interest for testing radiation hardening technologies, the spectral energy range where current x-ray sources are weak.

All project goals were met, as detailed in the next section.

2. Project Accomplishments

This project consisted of three principal tasks: laser target development, laser experiments, and computational design and modeling.

2.1. Laser Target Development

We pursued two parallel lines of target development, as follows.

2.1.1. We developed robust, lightweight Cu-loaded C foams tunable over a large range of densities, ~5 – 50 mg cm^{-3}. These foams were made using a couple of new techniques based on freeze-drying of a $CuSO_4$ carbon nanotube (CNT), and graphene-oxide (GO) solution, as illustrated schematically in Fig. 1. We submitted a patent application on this new process.

These C-Cu foams were fashioned into Omega-laser-scale (2 mm x 2 mm right cylinders) targets, and used to obtain x-ray emission data in Omega laser experiments. One of these Omega targets is shown in the photograph in Fig. 2, along with a scanning electron microscope (SEM) image of the foam. The little white dots in the SEM image are the Cu nanoparticles, having an average diameter of ~50 nm, embedded in the pores of the C foam.

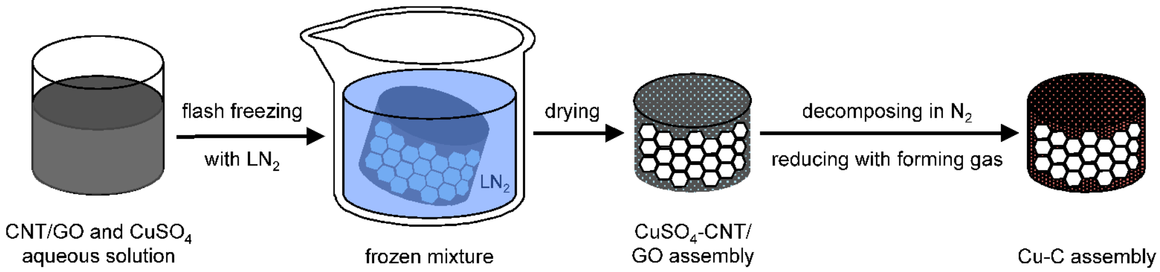

Figure 1. Schematic of the new synthesis process used to fabricate Omega targets with overall density 5 – 9 mg cm^{-3}.

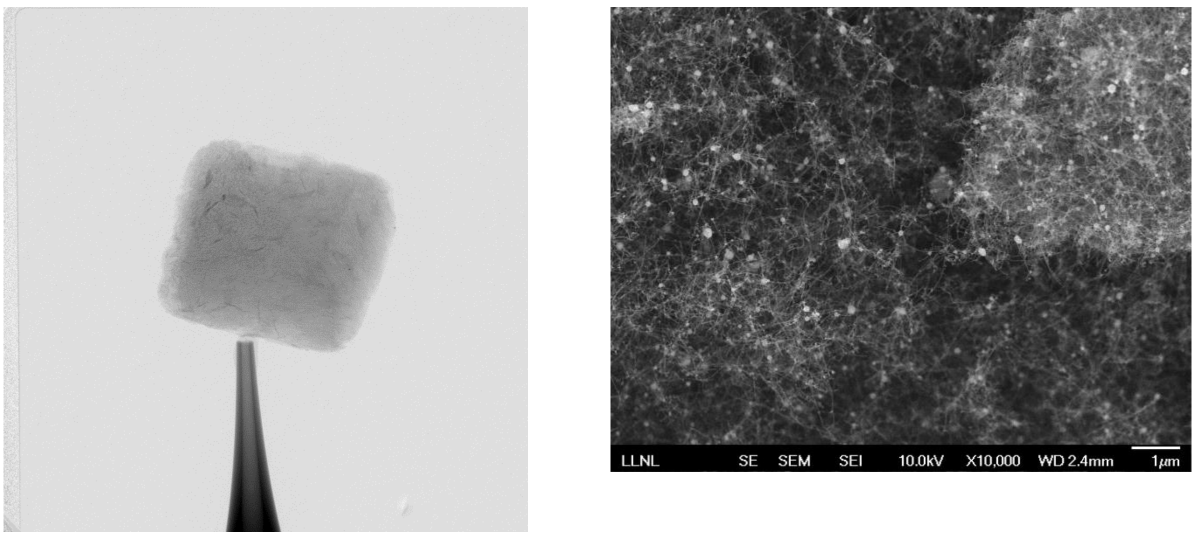

Figure 2. Left panel: 2 mm x 2 mm C/Cu foam Omega target on its stalk; Right panel: SEM image of C/Cu foam. The 1-μm scale bar is shown in the lower right corner of the image.

