Neutronic Analogues for Water Detection Studies in Spent Nuclear Fuel

B. J. Green, D. Folley, A. Parker, A. Kennedy, S. Croft, R. Mills and M. J. Joyce

***Abstract* – The identification of benign (not radioactive) materials to serve as neutronic analogues for prompt gamma activation analysis specifically relating to the identification of water in spent nuclear fuel is described. Production of 2.223 MeV γ rays via the (n,γ) reaction can be indicative of the presence of hydrogen and hence water. Using a simulated ^{252}Cf neutron field, this research compares the neutronic properties of UO_2 and capture γ-ray spectra to those of several candidate analogue materials. Of the tested materials, molybdenum yields the closest correlation to UO_2 in terms of its neutron transmission and (n,γ) reaction rate.**

***Index terms* – 2.223 MeV γ ray, GEANT4, neutron capture, prompt gamma activation analysis, spent nuclear fuel**

I. Introduction

WATER present in spent nuclear fuel (SNF) and fuel element debris (FED) can undermine the chemical and structural stability of these materials and complicate the ease with which they might be deemed safe prior to dry storage and permanent disposal in a repository. However, whilst drying of SNF is therefore necessary, detecting the presence of water in SNF or FED across a significant inventory prior to drying is complicated by the intense radiation they emit. This can require that such techniques are stand-off and radiation tolerant.

A frequently arising constituent of the radiation from SNF is the neutron component arising mainly from the minor actinide, curium-244. This can be manifest via activation of hydrogen within the water in the emission of prompt 2.223 MeV γ rays via the (n, γ) reaction which is compatible with prompt gamma activation analysis (PGAA).

Simulations of the detection of these characteristic γ rays and proof-of-principle as an indication of the presence and mass of water within nuclear fuels was explored by Binnersley et al. [1]. This suggested this approach was superior to the use of neutron or γ-ray sources for the purpose of transmission measurements. However, understanding this approach and quantifying it experimentally with SNF is complicated by the inherent radiological hazard.

To understand the use of PGAA in SNF/FED analysis, we have analyzed the properties of a set of non-active, candidate analogues with the objective that they have material and neutronic properties consistent with SNF/FED. The analogues have been produced using additive manufacturing processes that allow the composition and mass of hydrogenous materials with them to be controlled.

This work reports on research of the simulated PGAA of candidate analogues using the Monte Carlo nuclear code GEANT4.

II. Material and Methods

This research is focused on Advanced Gas-cooled Reactor (AGR) fuel assemblies due to the UK's large inventory of fuel AGR SNF but given the UO_2 composition of this material it is anticipated the work might be extrapolated qualitatively to light water reactor (LWR) SNF too.

In addition to the prospect of water derived PGAA effects, the effect of the high burnup structure formed due to irradiation and the production of gaseous fission products must also be considered. At the rim region of a spent fuel pellet, the effective density is often lower due to higher porosities in this region of up to 12% [2]. These lower-density regions will result in an increase in the mean free path of neutrons traveling relative to the lower porosity structure closer to the center with γ-ray attenuation being altered similarly.

Using GEANT4, a variety of metals, oxides and alloys have been simulated. The simulants were chosen for their ease of acquisition, affording a mean free path similar to spent UO_2 (AGR) pellets (via the n,γ macroscopic cross-section), and their potential for use in a variety of related manufacturing techniques. Cerium dioxide (CeO_2) was tested as it is often used as a chemical analogue for UO_2 and a variety of other materials were explored including: SNF, 316L stainless steel, CeO_2, molybdenum, TiO_2 and C360 brass. The simulated SNF was configured to have an initial enrichment of 3.8% ^{235}U when fresh and a burnup of 28 GWd/tU as per a typical AGR regime.

Simulations used a 1 m × 1 m, 50 cm deep target sheet with four ^{252}Cf neutron sources, equally spaced and offset 10 cm from the bottom face of the target. A detector volume was placed over the sheet and was set to register any incident γ rays and neutron transmissions through the target material. Figure 1 shows a 3D rendering of this basic test geometry with the four neutron point sources visible beneath.

III. Results and Discussion

Ignoring the neutron emissions from induced fission of ^{235}U, the neutron and γ-ray spectra of the SNF are seen in Figure 2. The mean neutron energy incident on the detector was 0.929 MeV whilst the mean γ-ray energy was 3.290 MeV. A notable γ-ray peak is seen at 205 keV which is indicative of the (n, γ) reaction in ^{235}U. The low (n,γ) cross-section of the oxygen isotopes means contributions to the observed γ-ray spectra are minimal.

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R. Mills is with the UK National Nuclear Laboratory, Central Laboratory, Sellafield, Seascale, Cumbria, CA20 1PG, United Kingdom (e-mail: robert.w.mills@uknnl.com)

Table I provides the comparative number of neutrons and prompt capture γ rays incident on the detector from the same number of simulated ^{252}Cf decays. Molybdenum demonstrated the closest agreement to SNF with the number of recorded γ rays and neutrons by the detector.

