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In–Pile Instrumentation Advanced Manufacturing: Development of Pt, Mo, and Nb inks

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In–Pile Instrumentation Advanced Manufacturing: Development of Pt, Mo and Nb inks

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SUMMARY

This report summarizes the nanoparticle ink synthesis for in–pile nuclear sensor applications. Bulk nanopowders of Pt, Mo, and Nb were dispersed in aqueous solvents and printed into in–pile nuclear sensor structures to demonstrate their compatibility with aerosol jet printing and plasma jet printing. While we did not directly demonstrate plasma jet printing, due to delays in equipment installation from the manufacturer, compatibility with aerosol jet printing technology is expected to transfer to the plasma jet printer based upon the manufacturer's specifications for ink compatibility.

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1. INTRODUCTION

The In-Pile Instrumentation (I2) program conducts research needed to develop and deploy unique instruments to characterize the behavior of fuels and materials during irradiation tests inside of nuclear test reactors. The mission of this program is to establish baseline instrumentation and novel sensors and measurements systems for in-pile applications that can provide real-time, accurate, spatially resolved information regarding test conditions and the performance of fuels and materials during irradiation. This mission is split into three objectives:

1. Demonstrate and maintain state-of-the-art instrumentation capabilities available to support irradiation testing of fuels and materials in support of DOE-NE programs.
2. Perform research and development of advanced sensor technologies and integrated measurement systems for in-pile applications.
3. Provide technical support to DOE activities in the area of nuclear instrumentation development and deployment.

A strong interdisciplinary scientific collaboration designed upon specific instrumentation research and development is required to meet these objectives. Completion of these objectives will provide technologies and instrumentation that can be rapidly developed and customized to suit the needs of individual irradiation tests.

The vision of the Advanced Manufacturing (AM) Work Package is to develop a diverse set of AM methods, equipment, and expertise to enable versatile sensor fabrication processes and meet the novel sensor designs developed in the other I2 work packages. It is anticipated that AM will significantly accelerate the feedback loop of design, modeling, fabrication, and testing by reducing the traditionally time-consuming manufacturing and prototyping time frame, thus enabling rapid advancements for in-pile instrumentation.

2. Advanced Manufacturing

The development of advanced manufacturing technologies which integrate electronic devices directly onto physical packaging stands to revolutionize the development of in-pile nuclear sensors. One such manufacturing method with emerging industrial impact is direct ink writing techniques such as aerosol jet printing (AJP) and plasma jet printing (PJP). AJP has recently been used to print conformal sensors, antennas, shielding and other active and passive components within the cellular telephone and aerospace industries.[1,2] Typical AJP materials (inks) include metals such as gold, silver, and nickel, as well as insulators such as polyimide and SU-8.[3,4] PJP is a much newer technique with fewer tested inks, though many AJP inks are expected to be compatible with PJP. The database of inks available for AJP is rapidly expanding and benefitting greatly from emerging nanomaterials development, e.g. development of graphene based inks.[5] Such material breakthroughs could have a significant impact on in-pile sensor development for real time monitoring of nuclear fuel and material behavior during a Material Test Reactor (MTR) irradiation test. In order to enable such an advance in sensor technology, much work remains to be done to develop novel material inks and understand the irradiation behavior of printed materials and devices.

Boise State University is uniquely positioned to address this knowledge gap. ***Hence, the objective of the crosscutting advanced manufacturing research at Boise State University is to develop novel nanomaterial inks for additive manufacturing of passive and active in-pile nuclear sensors.*** Additional efforts are underway through NSUF RTE's to investigate the effects of irradiation on printed electronic

materials in order to elucidate fundamental knowledge about structure-property-processing correlations of additively manufactured sensors and systems operating under extreme conditions.

Specifically, for this milestone we have developed Pt, Nb, and Mo nanoparticle inks for AJP and plasma jet printing PJP. Table 1 summarizes the priority materials for ink nanoparticle ink development, relative to passive neutron dosimeters. These sensors represent one of the widest ranges of material requirements as many of the developed inks for dosimetry can then be applied to temperature, strain, and thermal conductivity sensors.

Table 1. List of material requirements for ink development and additive manufacturing of dosimeters.

