Contract No:

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Disclaimer:

Laser-Based Means for Accelerating Nuclear Decay Rate

Project highlight. This project aims to reproduce and explain reports of using tabletop pulsed lasers and gold nanoparticles to promote faster decay of radioactive materials. Verification of this observation could be the first step in finding a new method for addressing long-term storage of nuclear waste.

Awards and Recognition

None.

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publicly published in its current form.

SRNL Legal Signature

Signature	**Date**

Laser-Based Means for Accelerating Nuclear Decay Rate

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Subcontractor: none

Project Type: Standard

Project Start Date: October 1, 2019
Project End Date: September 30, 2021

This project aims to replicate and build upon a reported series of results whereby nuclear decay of unstable species has been accelerated under laser irradiation in the presence of resonantly excited plasmonic nanoparticles. We are currently utilizing a laser system similar to that used within these reports as well as optimizing nanoparticle solution environments in an attempt to replicate reported results or provide alternative pathways that may explain some perceived discrepancies. The current work is being performed with equipment and techniques currently available at SRNL, including nanoparticle synthesis and pulsed lasers for plasmon excitation. The results of these experiments could provide quantitative data that can be used to resolve the discrepancies in the calculations and better estimate the ultimate utility of the process. If verified, this would represent a pathway for using readily available and scalable technology to reduce the long-term radiological storage requirements of certain forms of nuclear waste.

FY2020 Objectives

- Overall objective: investigate the accelerated α and β decay of radionuclides through exposure to the intense electric fields associated with laser-irradiated nanoparticles and nanopatterned surfaces.
- Reproduce observations reported in the literature.
- If observations are reproduced, establish quantitative relationships between experimental parameters and changes in the decay rates.
- If observations are not reproduced, propose and investigate alternative explanations for the reported results.

Introduction

This project aims to replicate and build upon a reported series of results whereby nuclear (α or β) decay of unstable species has been accelerated under laser irradiation in the presence of resonantly excited plasmonic nanoparticles. [1-2] Calculations support the possibility that a non-resonant interaction of an atom with an intense external electric field can alter nuclear processes, but disagree on the magnitude of the effect that can be expected. [3-7] Resonant excitement of plasmonic nanoparticles can lead to increases of electric field strength on the order of 10^5-10^6 near the nanoparticle surfaces. Laser pulses from commercially available, benchtop sources (such as a Nd:YAG laser) can thus be enhanced to 10^{18}-10^{19} W/cm^2, which is sufficient to excite relativistic energies in electrons. [8] In the reported work, fs, ps, and ns pulses directed into a solution of colloidal gold nanoparticles and dissolved UO_2Cl_2 altered the ^{238}U decay pathway, as detected by changes in the concentrations of daughter products ^{234}Th and ^{234m}Pa.

The significance of these results is that the equipment and materials used to generate changes in nuclear decay rates are much more readily accessible than synchrotron sources or free-electron lasers which

would access nuclear transitions directly [9]. The ability to accelerate decay rates would permit transformation of nuclear waste from a radiological and chemical problem at geologic time scales to a shorter-term chemical problem. Although significant engineering challenges would remain, not least of which being the determination of the form and materials for nanoparticles which can be used in massive quantities, the confirmation of these results would justify continued development in this field. Additionally, as implied in the above discussion, there is substantial disagreement between various calculations with respect to the magnitude of the effects that are possible. A more quantitative approach to irradiation parameters and the kinetics of decay and regrowth will inform and help to refine the calculations.

