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Novel passive and active tungsten-based identifiers for maintaining the continuity of knowledge of spent nuclear fuel copper canisters

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ABSTRACT

A new approach to provide a long-term safeguards identification of spent nuclear fuel containers, in particular copper canisters, is presented in this paper. The approach proposes the use of a tungsten insert marked with a binary code and placed inside the container. The insert is read with a combination of two independent techniques, radiation and ultrasonic measurements, in order to get a unique identification of the cask. Passive and active versions of the tag are considered. The passive version makes use of the radiation coming from the spent nuclear fuel itself. The active version of the tag is based on the use of an artificially introduced mixture of α-emitting isotopes, such as ^{241}Am with materials, ^{11}B and ^{23}Na, which easily undergo α-induced reactions with emission of specific γ-lines, 2313 keV and 1809 keV, respectively. The paper discusses results of the radiation and ultrasonic measurements and Monte-Carlo evaluations as the first proof of the concept. The results of the investigations show the strong potential for this concept to maintain the continuity of knowledge of spent nuclear fuel copper canisters for a time scale up to a few thousands years without compromising the environmental safety of the casks.

1. Introduction

One of the specific acceptance criteria for radioactive wastes to be disposed in a deep geological repository is related to the presence of a unique identifier for each waste package (CFR 60.135, 2013; Limited, 2004) in order to provide a feasible monitoring for detecting the escape of the emplaced material. Additionally, the identifier/tag/label should address the question of maintaining continuity of knowledge of nuclear waste for a time scale up to a few thousand years without compromising the environmental safety of the casks (Herva et al., 2010). Thus, as it is shown in Chernikova and Axell (2014) an ideal tagging technique must satisfy a few intuitive requirements: be environmentally safe (e.g. "…the identification shall not impair the integrity of the waste package" (CFR 60.135, 2013)); allow the use of non-contact reader system; have a long operation time (up to a few thousand of years); be secure against falsification of data, errors/multiple verification; provide a large and unique tag memory (preferably, but not necessary).

There are a number of existing techniques which might be used as tags for nuclear waste casks. Among them, conventional tagging techniques (etching characters, affixing identification plates, welding), radio frequency (RF) tagging systems, electronic tags, ultrasonic systems (Demyanuk et al., 2013), Reflective Particle Tags (RPT) (Bennett et al., 2009) and SERS-Active Nanoparticle Aggregates (SANAs; SERS: Surface Enhanced Raman Scattering) (Brown et al., 2010).

The main disadvantages of these techniques are well analyzed in Culbreth et al. (1993), Culbreth and Chagari (1992), Chernikova and Axell (2014). For example, RF tags encounter problems with battery life (active RF), interference of the metallization layer with the RF signal, locating methods and low transmission range. RPT and SANAs tags can not be easily located inside the canister, and in the long-term run they have difficulties connected to the reader system, the image degradation, inconsistent calibrations, occasional reader head instability and false rejection rate caused by corrosion, as shown in tests performed in Hill et al. (1994). At the same time, the ultrasonic inspection of the welding area of the cask might provide a unique fingerprint for each stored container, but the whole concept is based on the assumption of the uniqueness of the welding area. Therefore, this method is suffering from the problems with unknown long-term signature stability and material sensitivity, as well as problems with repeatability of the signature (Hill et al., 1994).

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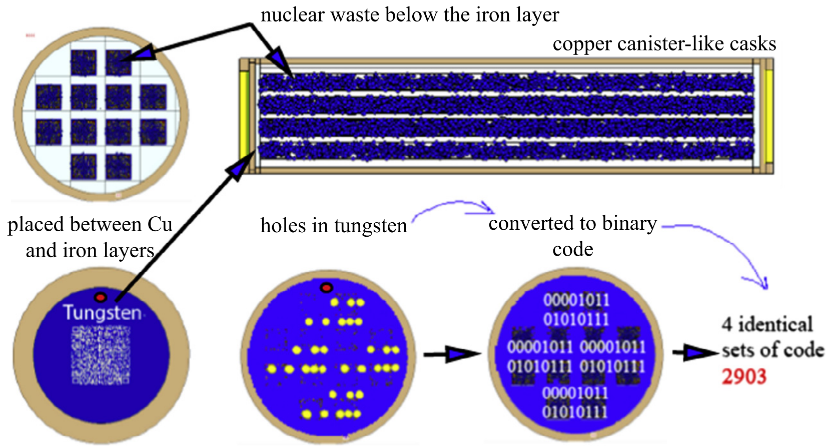

Fig. 1. A simplified illustration of an implementation of the proposed concept – part I (tungsten insert).