2.1.2 Our discovery of an entirely new and surprisingly simple technique for fabricating pure copper foams followed a long series of unsuccessful paths. These pure, unsupported foams are robust and can be tuned from high density to as low as 14 mg cm^{-3}. High quality targets of this foam were successfully shot at Omega and NIF. Briefly, the new technique involves freeze-drying an aerogel solution of high-aspect-ratio Cu nanowires and strengthening the nanowire interconnections via oxidation-reduction cycles. Using this new technique we succeeded in making pure Cu foams, spherical in shape and mechanically stable, with overall density <20 mg cm^{-3}, as shown in Figure 3. The spherical foams were mounted on a loop of Cu wire that was attached to the target stalk. We fabricated sets of targets of 2 mm diameter, suitable for Omega experiments, and sets of targets of 4 mm diameter, suitable for NIF experiments. We submitted a provisional patent application on this new foam fabrication process.

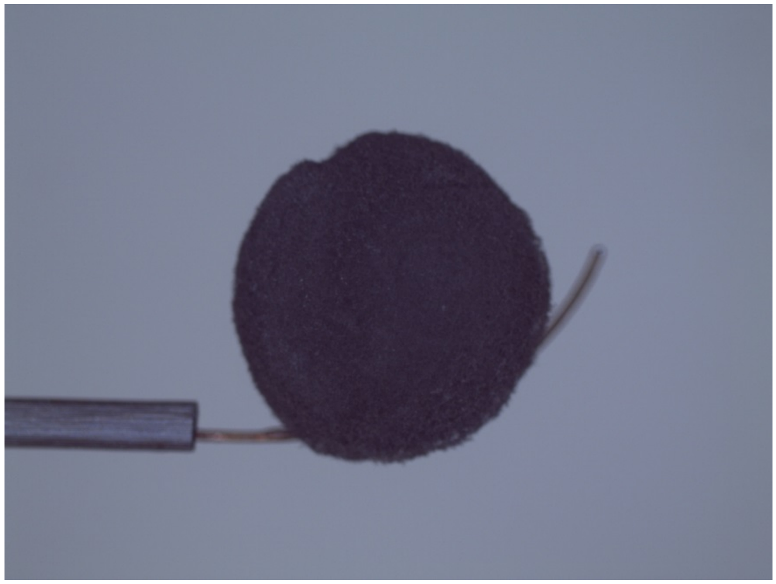

Figure 3. Pure Cu foam sphere of diameter 4 mm and density 14 mg cm^{-3}, affixed to a thin Cu wire loop for handling and mounting on target stalk.

In Fig. 4 we show SEM images at two different magnifications of the new pure Cu foam.

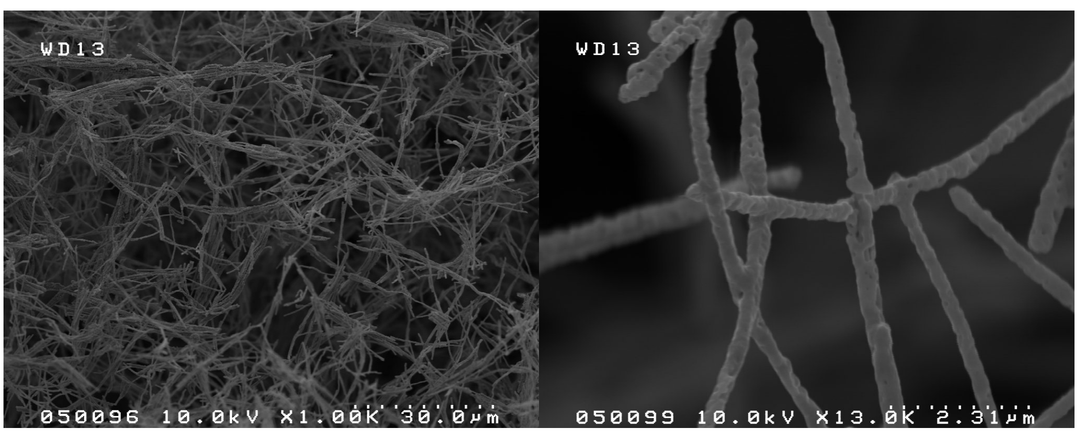

Figure 4. SEM images of pure Cu foam at two different magnifications.

