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Innovative Method to Authenticate Copper Canisters Used for Spent Nuclear Fuel Based on the Ultrasonic Investigation of the Friction Stir Weld

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ABSTRACT Sweden is planning to store nuclear spent fuel in long-term geological repositories. Copper canisters with a ductile iron insert will preserve the fuel for thousands of years at a depth of about 500 m in Swedish bedrock. The International Atomic Energy Agency (IAEA) and Euratom safeguards inspectorates have to maintain the continuity of knowledge (CoK) during transport and deposition of canisters from the encapsulation plant to the final repository. The aim of this paper is then oriented to provide such CoK for canisters by an identification and authentication based on ultrasound. This paper describes an authentication method oriented to prevent falsification of copper canisters. According to the ultrasonic investigation of the friction stir welding process of the copper lid onto the cylindrical tube, the ultrasonic response of the remaining gap partially filled during the welding is used as a unique pattern for authentication. The analysis of various measurements made on angular sections of copper lids friction stir welded onto the tube revealed the feasibility of acquisition of a valuable signal from the reflection of the internal gap. Following the observations on this data set, a possible technical design is detailed, where the identification and authentication methods can be combined with immersion probes.

INDEX TERMS Ultrasonic seal authentication, nuclear spent fuel, safeguard, copper canister, stir friction welding, immersion probe.

I. INTRODUCTION

The Seals & Identification Laboratory (SILab) of the Joint Research Centre (JRC) of the European Commission develops technologies and equipment based on ultrasonic control methods, suitable for sealing and identification of nuclear items. Nuclear safeguards applications require highly specialized and secure systems for identification, sealing, tracing, tracking and in general providing Continuity of Knowledge (CoK) in safeguarded installations. The simplest and oldest monitoring device is the seal, and it has to provide evidence of any access to the content of the sealed item, whether authorized or unauthorized [1]–[4]. In 2015 SILab started a collaboration with the Swedish Radiation Safety Authority (SSM) on the feasibility study of an ultrasonic identification system for copper canisters. The method for final disposal developed by Swedish Nuclear Fuel and Waste Management Company (SKB) involves disposing of spent nuclear fuel at a depth of approximately 500 meters in bedrock [5]. The copper canisters (Fig. 1) are nearly five meters high and one meter in diameter. The shell consists of a bottom plate, a tube and a lid and forms a 50 mm thick copper casing to protect against corrosion [6].

Copper is selected for a combination of several different factors, among which the heat transfer properties, chemical resistance, corrosion resistance, stress handling properties, stability in pure water. Inside the canister is an insert of nodular cast iron to provide the structural integrity required.

The final repository shall be designed so that it does not require maintenance or supervision. The reason for this is that there is no way to guarantee long-term (thousands of years) supervision and maintenance; this period of time is assumed necessary to protect people as well as the environment [7].

The possibility of applying some form of tag or seal to the copper canisters as well as to the transport casks are important steps in preserving the Continuity of Knowledge [8]. Several techniques might be used as tags for nuclear waste casks. Among them, conventional tagging techniques (etching characters, affixing identification plates,

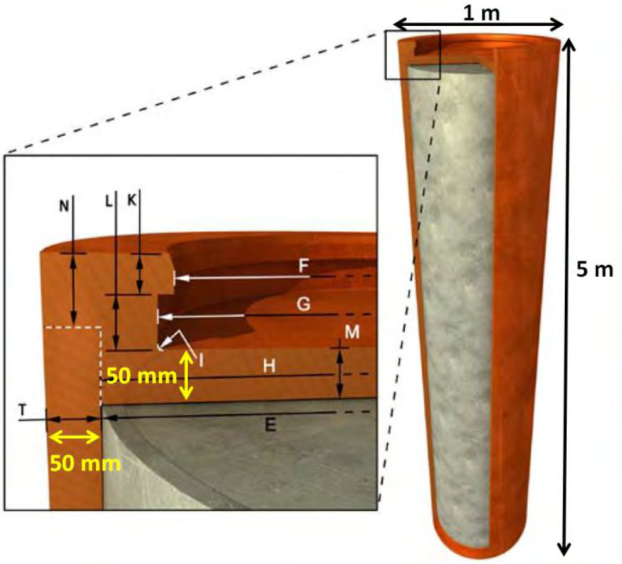

FIGURE 1. Copper canister geometry and section view. The relevant minimum values of copper thickness are reported T=M=50 mm [6].

welding), radio frequency (RF) tagging systems, electronic tags, Reflective Particle Tags (RPT) [9] and SERS-Active Nanoparticle Aggregates (SANAs; SERS: Surface Enhanced Raman Scattering) [10], tungsten-based identifier (active and passive) [11], reflective laser scanning and ultrasonic systems [12]. The ultrasonic tags can exploit both the ''unintentionally'' and/or ''naturally'' formed discontinuities in the material (the welding area of the cask, etc.) or could rely on the artificially created discontinuities by design (ultrasonic bolt, ''drilled holes of different depths'', etc.). Moreover, considering all possible methods, the ultrasonic approach is the only one that might have the natural uniqueness against duplication of fingerprints [13].

Nevertheless, this solution is based on the assumption of the uniqueness of the welding area that has not yet been demonstrated. A further step of research for the development of the ultrasonic authentication is the statistical analysis, which will determine the number of canisters that could be authenticated and the long-term stability and repeatability of the signature.

The aim of this paper is to report on the feasibility study of a new ultrasonic method for the identification and authentication of copper canisters used for long-term geological repository of nuclear spent fuel [8]. For corrosion reasons, external marking of canisters should be avoided. Therefore, the authors propose the ultrasonic reading of a series of chamfers (or flat bottom holes) milled on the internal side of the lid where the copper thickness is higher than 50 mm [14]. The chamfers are designed to maximize the reflection of the ultrasonic probing signal along optimal directions allowed by the canisters geometry. For the ultrasonic system, design has been proposed in [15] a 360° rotation of the probe along the circumference of the lid; this scanning method allows the acquisition of a binary code, different for each canister.

