

HIGHLY SCALABLE DEUTERATED POLYMER DEVELOPMENT FOR IFE TARGETS

George Larsen and Tyler Guin
Savannah River National Laboratory

Jorge Sanchez
Lawrence Livermore National Laboratory

Andrew Schmitt
Naval Research Laboratory

1. EXECUTIVE SUMMARY

Inertial fusion energy (IFE) relies on the laser-driven compression of a target to generate fusion conditions. For direct drive IFE, the target is a spherical capsule, and for indirect drive IFE, the spherical capsule is further placed within a metal hohlraum. The capsule typically consists of D-T ice encapsulated within a protiated (C-H) or deuterated (C-D) polymer. Materials other than polymers have been proposed for use as encapsulants, each having its own advantages and disadvantages.¹ However, polymer-based capsules have the advantages of a relatively high technology readiness level (TRL), lower cost, and simpler fabrication. Thus, they remain the leading candidate for IFE facilities, especially IFE power plants, where as many as 500,000 targets per day will be needed.² In spite of the high TRL for existing polymer capsules, the current target fabrication process cannot be scaled to meet the unprecedented fueling demands of an IFE power plant. Microfluidic-based fabrication³ and low-density polymer foams⁴ are representative of some concepts proposed in the literature to meet the scaling challenges. However, in addition to the challenges associated with scaling the capsule formation and enhancing D-T impregnation and ice solidification speed,⁵ there is also a need to develop highly scalable deuterated polymer production. The use of C-D polymer capsules will be critical to future IFE facilities, as the protium in C-H polymers may poison reactions in the fusion core and create an unmanageable load on subsequent isotope separation systems in the tritium processing plant. These effects are likely further compounded with the use of low-density polymer foams. It is therefore critical that deuterated polymer production capabilities are incorporated in and are developed to keep track of target design and manufacture.

2. BACKGROUND

Controlled hydrogen isotope exchange is applicable to a diverse array of industries. For example, replacing the normal protium (H) in an organic molecule with deuterium (D) or tritium (T) creates “tracers” to better understand chemical, biological, and pharmaceutical systems.⁶ The underlying process mechanisms can be elucidated by tracking the heavier isotope’s movement. Deuterated organics have also found use in organic light-emitting diodes (OLEDs) and optical fibers as means to increase stability and reduce data transmission losses, respectively.⁷ For IFE, deuterated polymers have been used to encapsulate hydrogen targets and map how shell material migrates into the fusion core. Historically, the production of isotopically exchanged organic material through catalytic isotope exchange is done by exposing an organic molecule-catalyst mixture in a heated, sealed vessel to D₂ gas or D₂O liquid.⁸ This batch process is inefficient and insufficient for the scalable production of highly deuterated organics as hydrogen isotope exchange is governed by the fundamental equilibria between the isotopologues in the two phases.⁹ Another issue is that the high pressure, high temperature environment of the sealed vessel may change the molecular structure of the target molecule. Alternatively, some simple deuterated organic molecules may be synthesized from deuterated precursors. However, the types of molecules that can be achieved in this manner are constrained by known chemical reaction pathways and the

availability of the necessary precursors.¹⁰ Thus, there remains a need to achieve selectively or highly exchanged complex organic molecules in a manner that does not require altering chemical structures.

As part of an ARPA-E GAMOW research project, SRNL has recently developed a record-breaking, continuous approach to synthesizing highly deuterated long chain aliphatic molecules from readily available precursors. Previous results in the literature for isotope exchange in similar long chain aliphatic molecules, only achieved <50 mol% conversion with very high catalyst loadings (>20 wt%). The SRNL team achieved >98 mol% exchange with less than 1 wt% catalyst (**Figure 1**). With further optimization, exchanges greater than 99 mol% are expected, and related work is ongoing. The SRNL process is highly scalable and can be readily adapted to polymers and precursors that are directly relevant to IFE targets. For example, SRNL's deuteration method is applicable to many conventional thermoplastic polyolefins, polyamides, and polyesters. Additionally, an important aspect of the SRNL process is that it leverages the expected hydrogen processing capabilities contained within the fuel processing plant and directly utilizes deuterium recovered within the fuel cycle (**Figure 2**).