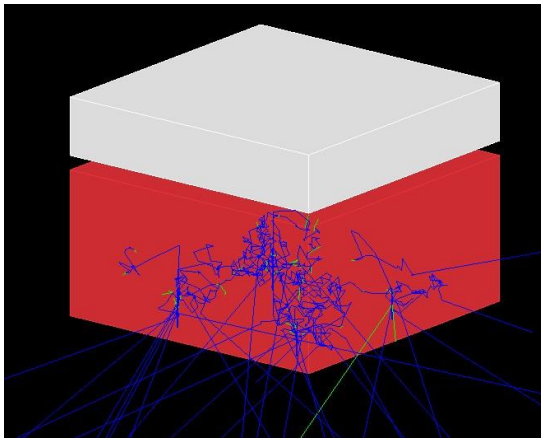

Fig. 1. The basic geometry model for assessment of SNF neutronic analogues. The red volume is the altered target material, the white is the detector volume, blue indicates neutrons and γ rays produced by (n, γ) reactions are green.

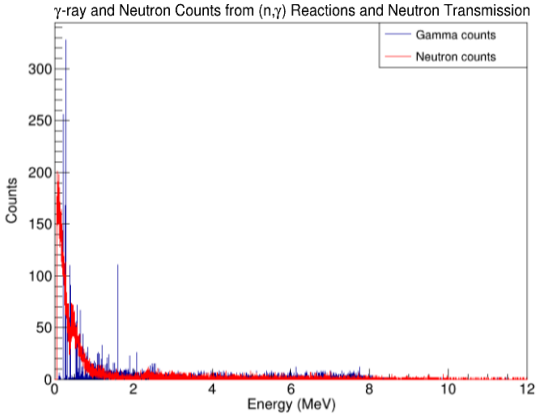

Fig. 2. Neutron and gamma energy spectra from ^{252}Cf decays passing through simulated SNF and (n, γ) reactions in SNF.

TABLE I
NEUTRONS AND GAMMAS RECORDED IN A DETECTOR FOLLOWING MATERIAL EXPOSURE TO CF-252 POINT SOURCES

Material	γ-rays	Neutrons
SNF	13,234	54,306
316L stainless steel	52,474	335,855
CeO_2	104,494	477,804
Molybdenum	11,273	48,815
TiO_2	154,922	263,455
C360 brass	34,897	197,802

The increased porosity observed in irradiated SNF was simulated by reducing the effective density of each material by 10%. The results of these changes on the number of neutrons and γ rays incident on the detector can be seen in Table II. The increase in recorded γ rays and neutrons varied greatly between materials from 6% for CeO_2 to 75% for SNF. Apart from SNF and stainless steel, the number of γ-rays incident on the detector for the lower-density materials increased by a smaller proportion than the number of recorded neutrons. This suggests that, for most materials, the change in γ-ray attenuation is less noticeable for small variations in material density.

TABLE II
NEUTRONS AND GAMMA RAYS RECORDED IN A DETECTOR FOLLOWING LOW-DENSITY MATERIAL EXPOSURE TO CF-252 POINT SOURCES

Material	γ rays	Neutrons
SNF	23,120	93,761
316L Stainless Steel	67,058	420,577
CeO_2	111,304	545,792
Molybdenum	18,325	81,212
TiO_2	193,515	366,309
C360 Brass	45,063	263,494

The molybdenum emission spectra are provided in Fig. 3. ^{95}Mo, ^{97}Mo, and ^{100}Mo responsible for most of the γ-ray emissions. Molybdenum-95 has the largest (n,γ) cross section with six γ-ray emissions between 2 MeV and 3 MeV. These may augment the observed 2.223 MeV count in a real detector due to attenuation and scattering effects. However, each molybdenum γ-ray emission cross-section is a factor of 10 or more smaller than hydrogen so the effect is not anticipated to be significant.

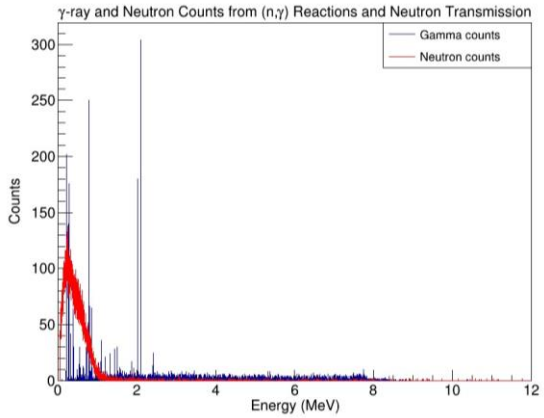

Fig. 3. Neutron and γ-ray energy spectra from ^{252}Cf decays passing through simulated molybdenum and (n, γ) reactions in molybdenum.

Verification of this research is limited currently by a lack of experimental data which forms the basis for future work.

IV. CONCLUSION

These simulations suggest that molybdenum may be a suitable neutronic analogue for spent nuclear fuel. In future, porous or mesh geometries will also be examined. AGR fuel pins and assemblies, both with and without water, will also be simulated to verify previous results [1].

Similar simulations will be carried out to determine an appropriate substitute for water that may be used in the laboratory with neutron sources to replicate the role of ^{244}Cm. Candidate analogues will be those with similar hydrogen densities to water but limited additional γ-ray emissions, particularly in and around the 2.223 MeV energy region.

REFERENCES

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