This paper is focused on the copper canisters authentication issue. The first paragraph describes the welding process of canisters by Friction Stir Welding (FSW) (II).

Afterwards the internal gap between lid and tube, partially filled by the material flow during welding, is investigated by contact and immersion methods. The experiments have been implemented on copper flanges that are portions of a full-scale copper lid welded onto the canister (III-A). Following the analysis of the internal gap by different transducers, additional investigations are accomplished to analyze the signal changes with temperature (III-B) and the reproducibility of measurements (III-C, III-D). Moreover, to validate the outcomes of the ultrasonic analysis, a comparison between ultrasonic and optical inspection is reported (III-E). Finally, a possible integration of the identification and authentication methods by an immersion scanning system is described (IV).

II. CHARACTERISTICS OF THE COPPER FILLED GAP CREATED BY FRICTION STIR WELDING (FSW)

An important background of this work is based on the welding process of the canister that is summarized in this section. Copper welding is usually difficult by conventional fusion welding processes because the copper has a high thermal diffusivity, which is about 10 to 100 times higher than many steel and nickel alloys. The heat input required is much higher than almost any other material, and weld speeds are quite low. To overcome these problems, the FSW is applied to the joining of copper.

FSW is a type of friction welding that was invented in 1991 at the Welding Institute in Cambridge, England [16]. FSW is a thermo-mechanical solid-state process, i.e. not a fusion welding method. This means that problems encountered in fusion welding – such as unfavourable grain structure, grain growth and segregation phenomena – can be avoided. The resulting microstructure in FSW of copper thus resembles the microstructure resulting from hot forming of the copper components in the canister.

The welding machine at the Canister Laboratory is designed for welding in air, however to investigate the effect on the welding process and the weld zone properties, welds in Argon gas have been produced using a provisional gas chamber. Before the welding begins, an inspection is made to ensure that the joint surfaces of the base and the tube are not damaged in any way and they are cleaned to reduce the probability of defects and impurities in the weld. The welding process consists of a rotating shoulder that heat up the metal by means of friction and, by its shape and rotation, forces the metal to flow around it, creating a weld between lid and tube. A slice of copper canister after the friction stir welding is shown in Fig. 2.

Once the FSW is completed, an extensive testing plan has been conducted at the Canister Laboratory to establish the performance of the welding process. Different Non-Destructive Testing (NDT) methods are employed from ultrasonic phased array to X-Ray [17], [18].

FIGURE 2. Demonstration of a piece of canister friction stir welded.

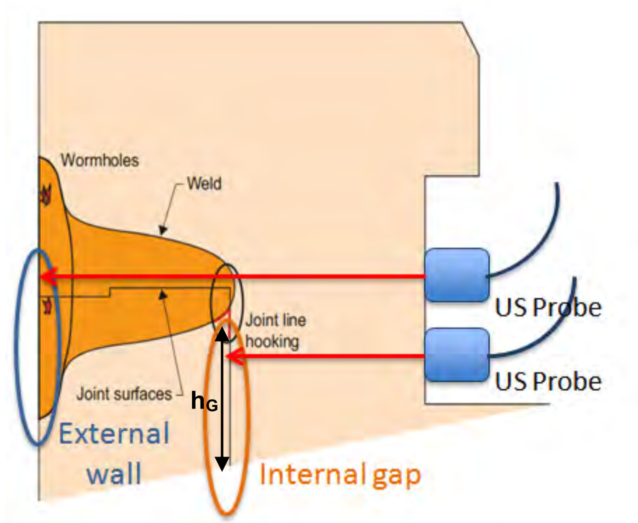

FIGURE 3. Welding spot shape due to the heating during FSW (orange area) and ultrasonic inspection schemes.

III. ULTRASONIC INVESTIGATION OF THE PARTIALLY FILLED INTERNAL GAP GENERATED BY THE FSW

The implementation of a method for the identification of copper canisters is not sufficient to prevent attempts of falsification or duplication of canisters. An authentication method is then necessary to ensure the originality of each container. The idea proposed in this paper is based on the research of a natural fingerprint, intrinsically contained inside copper and then unique for each canister. In particular, we focused our attention on the gap geometry between lid and tube of canisters after FSW. Tolerances for canisters machining should be such that there should not be any excess air gap between the lid and tube. In reality, to allow the installation of the lid on the tube before welding, a small gap of about 0.3-0.6 mm exists between them [19].

During the FSW process, because of the heat produced by the rotation of the shoulder, the gap between lid and tube could be partially welded.

Consequently, the canister results perfectly welded along the line of welding but beneath it, an air gap remains between the lid and the tube. Because of the presence of air in the gap, it represents a discontinuity detectable by an ultrasonic transducer placed as in Fig. 3.

Then the ultrasonic investigation of the gap with a rotating probe allows the acquisition of a pattern related to the change of the gap height (h_G) that is specific of the canister.

Even if FSW is a quite stable process, the quality of the weld depends on several fluctuating welding factors that could create imbalanced material flow. For that reason, the ultrasonic response of the internal gap could represent a very robust authentication fingerprint different for each canister.

The authors will develop statistical analysis to verify this condition when units will be available for testing. According to the assumption of uniqueness of fingerprints, several ultrasonic investigations are carried out with laboratory samples to verify the possibility of acquisition of the signal reflected by the internal

A. EVALUATION OF THE INTERNAL GAP ULTRASONIC RESPONSE

The results of several ultrasonic tests are illustrated in this paragraph. The aim of these measurements is to identify what is the best probe height for the acquisition of the ultrasonic echo reflected by the gap, while scanning the internal circumferential surface of the lid. According to a preliminary evaluation of the transducers characteristics for the high resolution scanning of the internal gap (12.7 mm probe focus diameter, 10 MHz frequency, 2.77 MHz bandwidth), we selected a V111-RM Fingertip Contact Transducers (Panametrics); this probe is then moved along the line A as shown in Fig. 4. Considering the element diameter D of 12.7 mm and the estimated velocity of sound in this copper material $V_C = 4651$ m/s, the near field length N of the probe in copper materials is calculated as in "(1)":

$$N = D^2/4 \cdot V_C = 86.7mm \tag{1}$$

This value represents the distance where the ultrasonic beam of the probe remains focused with a focal diameter equal to D.