Material	Commercial Ink Purchased	Ink Requires Synthesis	Estimated Dosimeter Complete	Priority per CCN 243151	Additional Notes
Titanium	N	Y	January 2019	1	Received as PVP capped powder
Iron	N	Y	January 2019	1	Received as PVP capped powder
Cobalt	N	Y	January 2019	1	Dispersed in water/PVP solution
Nickel	Y	N	January 2019	1	Commercial ink received and demonstrated printability with PA
Zinc	N	Y	2019	1	Not Purchased
Indium	N	N	2019	1	Not Purchased
Manganese	N	Y	TBD	2	Not Purchased
Copper	Y	N	January 2019	2	Commercial ink received and demonstrated printability with PA
Silver	Y	N	October 2018	2	Commercial ink received and demonstrated printability with UA and PA
Gold	N	Y/N	2019	2	Not purchased
Sodium	N	Y	TBD	3	Not purchased
Magnesium	N	Y	TBD	3	Not Purchased
Aluminum	N	Y	2019	3	Not Purchased
Scandium	N	Y	TBD	3	Not Purchased
Gallium	N	Y	TBD	3	Not Purchased
Arsenic	N	Y	TBD	3	Not Purchased
Rubidium	N	Y	TBD	3	Not Purchased
Yttrium	N	Y	TBD	3	Not Purchased
Zirconium	N	Y	TBD	3	Not Purchased
Niobium	N	Y	Janurary 2019	3	Dispersed in water and EG, demonstrated printability with UA
Molybdenum	N	Y	January 2018	3	Dispersed in water, demonstrated printability with PA
Ruthenium	N	Y	TBD	3	Not Purchased
Tantalum	N	Y	TBD	3	Not Purchased
Tungsten	N	Y	2019	3	Purchased as PVP capped nanopowder

3. Nanoparticle Ink Development

Boise State University was recently awarded two NSUF infrastructure awards. One award was to procure an Optomec 200 AJP system and another was to procure a suite of instruments for “top down” and “bottom up” nanoparticle ink synthesis used in this I2 work package. Additionally, Boise State University purchased a plasma jet printer from Space Foundry. Our nanoparticle ink development targets these two systems, with an understanding that demonstration of printing on the Optomec 200 should translate to printability in the PJP system, as per the manufacturer’s specifications. In this section we detail our “top down” approach to nanoparticle ink synthesis. Here, “top down” refers to ink development from purchased bulk nanopowders.

3.1.1 Top down Pt ink synthesis

Platinum ink synthesis proceeds as follow. Pt nanoparticles (powder) were purchased from US Research Nanomaterials (Fig. 1A). The hydrophobic Pt nanoparticles (Fig. 1B) were probe sonicated in a water/trisodium citrate solution for ~24 hours (total processing time). Trisodium citrate/water/Pt NPs were combined with 10,000 g/mol (Fig. 1C) polyvinylpyrrolidone solution and left to stir for 2 days. Excess salt and PVP were removed via ultracentrifugation (Figs. 1D,E). Pellets produced from the ultracentrifuge process were suspended in 10 mL of 90:10 Wt% water to ethylene glycol solution (Figs. 1F,G) to formulate an ink with a viscosity compatible with AJP and PJP. Ink was then dispersed on TEM grids for analysis of particle size. TEM of dispersed Pt demonstrates aggregation of Pt NPs revealed an average particle size of ~ 8 nm, as compared to dynamic light scattering which gave an average particle size of ~102 nm, likely due to measurement of aggregates.