2.2. Laser Experiments

The first set of laser experiments were conducted on the Omega laser, located at the University of Rochester in Rochester, New York, in January 2013 with C/Cu targets with pre-shot densities 30 – 50 mg cm^{-3}. Omega laser experiments with the new foam targets shown in Fig. 2 and fabricated by the process illustrated in Figure 1 were conducted in February 2014. All these targets had pre-shot densities <10 mg cm^{-3}. The Omega laser put 20 kJ of 1/3-micron laser light onto the target in three beam cones per each end of the cylindrical target. A total of seven shots were completed in February 2014. An array of x-ray and optical diagnostics were fielded.

Fig. 5 shows the framing camera images from one of the 2014 Omega shots in the left panel compared to the simulated images of the same shot in the right panel. The framing camera was filtered to record the >8 keV x-rays from the Cu K-shell. As is evident in this image, these lower-density foams (5 – 9 mg cm^{-3}) heated volumetrically, in agreement with the modeling simulations.

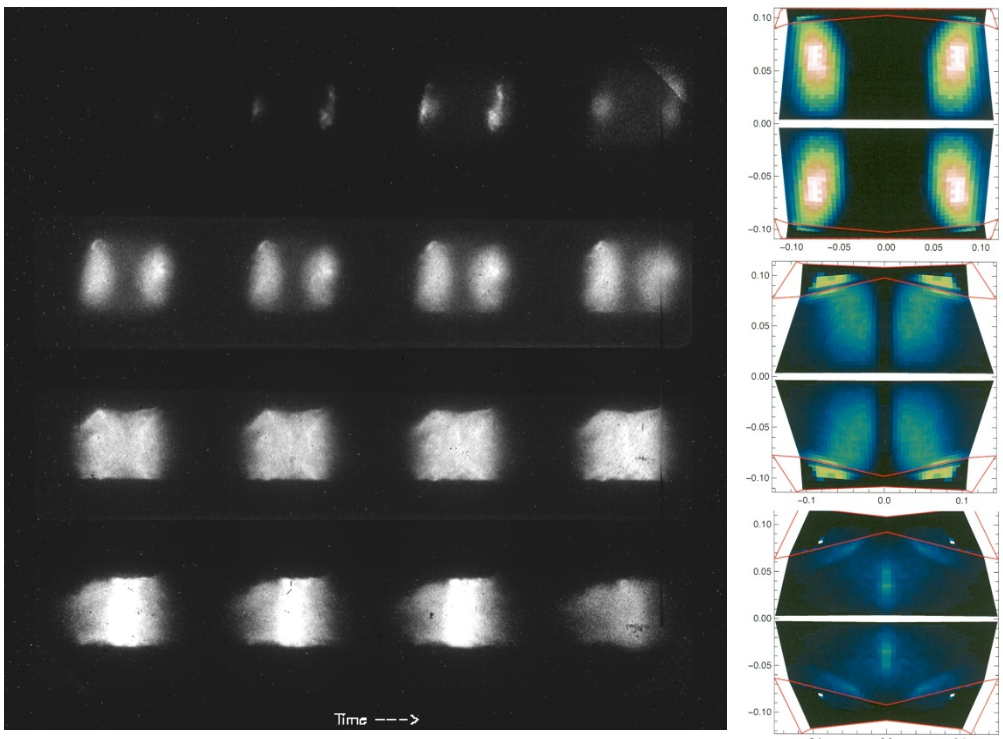

Figure 5. Left panel: x-ray framing camera images of Omega shot #72351, pre-shot foam density 6.5 mg cm^{-3}, 8 at% Cu. Time goes left to right and top to bottom, with the inter-frame time 80 ps and the four horizontal strips spaced at 0.5 ns intervals with 0 ns at the top left. Right panel: simulated x-ray framing camera images of Omega shot #72351, pre-shot foam density 6.5 mg cm^{-3}, 8 at% Cu. Times are 0.5 ns (top), 1.0 ns (middle), 1.5 ns (bottom). Pulse duration was 1 ns.

Fig. 6 shows a streak camera image of the same Omega shot as shown in Fig. 5. Like the framing camera, the streak camera was filtered to record the >8 keV Cu K-shell x-rays.