The distance d_W between the probe and the internal gap is around 50 mm, while the distance S_1 between the probe and the external wall of the canister is about 100 mm, as illustrated in Fig. 5. Therefore, the selected transducer is able to receive the echo scattered by the gap with a good signal to noise ratio.

Moreover, considering the geometry of the lid and the tube, the height of the internal gap should be around 75 mm from the bottom of the lid.

The ultrasonic echoes are obtained with the data acquisition software for the Lecoeur US-Box [20]. The settings of the US-Box are: voltage 200 V, emission width 11, P.R.F. 2 kHz, gain 33 dB The uncertainties of measure are about 1.9 % for the amplitude and 0.05 μs for the Time Of Flight (TOF). The following screenshot in Fig. 6 shows the relative amplitude (range $\pm 100\%$) of the echo received by the transducer

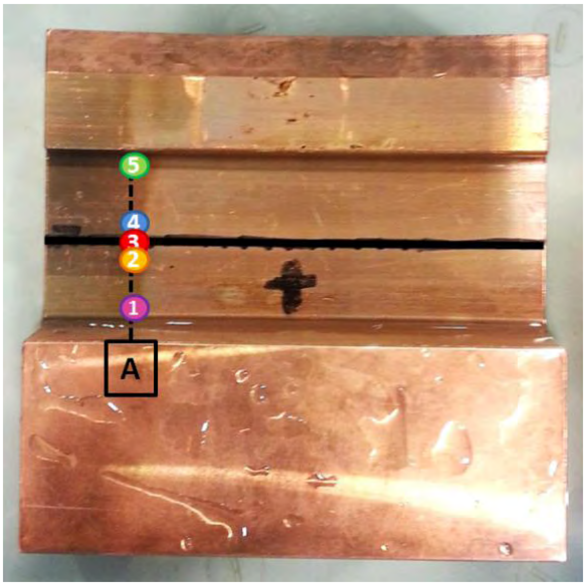

FIGURE 4. Positions of the probe along the inspection line "A". This line starts from the top plane of the copper lid. Its position has been chosen not too close to the left end of the sample to avoid the edge effect of multiple reflections.

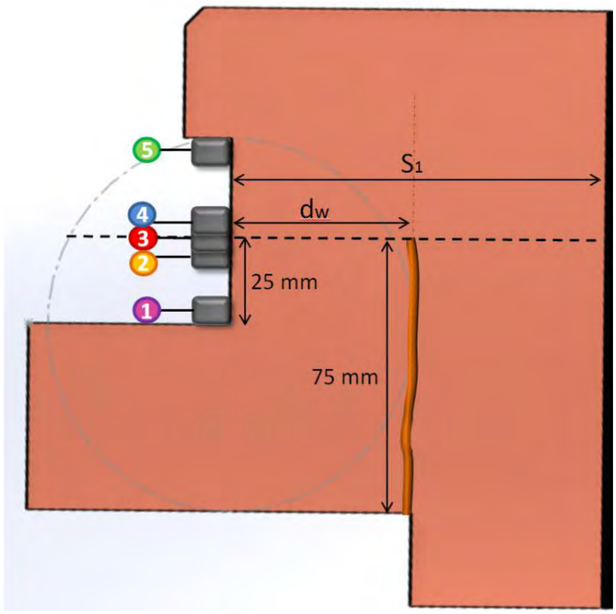

FIGURE 5. Positions of the transducer and distance of the internal gap. By design of copper canister, S_1 is equal to 100mm.

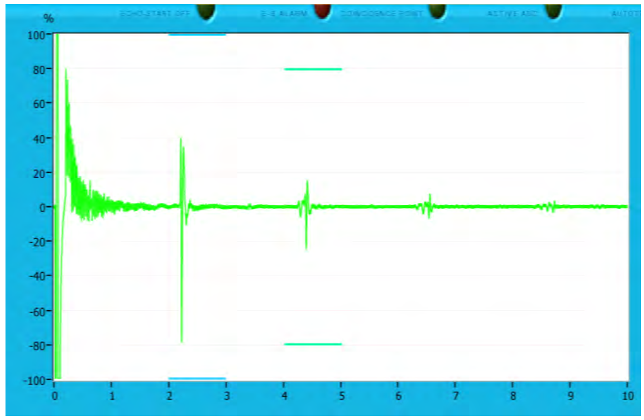

FIGURE 6. A-scan visualization of the echo due to the internal gap in position 1.

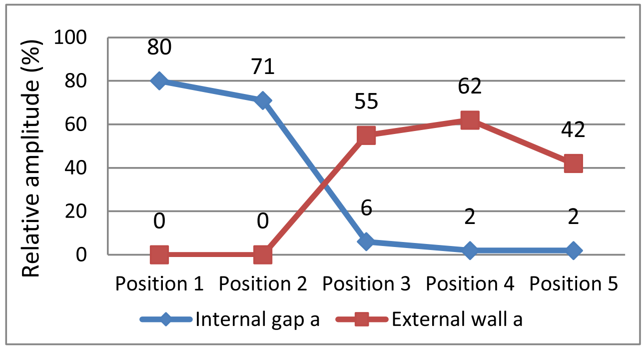

FIGURE 7. Relative amplitude of ultrasonic echoes of internal gap and external wall from positions 1 to 5 measured by transducer *a*.