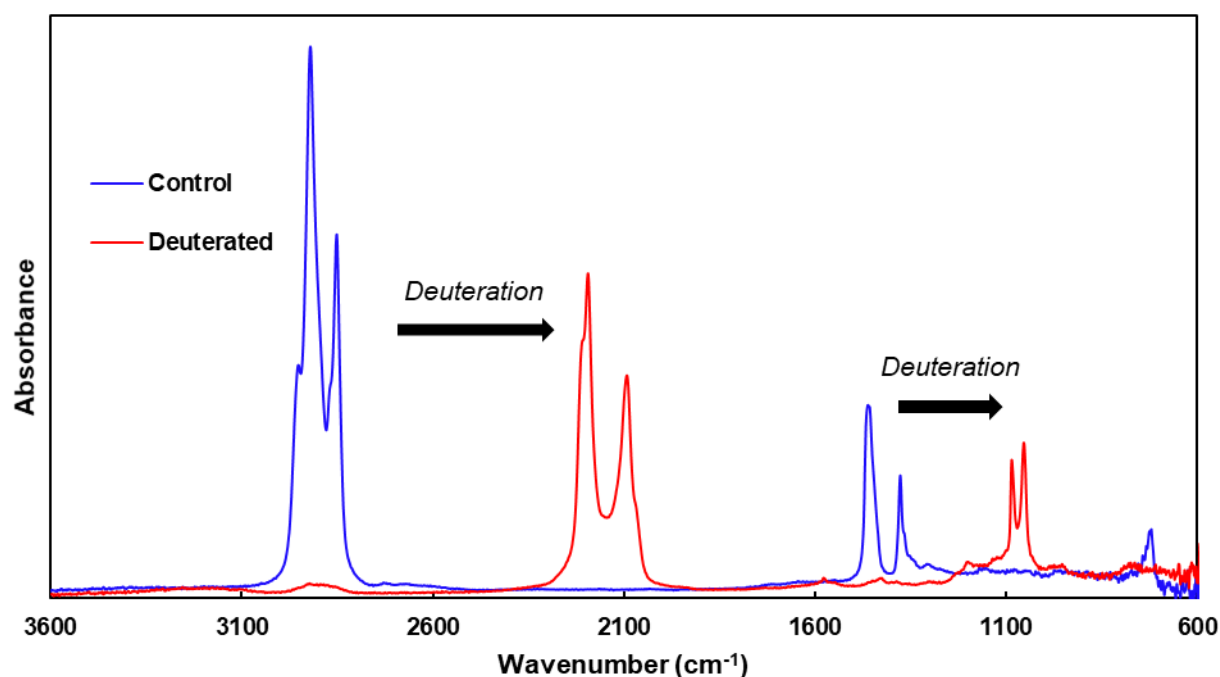


Figure 1. FTIR spectra of 500 M_n mineral oil $[(-CH_2-)_n]$ before (blue) and after (red) deuteration.