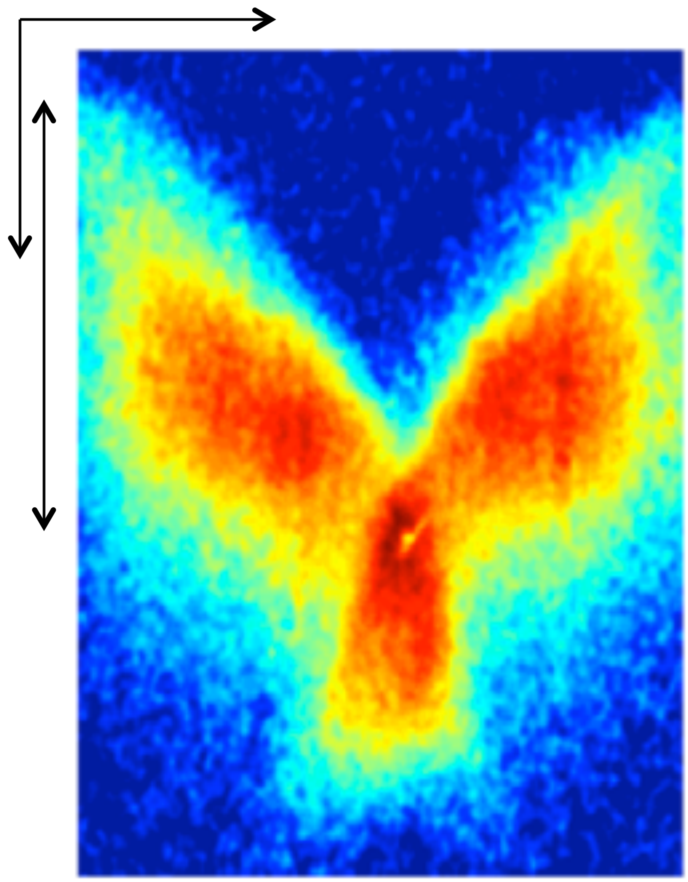

Figure 6. X-ray streak camera image of Omega shot #72351, pre-shot foam density 6.5 mg cm^{-3}, 8 at% Cu. Space increases to the right, time increases down, as indicated by the arrows in the upper left corner. The double-headed arrow indicates the laser pulse time duration, 1 ns.

As is evident in Figure 6, a heating front propagates towards the center of the target from the two heated ends, and eventually the whole foam is heated and emits x-rays. In addition, the x-ray emission continues after the end of the laser pulse. We showed in the full set of Omega shots that heat front propagation is slower in the higher density foams, consistent with simulations.

Cu K-shell x-ray conversion efficiency was measured by the absolutely calibrated DANTE broadband filtered x-ray diode array. We did not have any targets with a pre-shot foam density of 10 – 20 mg cm^{-3} (the targets for the 2013 experiments had pre-shot densities 30 – 50 mg cm^{-3} and the targets for the 2014 experiments had pre-shot densities <10 mg cm^{-3}), but the measured x-ray conversion efficiencies are consistent with there being a maximum of ~1% for pre-shot foam density in the range 10 – 20 mg cm^{-3}, as predicted by simulations. We published a detailed analysis and comparison with modeling in *Physics of Plasmas* in 2015 (see Section 4 for complete reference).

Five Omega shots were executed in November 2014 with the new pure Cu spherical foams. All these foams had diameter ~2 mm. Cu K-shell x-ray conversion efficiency was measured by DANTE to be ~2%, in good agreement with simulations.

Three NIF shots were executed in December 2014 with the new pure Cu spherical foams. All these foams had diameter ~4 mm. On these shots we obtained the first ever Cu K-shell x-ray spectra with the new time-resolved high-resolution NIF X-ray Spectrometer (NXS). Time resolution was 80 ps.

We show in Fig. 7 lineouts at one particular time of the spectral data for the three NIF shots with the background subtracted out. There is not yet an absolute calibration for this diagnostic. Most of the K-shell emission is He-α emission at ~8.4 keV from He-like Cu ions. This He-α emission increases with foam density up to 18 mg cm^{-3}, which is the density for near-maximum x-ray conversion efficiency. Note also that the Ly-α emission (from hydrogen-like Cu ions) at ~8.7 keV is visible but not much above background. We also observed that the peak emission shifts later in time as the pre-shot density increases.