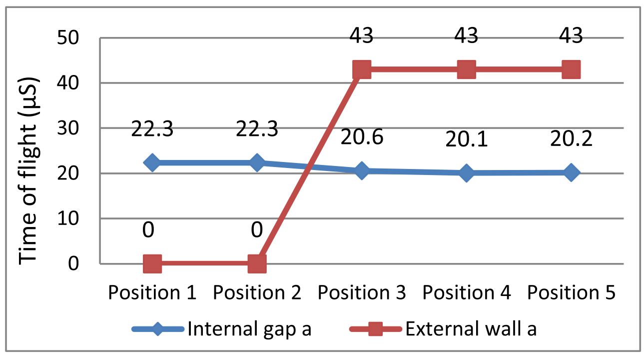

FIGURE 8. Time of flight of ultrasonic echoes of internal gap and external wall from position 1 to 5 measured by transducer *a*.

placed in position 1 (see Fig. 4), in the acquisition time interval of 100 μs.

In this probe position, the echo of the internal gap and its multiple path repetitions can be clearly identified at 22.3 μs and 44 μs respectively; instead, there is no detectable reflection from the external wall of the flange. By the TOF automatically measured with the gate function available in the US-Box software, dw is calculated with "(2)"

$$d_W = (V_C \cdot \text{TOF})/2 = ((4.651\text{mm}/(\mu s)) \cdot 22.3\mu s)/2 = 51.9\text{mm} \quad (2)$$

The results of the inspections in each position are shown in the graphs in Fig. 7 and Fig. 8 in terms of relative amplitude and time of flight respectively.

The results of the investigation in position 2 show that the signal of the internal gap is appreciable while the external wall echo is still not detectable.

In position number 3, instead, the amplitude echo of the gap is lower than in position 1 and the external wall echo (at distance S_1) is seen. This means that moving from position 1 to 3 the internal gap gradually disappears and then its ultrasonic echo decreases in amplitude. In position 3, it is possible to evaluate the distance S_1 as in "(3)":

$$S_1 = ((4.651\text{mm}/\mu\text{s}) \cdot 43\mu\text{s})/2 = 99.997\text{mm} \qquad (3)$$

We observe this value is very close to the theoretical one of 100 mm. In positions 4 and 5, the echo of the internal gap has completely disappeared (relative amplitude close to zero); it means that the gap between lid and tube is completely filled by the material flow during welding. In fact, the amplitude of the external wall echo increases while the TOF remains unchanged, as expected by the interpretation of physical model described in Fig. 5.

In conclusion, the internal gap height can be estimated by scanning along the line A. With a tolerance of the focal spot dimension related to probe radius (in this case equal to 8.25 mm), it has been verified that the internal gap echo disappears moving the probe between positions 2 and 3, then, in correspondence of line A, the gap height is between 66.75 mm and 75 mm.

The repeatability of these measurements is important for the realization of a method that can work in the field.

Furthermore, to guarantee a good acquisition of the signal, the most suitable transducer should be selected. Therefore, the same ultrasonic testing has been reproduced using different transducers with different dimensions and operating frequencies. For these reasons, the scan along the same line "A" has been repeated with the following transducers:

- Fingertip Contact Transducers – F Style (GE), Alpha series (126-000), 10 MHz, 0.5 in (transducer *b*);
- AeroTech GAMMA Transducer, 5 MHz, 0.25 in (transducer *c*);

Although it has been proved that at 5 MHz the attenuation of ultrasound in copper is quite low (less than 50 dB/m [21]), it is believed important to use a wide variety of frequencies, including high frequency probes (10 MHz) that provide higher axial resolution. The internal gap echoes obtained with these two different probes are compared with the data of the first investigation with the V111-RM probe (transducer *a*) and illustrated in the following Fig. 9 and Fig. 10.

The correlation indexes between the three curves are about 0.9 for the relative amplitude and 0.7 for the time of flight.

Therefore depending on the transducer used for the inspection there are some differences in measurements that could not be negligible. The selection of the most suitable transducer is then essential to acquire echoes with a good signal to noise ratio. Thus, transducer *a* best fitted our application and has been chosen for the following investigations.

B. STUDY OF THE INFLUENCE OF TEMPERATURE ON THE ULTRASONIC INVESTIGATION OF THE INTERNAL GAP

In the design of the new ultrasonic system for the identification and authentication of copper canisters, it is worth considering the temperature changes in the surface of canisters from encapsulation to deposition. Canisters are designed to withstand high temperatures caused by the radioactivity decay of the internal nuclear spent fuel [22].

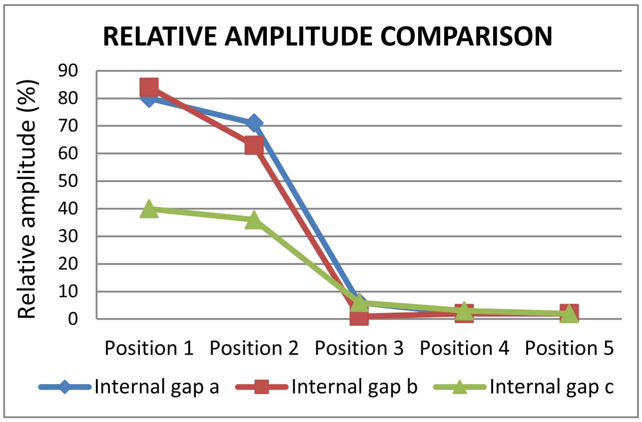

FIGURE 9. **Comparison between transducer *a* (V111-RM Fingertip Contact Transducers), *b* (F Style (GE), Alpha series 126-000), *c* (AeroTech GAMMA Transducer) in terms of relative amplitude.**

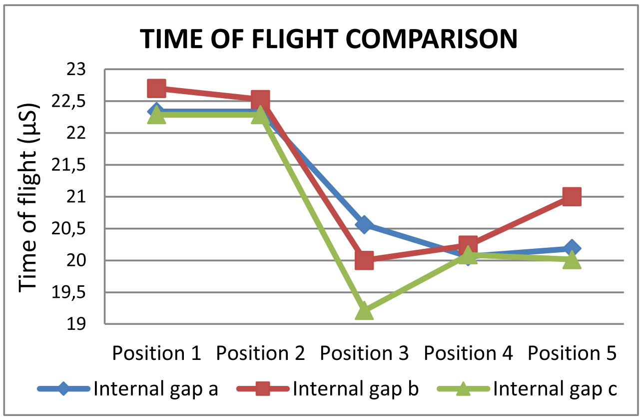

FIGURE 10. **Comparison between transducer *a* (V111-RM Fingertip Contact Transducers), *b* (F Style (GE), Alpha series 126-000), *c* (AeroTech GAMMA Transducer) in terms of time of flight.**

The temperature variation on the flange surface could affect the accuracy of the ultrasonic testing and it is therefore important to verify if there are some changes to the reflected echoes. The temperature on the outer surface of the canister is designed to never exceed 100° C. From the ultrasound point of view, the temperature variation could have a strong impact on the ultrasonic signals. The changes on ultrasound velocity in water or in copper could modify the relative amplitude and time of flight of the ultrasonic fingerprints. For the estimation of the operational temperature range, the effects of temperature on the velocity of sound are examined.