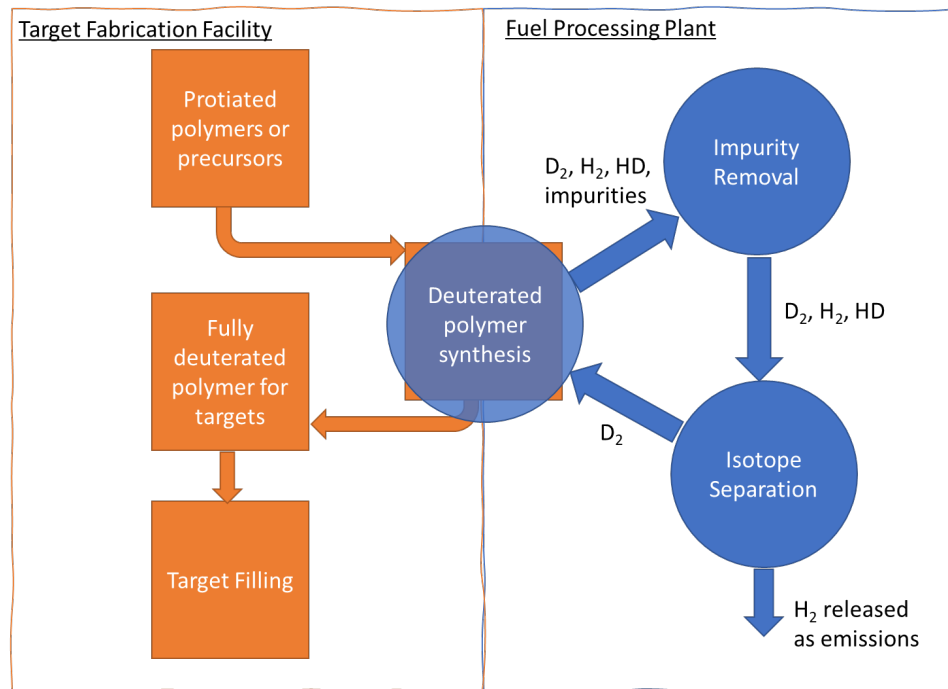


Figure 2. Schematic showing integration of deuterated polymer synthesis with target fabrication facility and fuel processing plant

3. RECOMMENDATIONS

Low density deuterated polymers with high deuterium/carbon ratios will substantially benefit both the core physics and tritium processing of IFE facilities. Therefore, deuterated polymer synthesis approaches should be developed alongside target design and manufacture. Further, deuterated polymer synthesis should be an integral part of any IFE target fabrication facility in order to take advantage of existing hydrogen processing infrastructure and maximally utilize the deuterium recovered from the fuel processing plant.

4. TEAM AND CAPABILITIES

Dr. George Larsen is a principal scientist in the Hydrogen Isotope Process Science Group at SRNL, where he provides tritium science expertise. His research covers all aspects of tritium processing, from basic to applied science and has been conducted in support of both NNSA and DOE Office of Fusion Energy Science. Dr. Larsen is also a PI on ARPA-E and INFUSE projects related to the tritium fuel cycle.

Dr. Tyler Guin is a senior engineer in the Hydrogen Isotope Process Science Group at SRNL, where he develops new materials for tritium relevant applications. His research primarily explores tritium compatible materials, tritium gas processing, novel catalysts, and polymer synthesis/upcycling.

Jorge Sanchez is a Research Scientist in the Materials Science Division at LLNL and is an expert in cryogenics, low temperature physics and engineering. His substantial research experience includes the development and design of cryogenic target fielding for ICF experiments. He is currently exploring better gas processing and handling for the Tritium Facility at LLNL.

Dr. Andrew Schmitt, the Head of the Theory Section in the Laser Plasma group at NRL, has significant experience in designing and modeling targets for direct drive inertial confinement fusion (ICF) applications. He also has experience identifying problem areas in ICF materials and systems.

Savannah River National Laboratory (SRNL) has over 50 years of hydrogen and tritium processing experience – more than 80 scientists and engineers and a \$20 million annual budget are dedicated to hydrogen and tritium defense missions. Over 50% of its more than 750 technical employees have advanced degrees—the majority in chemistry, engineering, and material science. SRNL has high-pressure hydrogen laboratories, a heat transfer lab, bench- and pilot-scale hydrogen storage and pumping systems as well as spectroscopic laboratories. SRNL’s relevant analytical capabilities for hydrogen studies, including ICP-ES, FTIR, and Raman spectrometers, and DSC instruments, are staffed by research-level professionals. SRNL also maintains TEM, SEM and X-ray diffraction (XRD) instrumentation for expert and detailed analysis of materials. The SRNL scanned probe microscopy laboratory (SPM) is housed in the HTRL. The SPM lab is equipped with both AFM and STM instrumentation that can be operated in ambient and controlled atmosphere. TVA and TGA analyzers are available to measure hydrogen uptake and release at a wide range of temperature and pressures. Three “inert gas glove boxes” are installed in a lab designed for synthesis and characterization of air-sensitive compounds. The majority of this work will be performed in the SRNL’s new Hydrogen Technology Research Laboratory, a 30,000 sq. ft. facility located in Aiken County’s Center for Hydrogen Research (CHR), which is dedicated to collaborative hydrogen research with university and industrial partners. The new CHR opened in February of 2006 and is a state-of-the-art facility committed to the advancement of hydrogen research, technology development and education.