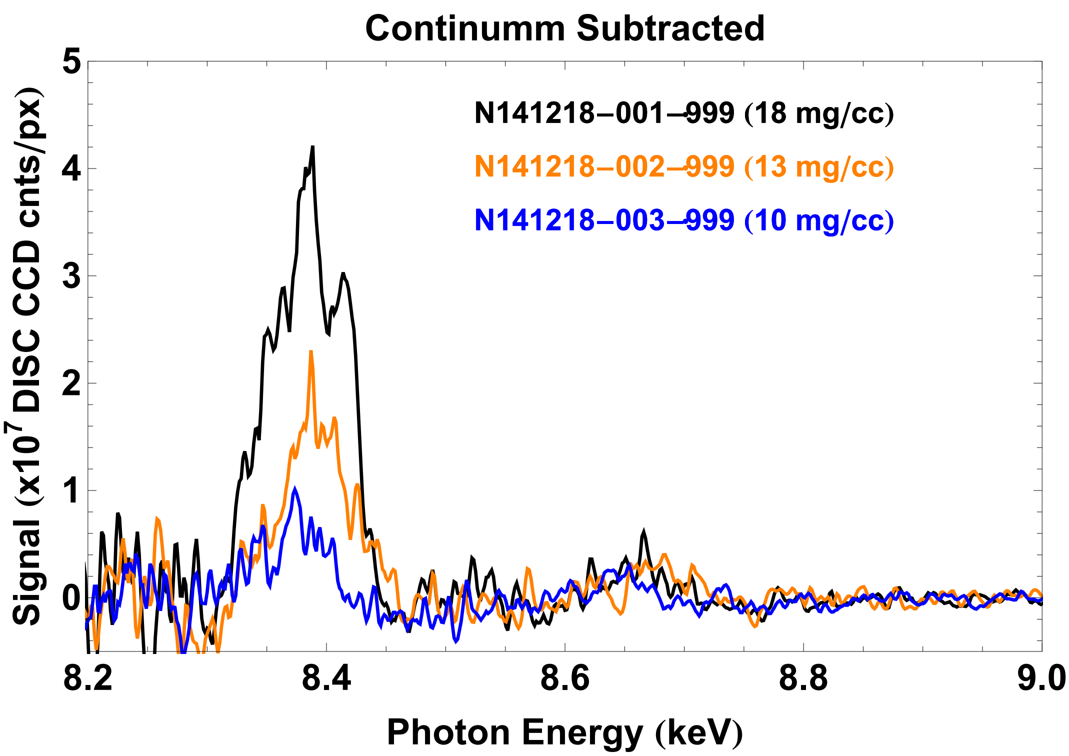

Figure 7. Lineouts at one particular time of the very first NIF/NXS spectral data obtained using our pure Cu foam spheres

Cu K-shell x-ray conversion efficiency was measured by DANTE to be 4 – 6% on the three NIF shots, which is consistent with the range inferred from the time-integrated NXS data. This efficiency is less than what was predicted in the simulations, for a reason we do not yet fully understand.

Nonetheless, the success with Cu emboldened us to adapt the same newly developed techniques to making very low-density pure Ag foams in the last months of this project. We not only succeeded in fabricating Ag foams but we executed one NIF shot in October 2015 with a spherical Ag foam with pre-shot density 24 mg cm^{-3} and three NIF shots in January 2016 with cylindrical Ag foams having pre-shot densities 22, 26, and 26 mg cm^{-3}. As with the NIF Cu spheres, measured L-shell x-ray conversion efficiency for all the Ag foam shots was less than predicted by simulations. The extended period of performance of this research grant ended on January 31, 2016, but we now have in hand a wealth of new data that, with further analysis, will allow us to define research directions for the future.

2.3. Computational Design and Modeling

Simulations were done with a non-LTE super-configuration atomic model incorporated in both our 2D and our 3D radiation-hydrodynamics computer codes. Population densities of each ionization level in the laser-heated plasma, in a principal quantum number description (i.e., super-configurations), are obtained from non-LTE rate equations, taking into account every possible interaction between electrons, ions, and photons --- including radiative transitions. We account for absorption of the laser beam energy by inverse bremsstrahlung and heat transport by flux-limited thermal conduction.