In our study, the couplant agent used for the ultrasonic testing is water that has a different dependence on the ultrasound velocity with temperature respect to copper. The reason for this choice lies on the easier automatic scanning of canisters and on the lower risk of contamination due to the evaporation of water after the ultrasonic reading. The speed of sound in water increases with rising temperature, as shown in the graph below (Fig. 11). We can observe that the maximum speed in pure water under atmospheric pressure is attained at about 74 °C. Ultrasound travels slower in hotter water after that point [23].

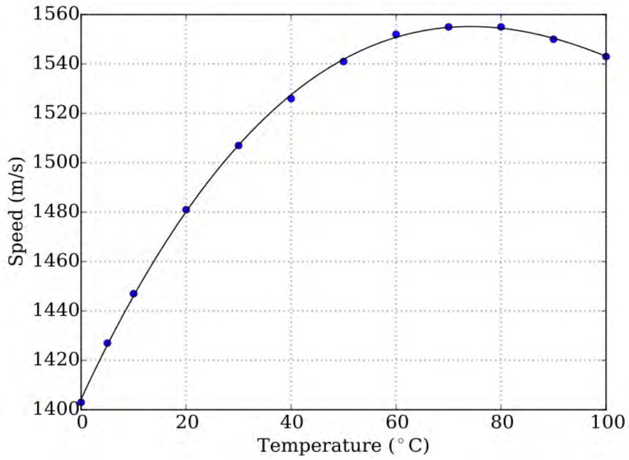

FIGURE 11. Velocity of sound in water with a temperature between 0°C to 100°C.

According to the canister design, the outer surface temperature of the canister is always lower than 100°C.

Therefore, the temperature of water involved in the ultrasonic readings could vary in a range from 0°C to 100°C. Passing these limits causes the inspection failure due to the water change of status. The velocity of sound in water could fluctuate from 1403 m/s at 0°C up to a maximum of 1555 m/s at 74°C with a percent variation of about 11%. Moreover, it is worth accounting the effects of temperature on the velocity of sound inside copper. The speed of sound depends on the Young's modulus and the density. Canisters are made of pure copper and the content of other elements is limited (phosphorus 30–100 ppm, Sulphur $<$ 12 ppm). The variation of the Young's modulus of copper with temperature between 0°C to 100°C is roughly 2% while the variation of density in the same range is around 0.5% [24], [25]. Then the percent variation of speed of sound in copper due to temperature (between 0°C to 100°C) is reported in "(4)":

$$\Delta V_c = 1.02/1.005 = 1.015 \tag{4}$$

This value is negligible with respect to the variation of the speed of sound in water. As a result, the temperature could influence the ultrasonic investigation of copper canisters because the variation of water temperature affects measurements.

However, the influence can be compensated using a temperature sensor attached on the external wall of the lid, this allows to measure the temperature during the ultrasonic testing and then use an algorithm to calibrate the signals which would then be referred on a unique scale.

C. COMPARISON BETWEEN DIFFERENT COPPER FLANGES

Aiming to fulfill a preliminary evaluation of the authentication capability of each canister, the ultrasonic scanning on two different copper flanges is carried out and the results are compared.

According to the analysis of the internal gap ultrasonic response in section III-A, the contact probe is placed at different heights corresponding to different circumferential lines on the internal part of the flange. The measurements are carried out with gel coupling at room temperature. As illustrated in Fig. 12, each flange is scanned along five lines at height h = 25, 23, 21, 19, 17 mm, from the top plane of copper lid.

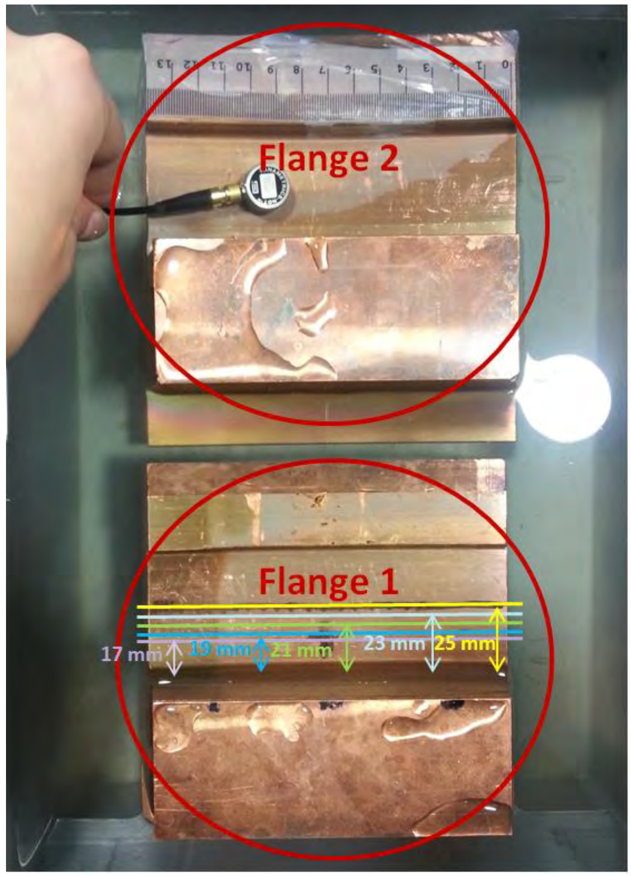

FIGURE 12. Investigation of two copper flanges at different heights.