5. REFERENCES

1. Goodin, D.; Nobile, A.; Alexander, N.; Petzoldt, R. In *Progress towards demonstrating IFE target fabrication and injection*, Proc. of the 2nd International Conference on Inertial Fusion Sciences and Applications, Citeseer: 2001.
2. Goodin, D.; Nobile, A.; Schroen, D.; Maxwell, J.; Rickman, W. *Cost-effective target fabrication for inertial fusion energy*; General Atomics, San Diego, CA (United States): 2004.
3. Li, J.; Lindley-Start, J.; Porch, A.; Barrow, D., Continuous and scalable polymer capsule processing for inertial fusion energy target shell fabrication using droplet microfluidics. *Scientific reports* **2017**, 7 (1), 1-10.
4. Dezulian, R.; Canova, F.; Barbanotti, S.; Orsenigo, F.; Redaelli, R.; Vinci, T.; Lucchini, G.; Batani, D.; Rus, B.; Polan, J. In *Equation of state data of plastic foams at Mbar pressures*, Frontiers of plasma physics and technology. Proceedings of an international conference, 2008.
5. Aglitskiy, Y.; Velikovich, A.; Karasik, M.; Schmitt, A.; Serlin, V.; Weaver, J.; Oh, J.; Obenschain, S.; Cochrane, K., Absolute Hugoniot measurements for CH foams in the 2–9 Mbar range. *Physics of Plasmas* **2018**, 25 (3), 032705.
6. (a) Daniel-Bertrand, M.; Garcia-Argote, S.; Palazzolo, A.; Mustieles Marin, I.; Fazzini, P. F.; Tricard, S.; Chaudret, B.; Deraud, V.; Feuillastre, S.; Pieters, G., Multiple Site Hydrogen Isotope Labelling of Pharmaceuticals. *Angewandte Chemie International Edition* **2020**, 59 (47), 21114-21120; (b) Bowen, G. J.; Wassenaar, L. I.; Hobson, K. A., Global application of stable hydrogen and oxygen isotopes to wildlife forensics. *Oecologia* **2005**, 143 (3), 337-348; (c) Lachawiec Jr, A. J.; Yang, R. T., Isotope tracer study of hydrogen spillover on carbon-based adsorbents for hydrogen storage. *Langmuir* **2008**, 24 (12), 6159-6165.

7. (a) Tsuji, H.; Mitsui, C.; Nakamura, E., The hydrogen/deuterium isotope effect of the host material on the lifetime of organic light-emitting diodes. *Chemical Communications* **2014**, 50 (94), 14870-14872; (b) Kaino, T., Preparation of plastic optical fibers for near-IR region transmission. *Journal of Polymer Science Part A: Polymer Chemistry* **1987**, 25 (1), 37-46.
8. Atzrodt, J.; Derdau, V.; Kerr, W. J.; Reid, M., C–H Functionalisation for Hydrogen Isotope Exchange. *Angewandte Chemie International Edition* **2018**, 57 (12), 3022-3047.
9. Sattler, A., Hydrogen/deuterium (H/D) exchange catalysis in alkanes. *ACS Catalysis* **2018**, 8 (3), 2296-2312.
10. Klenner, M. A.; Cagnes, M.; Wood, K.; Mita, K.; Kishimoto, M.; Darwish, T., Decagram scale production of deuterated mineral oil and polydecene as solvents for polymer studies in neutron scattering. *Polymer Chemistry* **2020**, 11 (31), 4986-4994.