Simulations were done for design of all the experiments and for estimates of expected outputs to aid setup of diagnostics. In addition, post-shot simulations were done to compare with actual data. Over the course of this research we made several modifications to our baseline computational model in order to get simulations to agree with data. This work has led to the most advanced computational model in existence for under-dense non-LTE high-Z plasma that has been benchmarked with actual data. The new model is beginning to be adopted by other research programs at LLNL.

In addition, as part of this research project we conducted a simulation parameter study to compare L-shell emission from pure Ag foams (3 – 5 keV) to Ar K-shell emission (also 3 – 5 keV). We used the same target and laser beam setup as used in previous NIF gas target shots. In the parameter study we varied the pre-shot density, the laser beam energy, and the laser pulse duration. We found that Ag is 10 to 20 times more efficient per ion in 3 – 5 keV x-ray emission than is Ar. Maximum x-ray conversion efficiency in the 3 – 5 keV photon energy band is 36% for Ag and 18% for Ar. We have thus found a new path to higher x-ray conversion efficiencies: substituting a higher-Z equivalent photon energy L-shell source for a K-shell source.

3. Personnel Supported, Training and Professional Development

The following people have been partly supported by this grant during all or some period of time in the five-year course of this research.

At LLNL:

Dr. Jeff Colvin, PI, staff scientist
Dr. Sergei Kucheyev, staff scientist
Dr. Kevin Fournier, staff scientist
Dr. Mark May, staff scientist
Dr. Supakit Charnvanichborikarn**,** postdoctoral research fellow
Dr. Michael Bagge-Hansen, postdoctoral research fellow
Dr. Frédéric Pérez, postdoctoral research fellow
Dr. Maria Barrios, postdoctoral research fellow
Dr. G. Elijah Kemp, postdoctoral research fellow

At University of California-Davis:

Prof. Kai Liu
Mr. Edward Burks, graduate student, presently Lawrence Graduate Scholar at LLNL
Mr Dustin Gilbert, graduate student, presently NRC postdoctoral research fellow at NIST
Mr. Chad Flores, graduate student

At Sandia National Laboratories-California:

Dr. Tom Felter, staff scientist

This project has formed part of the basis for the training and thesis work for the UCD graduate student Edward Burks. Of the 5 LLNL postdoctoral fellows who at one time or another worked on this research grant, Charnvanichborikarn and Pérez have taken staff scientist positions elsewhere (the former in South Korea, the latter in France). The other three have been converted to staff scientist positions at LLNL, largely on the basis of their work on this research project.

4. Dissemination of Research Results

Some of the research results generated by this project have been presented in four refereed journal publications, as follows.

- F. Pérez, J. D. Colvin, M. J. May, S. Charnvanichborikarn, S. O. Kucheyev, T. E. Felter, and K. B. Fournier, "High-power laser interaction with low-density C/Cu foams", *Phys. Plasmas* **22**, 113112 (2015).
- G. E. Kemp, J. D. Colvin, K. B. Fournier, M. J. May, M. A. Barrios, M. V. Patel, H. A. Scott, and M. M. Marinak, "Simulation study of 3-5 keV x-ray conversion efficiency from Ar K-shell vs. Ag L-shell targets on the National Ignition Facility", *Phys. Plasmas* **22**, 053110 (2015).
- S. Charnvanichborikarn, M. A. Worsley, M. Bagge-Hansen, J. D. Colvin, T. Felter, S. Kucheyev, "Ice-templating synthesis of low-density porous Cu-C composites", *J. Mater. Chem. A* **2**, 18600 (2014).
- S. Charnvanichborikarn, S. J. Shin, M. A. Worsley, I. C. Tran, T. M. Willey, T. van Buuren, T. E. Felter, J. D. Colvin, and S. O. Kucheyev, "Nanoporous Cu-C composites based on carbon-nanotube aerogels", *J. Mater. Chem. A* **2**, 962 (2014).

Some of the research results generated by this project have been presented at several scientific conferences.

We also submitted the following three Patent Applications.

"Low-density interconnected metal foams", Provisional Patent Application Number 62/261211, November 2015

"Porous materials via freeze-casting of metal salt solutions", Patent Application Number 14/741334, June 2015

"Nano-porous metal-carbon composite", Patent Application Number 14/485474, September 2014

5. Project Transition

This research project is continuing with funding now from the Nuclear Survivability Branch of the U. S. Department of Energy National Nuclear Security Administration. Jeff Colvin of LLNL is the project PI.