All the acquisitions of TOF and relative amplitude of the internal gap echo and the external wall echo are summarized in charts reported in (Fig. 13 and Fig. 14).

The results analysis shows that each flange has a different signal pattern. This result is important because it opens up the possibility of authentication of each flange with a unique fingerprint depending only on the randomness of the FSW process. However, it is worth remembering that flanges are just laboratory samples and that the variation of patterns in samples coming from a production-like process could be lower.

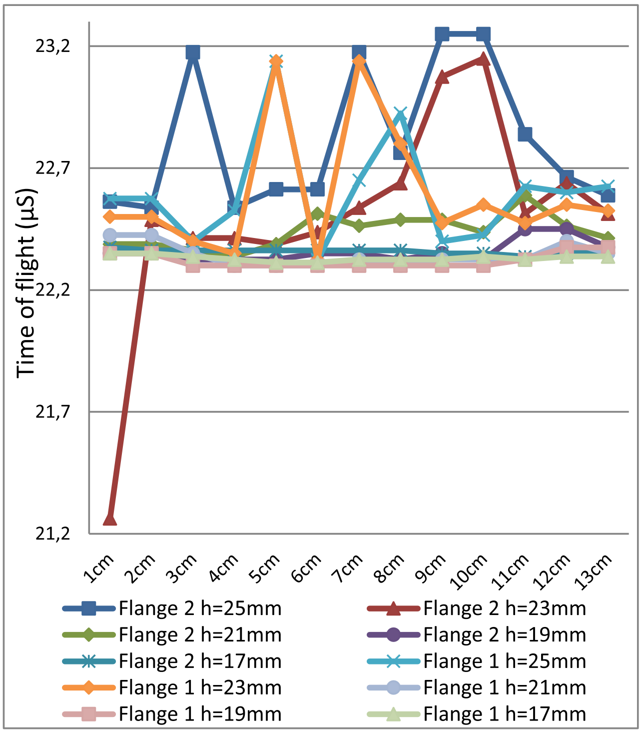

FIGURE 13. Comparison of TOF of the internal gap echo between flanges 1 and 2.

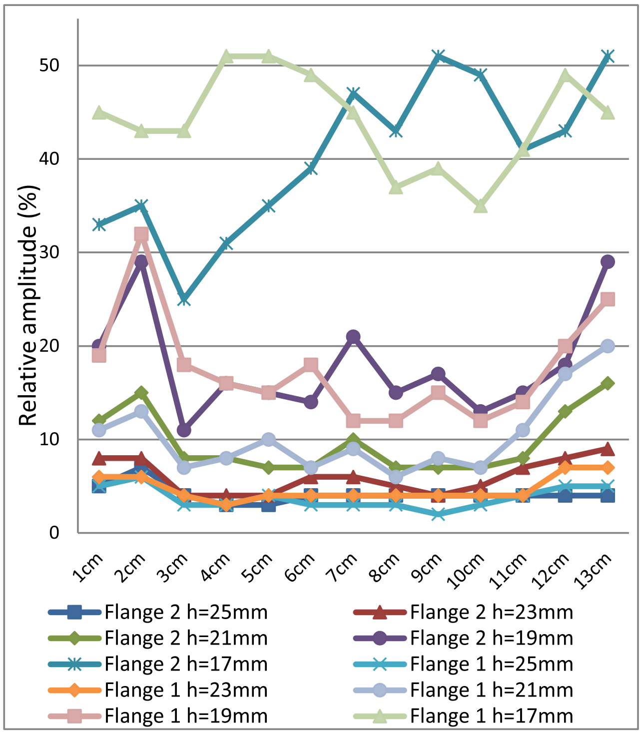

FIGURE 14. Comparison of relative amplitude of the internal gap echo between flanges 1 and 2.

D. VALIDATION OF THE METHOD WITH THE INVESTIGATION OF AN ANGULAR SECTION 50° WIDE OF A COPPER FLANGE

To validate the preliminary results obtained with small samples (22.3° slice), the investigation continued on a larger sample corresponding to a 50° slice of the copper lid welded with the canister tube. The following picture (Fig. 15) shows this additional flange sample. This new sample allows the inspection along a longer circumferential line and the edge effects at the two ends can be neglected.

Although the copper flange is scanned at different heights from the previous two flanges, it is now possible to collect more data than before because the length of each scan line is around 38 cm (instead of 13 cm, as in the previous case). Depending on the gap height due to the welding, the amplitude of the internal gap echo changes. Fig. 15 shows the copper lid scan by the ultrasonic probe along a line. The red circles marked as "A" and "B", represent the focal spots of the transducer. In case "A", the internal gap amplitude is higher by consequence that the external wall echo amplitude; vice versa, in case "B" the internal gap is lower and then the internal gap amplitude decreases while the external wall amplitude increases.

To access the reliability of the method, a comparison between different tests on the same line is carried out. The chart (Fig. 16) shows the received echoes in three different measurements along a line at height h = 17mm.

Observing Fig. 16, the repeatability of the ultrasonic investigation is verified because the trend of the signals acquired at the same line in different experiments is similar.

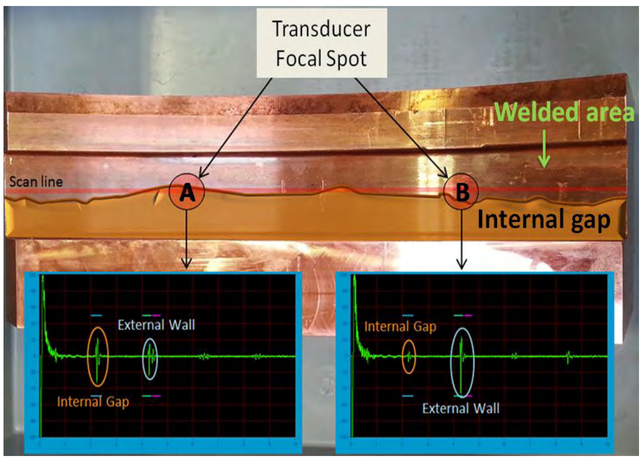

FIGURE 15. Ultrasonic investigation of an angular section 50° wide of a copper flange.

