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Assessment of the doctoral thesis prepared by Zuzanna Krajewska

The thesis prepared by Zuzanna Krajewska is titled "Front-end investigations of the coated particles nuclear fuel for high temperature gas cooled reactors" and centers on the irradiation behavior of TRISO fuel particles, particularly the buffer and inner pyrolytic carbon layers of TRISO fuel particles. The thesis consists of seven chapters over 108 pages, including the introduction and list of references.

In my opinion, the main conclusion in the thesis is that ion irradiation and subsequent analysis of TRISO fuel particles show promise as an efficient method for evaluating fuel particle quality before insertion in a nuclear power plant.

Zuzanna Krajewska's thesis is built on five journal articles, three of which have been published:

1. Z.M. Krajewska, W. Gudowski, Raman Spectroscopy Studies of TRISO -Particle Fuel, International Conference on High Temperature Reactor Technology (HTR) 2021, Journal of Physics: Conference Series, 2048 (2021) https://iopscience.iop.org/article/10.1088/1742- 6596/2048/1/012007
2. Z.M. Krajewska, T.Buchwald, T. Tokarski, W. Gudowski, Front -end investigations of the coated particles of nuclear fuel samples – ion polishing method, Nuclear Engineering and Technology, Vol.54, p.1935-1946 (2022) https://doi.org/10.1016/j.net.2021.12.003
3. Z.M. Krajewska, T.Buchwald, A. Droździel, W. Gudowski, K. Pyszniak, T. Tokarski, M. Turek, The Influence of the Ion Implantation on the Degradation level of the Coated Particles of Nuclear Fuel Samples, Coatings 13, 556 (2023) https://doi.org/10.3390/coatings13030556

Two other papers, on ion polishing and ion implantation associated with TRISO fuel particles, have been submitted to journals. It is worth noting that Zuzanna Krajewska is the first author on all three published papers. These publications, and her leadership in each, are evidence of her talents and independence as a scientific researcher.

The first chapter of the thesis, the introduction, begins by reviewing the energy needs and nuclear power developments in Poland. TRISO fuel particles are introduced, including a listing of entities with current TRISO research work underway. The introduction touches on topics such as the need for spent fuel storage, past irradiation experiments, and the need for inspection of fresh fuel particles. Then, the goals of the research are listed: whether damage can be detected via Raman spectroscopy and whether ion irradiation can mimic neutron irradiation.

Chapter 2 centers on material damage due to irradiation. The chapter introduces the subjects of uranium isotopes, neutron interactions, ion interactions, and DPA (displacement per atom). Crystal damage is discussed, including Frenkel pairs and Schottky defects. The primary knock-on atom, or PKA, and displacement cascades are introduced. This is followed by an overview of nuclear reactors with an emphasis on high temperature gas cooled reactors (HTGRs). The fuel of HTGRs, pebbles and prismatic blocks, each containing thousands of TRISO fuel particles, is explained.

Chapter 3 gives a detailed review of TRISO fuel particles and their constituent layers. The composition and purpose of each layer is given. Failure modes are reviewed.

Diagnostic tools are the subject of the following chapter. Two polishing methods, mechanical and ion, are reviewed. (Mechanical polishing was discarded due to its tendency to damage the buffer layer. See Chapter 5.) Raman spectroscopy's characteristics are given in the context of evaluating fuel particles. The computational tools SRIM (Stopping and Range of Ions in Matter) and TRIM (Transport and Range of Ions in Matter) are explained.

Chapter 5 explains the experiments conducted. Three types of surrogate TRISO fuel particles were examined, one with kernel and buffer, one with kernel and pyrolytic carbon, and one with kernel, buffer, and pyrolytic carbon. Given an unirradiated sample, Raman spectroscopy was performed to give a baseline for comparison. A study was conducted to determine the energy to be used for ion implantation. The effects of different fluence levels on graphite were shown, leading to determination of a starting fluence for the particle irradiation. Next comes a central topic in the work, that of finding an ion irradiation level that will give comparable damage to a given neutron fluence. This general topic is one long discussed in the scientific community, and the thesis clearly explains the methods used to determine the equivalent ion irradiation fluence. At a level of 10^{14} ions/cm^2, noticeable damage has occurred. This is further reflected in Raman map images of the interface between buffer and pyrolytic carbon layers. SEM studies further clarified the extent of damage in the samples. One conclusion from this chapter is that after the equivalent of one year of neutron irradiation, crystalline damage has occurred with no noticeable additional damage with further irradiation.

Chapter 6 summarizes the findings. These include the need for high-quality polishing, the procedure for determining ion irradiation parameters that will lead to equivalent damage from set neutron irradiation levels, identification of 10^{14} ions/cm^2 as a threshold value for damage, and the evolution of the interface characteristics between the buffer and pyrolytic carbon with increased irradiation. Suggestions for further research areas are listed.

Chapter 7 lists the 177 references used in the thesis.

Three critical remarks are given below:

1. On page 96, "This primary fluence creates an interlayer between the Buffer-IPyC interface, which may lead to creating a gap between those layers." It is well known that a gap does indeed form between the buffer and IPyC, and that gap is driven by the shrinkage of the buffer. Whether gap formation is accelerated or decelerated by fluence is unknown. Further evidence on the connection, if any, between interface irradiation behavior and gap creation is needed.
2. Also on page 96, "In all analyzed cases, as the irradiation time of the samples increases, the Buffer-IPyC transitional layer is displaced, suggesting the formation of a Buffer - IPyC debonding." Like the previous item, more evidence is needed to support this claim.
3. The following statement is not justified from the results presented: "The research proves that for a fast screening method of produced but never used fuel or for a newly manufactured fuel, the ion implantation technique may be used to verify the quality of TRISO-particles before being placed the reactor core." Importantly, on page 97, this is clarified: "However, the results obtained in the mentioned scope should be compared with freshly manufactured TRISO samples within the occurrence of damage in the coating layers. Then the question of whether the damage in the TRISO sample layers is more progressive in 'old' (stored, but unirradiated) fuel than in 'new' (freshly manufactured) fuel could be answered." The second statement is much more satisfying and should, of the two, be the one emphasized.

Lest there be any mistake, I view these three criticisms as minor. The thesis presents much by way of background and original work that far outweighs these items. The first two are items for future research, and the third is editorial in nature.

The investigation of TRISO fuel particles is an activity underway by many organizations across the world, including my own. Thus, Zuzanna Krajewska's findings in this thesis contribute to widely-recognized need, that of developing economically feasible, low-carbon energy required internationally. It is relevant research.

To conclude, Zuzanna Krajewska's research, as captured in her thesis, presents a viable and repeatable procedure for sample preparation, ion irradiation, and microstructural evaluation of TRISO fuel particles. Her work, in my opinion, is both novel and valuable. I recommend that Zuzanna Krajewska be awarded the doctoral degree.

Respectfully submitted,

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