Work in FY20 followed two parallel paths. The main direction of work was towards setting up an experiment where highly energetic laser pulses could be safety focused into a radiological solution. This work was slowed by necessary repairs of the Nd:YAG laser that was procured for this purpose. We were able to design and test a reaction cell and focusing optics that allow us to maintain a large beam diameter at the window faces (preventing cell damage) and a tight focus inside the solution (maximizing power density). This arrangement should be ready for radiological testing in early FY21. The second path was an initial investigation of the chemical effects of laser-induced plasmas inside the solution on both the nanoparticles and dissolved materials. A possible alternate explanation for the disappearance of radionuclides is their incorporation into agglomerated precipitates, which were observed in the original experiments. It is likely that in the previous work, the agglomerated particles were not included in the radiological characterization. We have demonstrated the ability to make gold agglomerates from dissolved gold salts and will rapidly expand this work to determine if co-dissolved materials also are removed from solution.

Approach

Achieving the goals of the project brings together SRNL capabilities in nanoparticle synthesis and evaluation, lasers, radiochemical solution handling, and analytical measurements (rad and non-rad). Replication of the reported results will be accomplished by laser irradiation of uranyl chloride solutions with natural abundance ^{235}U. Solutions will be characterized before and after irradiation by gamma spectroscopy and mass spectrometry. If the equilibrium of the decay products is found to be altered, then variations in experimental parameters (laser characteristics, exposure time, types of nanoparticles) will be explored to try to improve yields and enhance mechanistic understanding. Not observing the same results would lead to an investigation of the chemistry of irradiated solutions, to try to find an alternative explanation for the reported observation.

Results/Discussion

Initial evaluation of the procured Nd:YAG nanosecond (ns) laser found repairs necessary for proper function at high output power (>2W). As of the end of FY20, all parts needed to fully refurbish the laser have been procured. A similar laser was used in experiments up to this point (Nd:YAG ns laser using the second harmonic at 1.5W output power with a 10 Hz pulse repetition interval). After constructing an optical pathway post-laser output (Figure 1), we found that standard cell configurations led to issues in both heat production and cell damage. Thus, we designed a new cell, which was made by the SRNL Glass Shop (see Figure 2). The cell was able to keep temperatures of irradiated solution around room temperature and was not damaged during experiments, which lasted up to six hours. During some experiments, where a transparent solution was used, the focus of the laser could be seen (Figure 3) and the laser-nanoparticle interaction could also be witnessed, as seen in Figure 4.

Figure 1: Optical setup. Pathway: laser tuner, turning mirrors, Galilean beam expander, beam focuser, sample cell, beam dump.

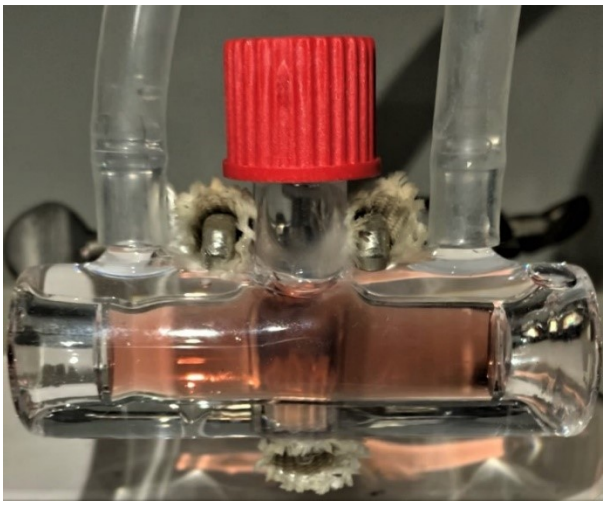

Figure 2: Designed cell containing a solution of gold nanoparticles with water flowing through the water jacket.