The correlation index between the internal gap curves is 0.95 while between the external wall curves is 0.93.

Calculating the ratio between the external wall echo and the internal gap echo at each measurement point, it is easier to appreciate that the difference between measurements is always less than ±3 %. This tolerance is also due to the manual repositioning of the probe which affects the quality of measurements. In the future, automatic scanning will improve the precision of the testing.

Moreover, considering the internal gap amplitude along the lines at height h = 15, 17, 19, 21, 23, 25 mm, an area chart of the heights of the internal gap is created (Fig. 17). This image illustrates the morphology of the internal gap that is a natural fingerprint of the canister. To conclude, we believe

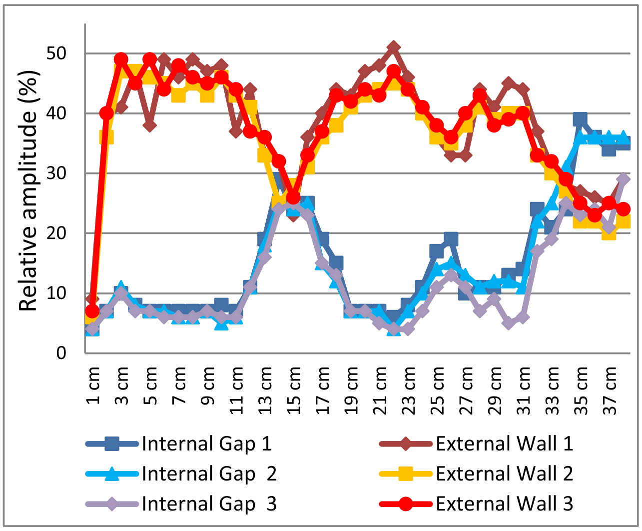

FIGURE 16. Comparison of the relative amplitude of three repeated tests at the same height h = 17 mm.

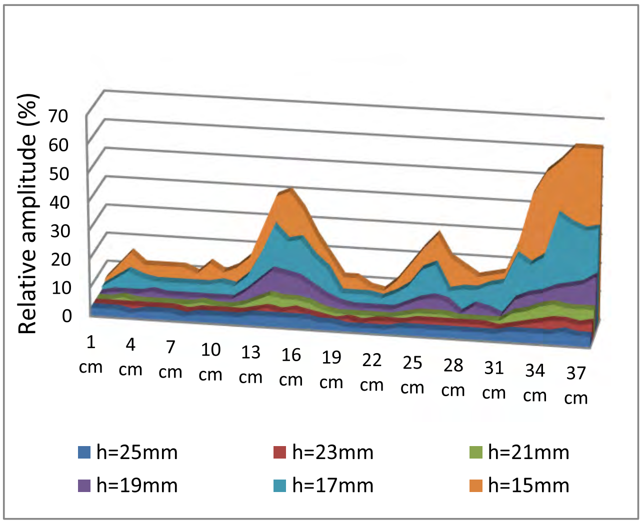

FIGURE 17. Area chart of the relative amplitude of the internal gap of the 50° wide flange sample.

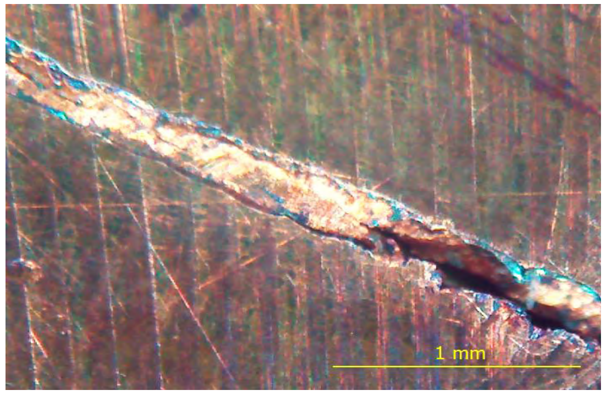

FIGURE 18. A zoommed image of the optical investigation of the internal gap of one section of flange 2.

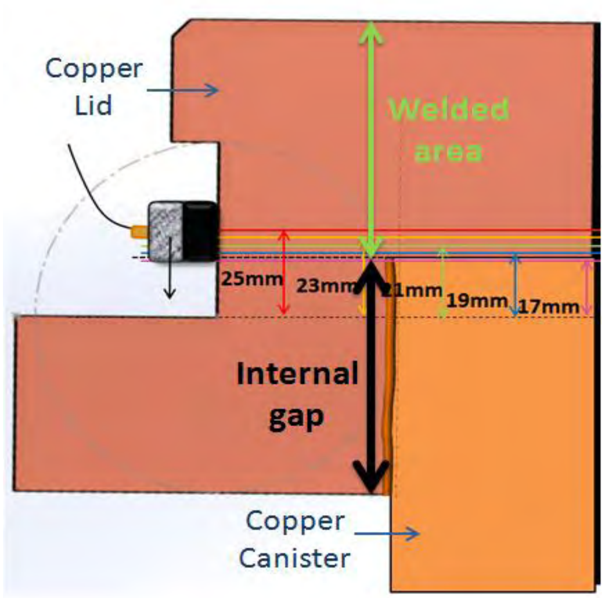

FIGURE 19. Ultrasonic investigation of the internal gap of one section of flange 2 across the end of the internal gap and the welded area.

that the internal gap between tube and lid after FSW could be used for authentication of copper canisters since the signal reflected by the gap is repeatable and it gives a specific pattern depending on the weld shape.