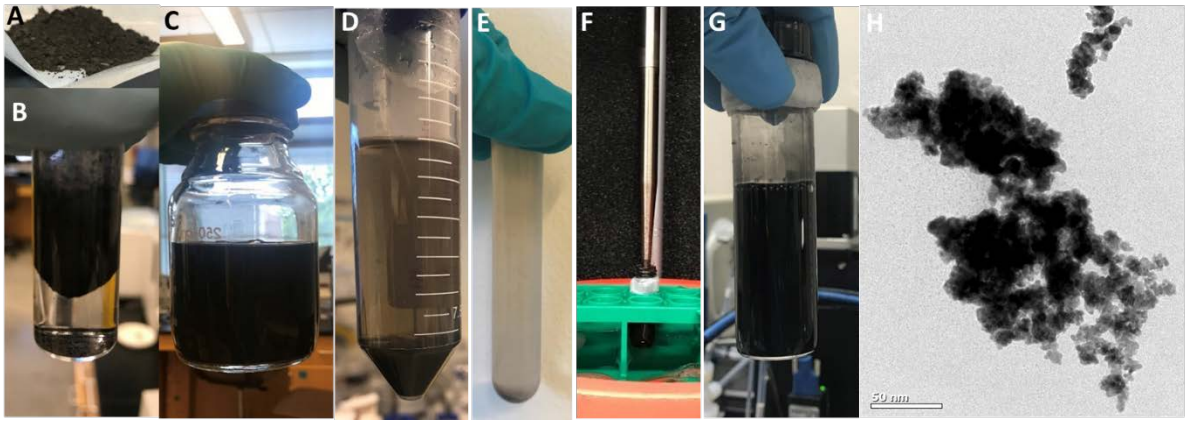
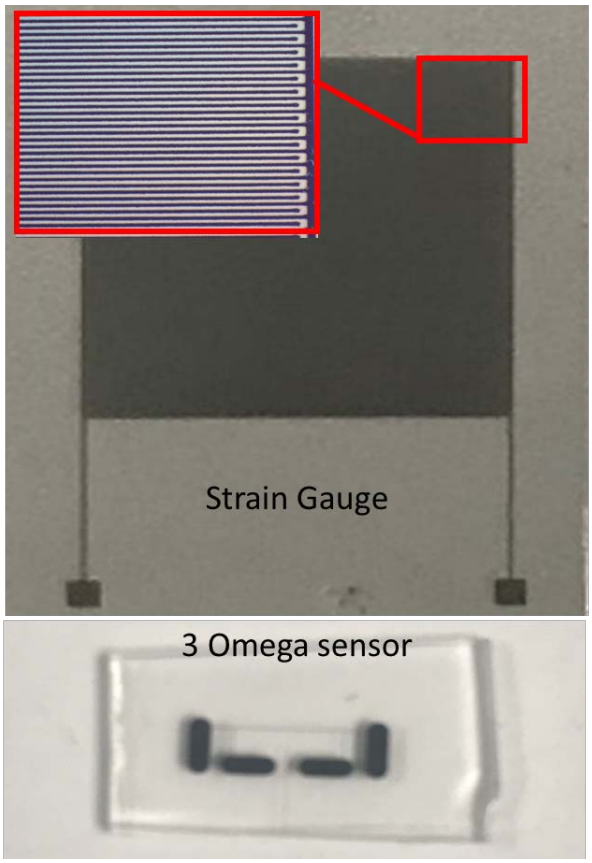


Figure 1. Top down Pt ink synthesis. **A)** PVP Pt nanopowder. **B)** PVP capped powder in water. **C-G)** Images of Pt ink synthesis process steps. **H)** TEM images of Pt particles from ink solution



Pt nanoparticle ink was then placed in the UA-MAX ultrasonic atomizer of an Optomec AJ-200 aerosol jet printer to demonstrate its compatibility with AJP. A recirculating bath temperature of 15 °C was used to help stabilize the ink temperature and to maximize the output concentration. The tool platen, nozzle diameter, and atomizer and sheath gas flows were optimized to ensure the line widths and material deposition were sufficient per device constraints. Two types of sensors were printed. An interdigitated capacitive strain gauge was printed. The device consisted of 100 digit pairs for the device electrodes with a 25 µm spacing (Fig. 2). Additionally, a 3-omega sensor was printed for insertion into the NCSU reactor through a separate NEUP/NSUF award (Fig. 2).

Figure 2. Pt based in – pile nuclear sensors printed at Boise State University using in – house manufactured Pt ink. (Top) – Interdigitated Capacitive Strain Gauge. (Bottom) 3 – omega sensor for insertion into the NCSU reactor.

3.1.2 Top down Mo ink synthesis

Based upon our results developing a Pt ink, we adopted similar methods for Mo ink development. PVP capped Mo nanoparticles (powder) were purchased from US Research Nanomaterials (Fig. 3A). 20 g of Mo powder was slowly added to 20 mL of vigorously stirring nanopure water (1 g every 30 minutes) to disperse the particles for ink development (Figs. 3 B,C). Once developed, the ink was analyzed by TEM to evaluate the particle size. Particles were found to range from ~10 nm to >100 nm (Fig. 3D). The ink was then loaded into the pneumatic atomizer of the Optomec 200 AJP system to print Mo based dosimeters of 1 cm by 1 cm dimension (Figs. 3 E,F).