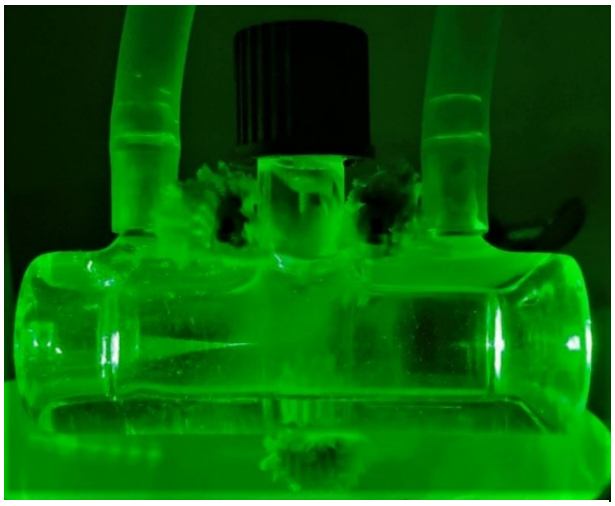

Figure 3: Focused laser light is witnessed in a gold nanoparticle solution demonstrating proper optical setup for both beam expander and focus lens.

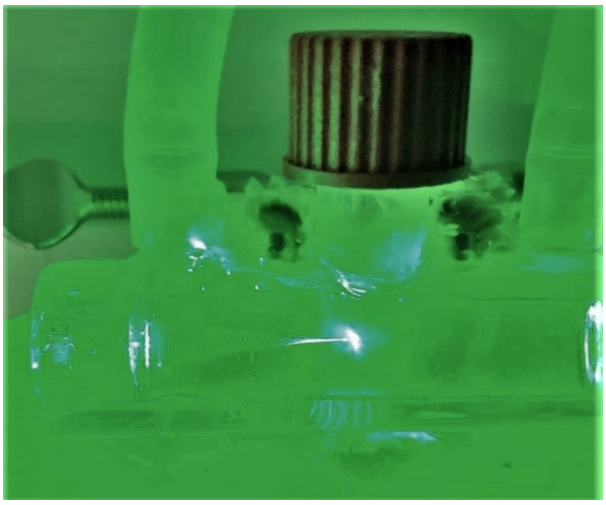

Figure 4: Focused beam interaction with gold nanoparticles creating a resonant plasmonic effect is witnessed. Note the bright flash and large area of effect where interaction occurs.

With a properly functioning cell in hand, baseline studies were conducted to ensure proper setup of optical system. A solution of gold tetrachloride ($HAuCl_4$) was irradiated by a Nd:YAG ns laser using the second harmonic (532 nm) at a power output of 1.5W for 2.5 hours. The characteristic plasmon absorbance peak for gold nanoparticles appeared, leveling off over time, indicating the formation of gold nanoparticles from the solution (Figure 5). Visually, there was a color change of the solution, a further indication that a product or mixture of products was produced. Other particulates that fell out of the solution when irradiated have yet to be characterized.