Thus, none of these existing tagging systems fully satisfy all "intuitive requirements", and there is a recognized and emerging need for another type of tagging method.

This study first proposes the new tungsten-based tagging approach for maintaining of continuity of knowledge of spent nuclear fuel copper canisters in the conditions of deep geological repository. Then, the proof of the concept is provided and discussed based on the results of experimental and numerical investigations. Finally, the specific future challenges associated with the problem and the possible ways of addressing them are mentioned.

2. The main concept

2.1. Description of approach

The new tagging approach consists of two parts. The first part is related to the use of a tungsten/lead insert marked with a binary or QR code[1] and placed inside the container between an iron layer and a 5 cm copper lid, as shown in Fig. 1.

Originally, two materials, tungsten and lead, were considered as inserts. The tungsten alloy was chosen after a preliminary study. Attenuation properties of tungsten are superior compared to those for lead, e.g. an attenuation coefficient in a lead for 500 keV gamma rays is 1.7 cm^{-1}, while for tungsten it is 2.14 cm^{-1} (Nelson and Reilly, 1991). Tungsten has also low toxicity and damage resistance (due to higher mechanical properties) compared to lead. Finally, tungsten has a good corrosion resistance, e.g. tungsten alloy with a composition of 93% W, 4.5% Ni, 1% Fe and 1.5% Co shows the lowest corrosion rate (Abdel Hamid and Hassan, 2013).

The tungsten disc does not impair the integrity of the waste package and can not be removed without breaking the integrity of the whole cask. If only a unique identification number is required for keeping records, we propose to use an ordinary binary code. One of the possible implementation of this tagging approach is shown in Fig. 1, where the particular waste package is identified as # 2903, or 0000101101010111 in binary representation. The binary numbers are implemented as combinations of holes in the tungsten plate, i.e. a presence of the hole in the tungsten insert reads as one (1) in a binary code. In order to provide a redundancy the tungsten insert is marked with four identical binary numbers.

The second part of the approach considers using a combination of two different reading techniques, radiation and ultrasonic measurements, in order to get the same unique identifier of the cask (see Fig. 2). The ultrasonic detector reads the discontinuities (in our case - holes) in the tungsten layer, thus providing the image of the distribution of the holes (unique identifier) in the tungsten.

In the radiation measurements the intrinsic or artificially included source of gamma rays can be used that corresponds to the passive (Fig. 3, r.h.s.) or active (Fig. 3, l.h.s.) versions of the tag. Thus, due to the strong attenuation properties of tungsten, in other words low transmission of gamma rays through tungsten tag, the variation in the gamma counting rate provides the distribution of the holes in tungsten insert.

2.2. Passive or active tungsten-based tags

An active version of the tag is based on the use of a mixture of α-emitting isotopes with long half-lives, such as ^{241}Am with materials, e.g. ^{11}B and ^{23}Na, which easily undergo α-induced reactions with emission of specific γ-lines, i.e. 2313 keV and 1809 keV, respectively. Thus, the use of a high-energy signature eliminates the problem with radiation background from the fuel, which is one of the advantages of the concept of the active tag. Among other advantages one may mention the long-term stability of the tag signatures (up to few thousand years) due to the long half-life of ^{241}Am. Apart from this, the use of an active tag can solve the existing problem of disposing smoke detectors (Chernikova and Axell, 2014; The European Commission, 2007) through recycling them into active tags. That, by turn, will partly cover the costs of the waste disposing. However, despite all advantages, the practical implementation of the active tag approach will require creation of a lab for radioisotope handling on the site of the encapsulation plant and it will also lead to longer encapsulation times and raise the additional safety problems with introducing extra radioactivity to the cask. Overall, in the present situation the practical use of active tags seems to be unrealistic.

The passive version of the tag, on the contrary, only requires radiation from the spent nuclear fuel, which depends on a burnup and a cooling time of the fuel. As an estimate, the intensity of gamma emission from typical spent fuel will be $\sim 3 \cdot 10^{14}$ photons/s/assembly (a BWR spent fuel with 37.8 MWd/kgU burnup at discharge, at 4091.7 days) (Tanskanen, 2000) which is enough for the passive tungsten-based tag to function. The gamma emission of the spent fuel will be changing with time. For a fuel cooled for a period between ten years and a few hundred years, ^{137m}Ba, the daughter nuclide of ^{137}Cs, will be the main gamma emitter. After a period of approximately a few hundred years the integrated gamma dose will be dominated by the gamma radiation coming from transuranic elements. The time evolution of the contribution of transuranic elements to the neutron emission (which is

[1] Quick Response code/a type of two-dimensional barcodes.