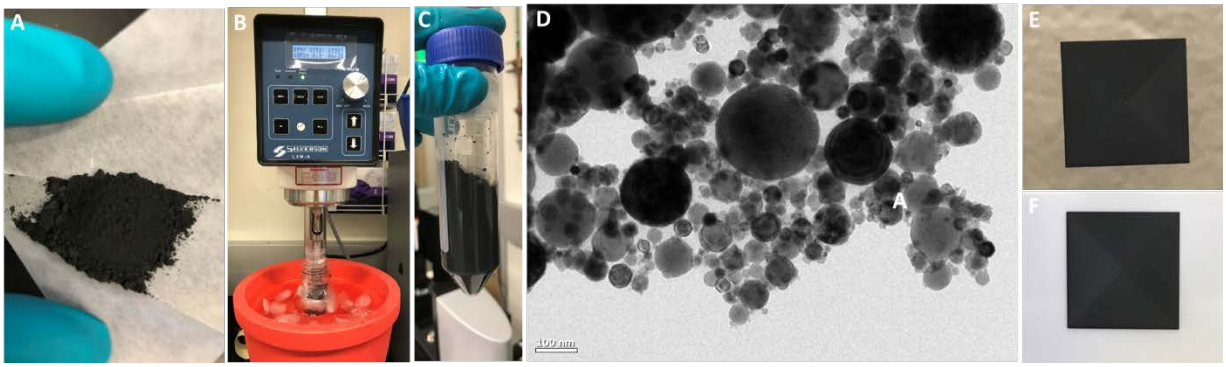


Figure 3. **A-C**) Mo based nanoparticle ink process. **D)** TEM Image of Mo nanoparticles. **E,F)** Mo dosimeters before (E) and after annealing at 300 °C for 30 minutes.

3.1.3 Top down Nb ink synthesis

Based upon our results developing a Mo ink we developed a Nb ink for printing passive dosimeters. The process was identical to Mo ink development. Figure 4 highlights our Nb process as well as TEM characterization of Nb nanoparticles. The ink was printed in 1 cm by 1 cm Nb passive dosimeters using the ultrasonic atomizer of the Optomec 200 AJP, highlighting versatility of our process with the AJP system.

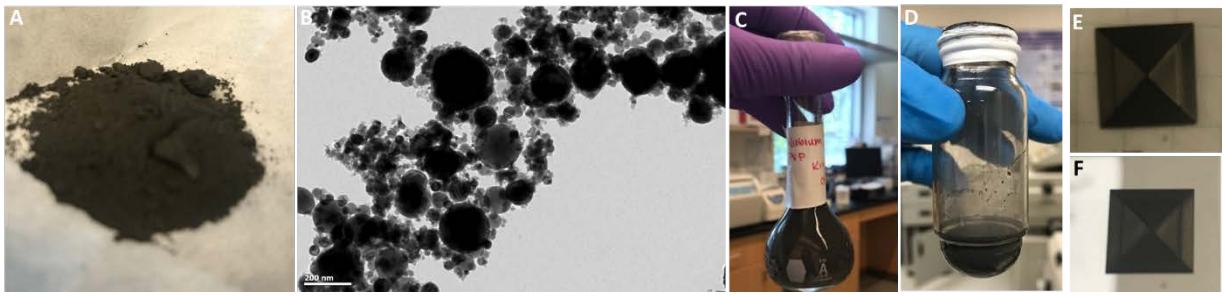


Figure 4. **A, B**) Nb nanoparticle powder and TEM images of Nb nanoparticles. **C,D)** Large scale Nb ink synthesis and Nb ink the UA vial of the AJP 200 system. **E,F)** Nb dosimeters before (E) and after annealing at 300 °C for 30 minutes.

4. Conclusions

Our results highlight our ability to synthesis inks compatible with AJP and PJP. We synthesized Pt, Mo, and Nb inks and demonstrated their printability for in – pile nuclear sensor applications. These results meet our milestone requirements for ink development and the inks are now available for manufacturing sensors designed in the other I2 work packages. Work will continue developing additional metallic and ceramic ink and use the produced ink in sensor production.