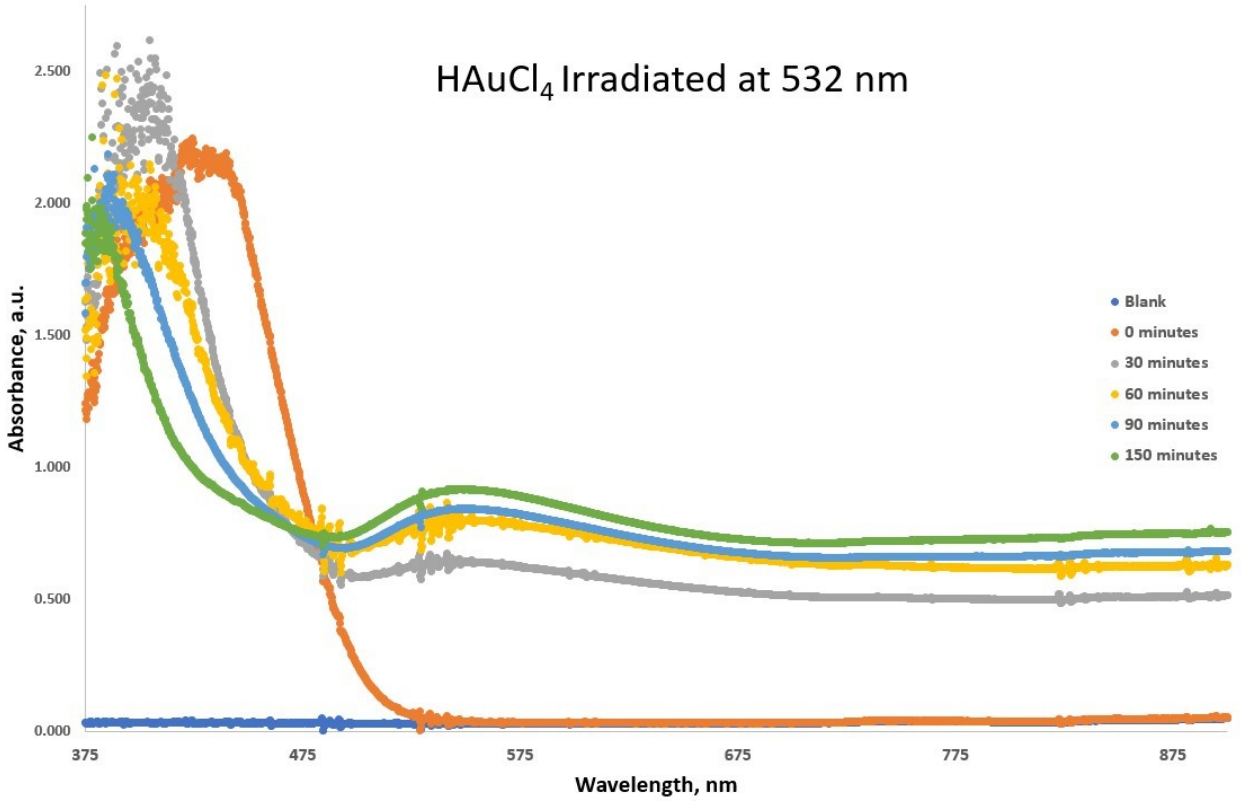

Figure 5: Absorbance plot of $HAuCl_4$ solution irradiated at 532 nm at 1.5W with a pulse reputation rate of 10 Hz for 2.5 hours. Note the growth and blue shift of the peak around 530-550 nm, characteristic of gold nanoparticles.

We believe this crashing out mechanism may be one of the pathways that led to discrepancies in the reported literature. Other metals and mixtures of metals are being tested to determine their ability to create nanoparticles, metal amalgams, or other types of mixtures that will crash out of solution.

FY2020 Accomplishments

- Refurbished Nd:YAG laser to optimize pulse power.
- Designed and tested optical and sampling handling setup. Developed new sample cell to withstand laser fluence and dissipate heat associated with sample absorbance.
- Conducted preliminary experiments to demonstrate that co-agglomeration is a potential pathway for removal of co-dissolved species from solution.

Future Directions

This project has been continued for FY21. Primary research efforts will include:

- Pursuing the original objective of replicating the reported observations and claims of accelerated nuclear decay. More rigorous chemical and radiological characterization of all products of the process will provide evidence for the fate of the radionuclides.
- A continued examination of the types of nanoparticles and agglomerates formed from solution under pulsed laser irradiation will not only help explain the original results, but also suggest a way to make new materials with interesting plasmonic properties.
- We will also pursue the modification of nanoparticles with actinide-sorbent shells. [10] The shells will be based on materials that have been shown to be selectively sorbent to uranyl nitrate in acidic conditions. By increasing the concentration of actinides near the nanoparticle

surface, it is thought that plasmon-mediated effects can be introduced more efficiently than if relying on random proximity within a solution.

FY 2020 Peer-reviewed/Non-peer reviewed Publications

None produced during FY20.

Presentations

None made during FY20.

References

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Acronyms

Nd:YAG – Neodymium: yttrium-aluminum-garnet

Intellectual Property

N/A

Total Number of Post-Doctoral Researchers

One post-doctoral researcher supported (Michael Thomas, started work at SRNL June 2020).

Total Number of Student Researchers

None.

External Collaborators (Universities, etc.)

None.