E. COMPARISON OF OPTICAL AND ULTRASONIC INSPECTIONS

To validate the ultrasonic investigation results and evaluate the internal gap height due to the FSW, optical testing is carried out using a Conrad digital microscope. The tiny gap (Fig. 18) between the lid and the canister welded together is barely visible to naked eye; however, the microscopic analysis can better show the microstructure of interface region in the friction welded joint.

Furthermore, the optical scanning allows the detection of welding defects like joint line hookings and wormholes.

After the optical inspection of flanges 1 and 2, the internal gap height results range from 66 to 68 mm. Therefore, in order to select a suitable transducer for the internal gap investigation, it is important to consider that the range of variability of the gap heights is limited to only 2 mm.

This value means the probe focal spot must be lower than 2 mm in order to guarantee the acquisition of a signal, which accurately reproduces the shape of the welding.

Fig. 18 shows a view of the internal gap scanned by the microscope on flange 2. The image reveals the presence of a gap (see bottom right) which is easily distinguished from the welded channel. Then, the discontinuity of acoustical impedance generates the echo signals that are used in this method to authenticate the copper canister.

The results of the optical investigation were compared with the ultrasonic inspection to determine the internal gap maximum height. As observed in section III, the amplitude of the internal gap echo varies along the different positions as well as the external wall echo; with reference to Fig. 17 the amplitude of the internal gap is more evident and clearly decreasing in height, around 19-15 mm. This means the end of the gap due to welding is located approximately at 69-67 mm from the bottom of the lid. The simplified drawing in Fig. 19 shows the transition between the welded area and the gap measured by the digital microscope (in black). We can conclude that there is a good correlation between the optical and the ultrasonic inspections.

IV. PROPOSAL OF AN ULTRASONIC SYSTEM FOR IDENTIFICATION AND AUTHENTICATION

The full task of the investigation of copper canister includes the study of solutions capable of integrating both the identification and the authentication methods. The results obtained in a previous work on an identification method using binary codes generated by 40° chamfers, reproducing the behavior of inclined flat holes on the inner side of the copper lid were described in [15]. The solution proposed by authors is the combination of the two methods by a rotating scanning system where two immersion probes can be used: one to investigate the internal gap for authentication and the other to read the codes generated by series of cavity milled on the chamfers of the copper lid (see Fig. 20).

The solution proposed for the ultrasonic scanning system for field inspections is illustrated in Fig. 21.

The curve giving the echo amplitude of the welding gap and the binary code will be registered on a 360° rotation. The internal gap fingerprint is then matched angularly with the binary code creating a third unique, random, robust identification and authentication solution.

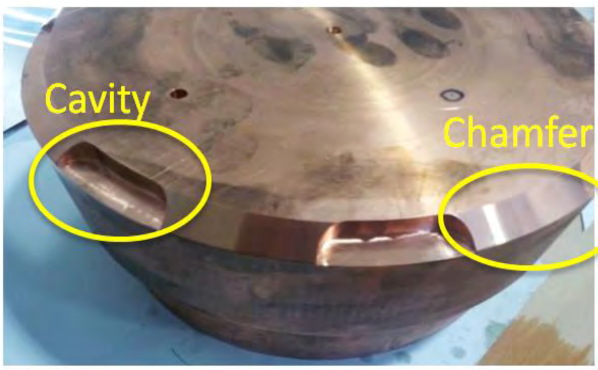

FIGURE 20. Identification with inclined reflectors: Cavity means no ultrasonic echo while chamfers means ultrasonic echo.

The internal marking of the binary codes is a low cost mechanical operation which is completed during the turning/milling of the copper lid. The reading system will operate remotely and will be put in place by a robotized crane and automatically centered on the lid. Once in place, water included in a tank will be released and poured on the external concave lid part, making an ideal and easy solution for the ultrasonic probe coupling. The immersion method should also improve the reproducibility of measurements. After a 360° inspection, the two curves are registered, stored or sent wirelessly to a control station. Water could be pumped back or left on the lid; in fact, the heat of the inner spent fuel will evaporate the small quantity of water rapidly. Moreover, electronics will be properly shielded in order to avoid problems due to the dose rate at the canister surface.

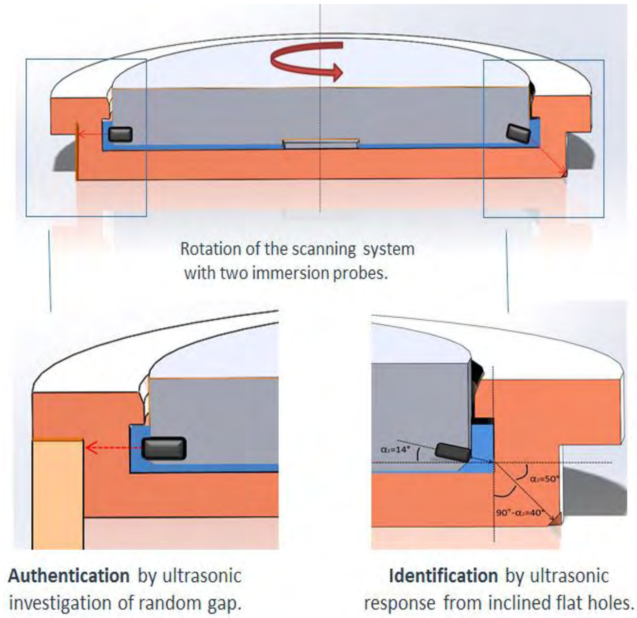

FIGURE 21. Illustration of the rotating scanning system with two immersion ultrasonic probes.

The probe revolution period (on the prototype scale 1/4) around the entire circumference is about 30 s. In the final system of identification and authentication design on a full-scale canister, the revolution period will be about 2 minute in order to keep the same resolution as was used for the prototype reading.

V. CONCLUSIONS

The internal gap ultrasonic investigation after friction stir welding of copper canisters has been experimentally demonstrated in the laboratory using a scanner with 10 MHz central frequency probes. The relative amplitude response due to the discontinuity introduced by the gap seems adequate for an authentication method. The characteristics of the internal gap have been investigated by the pulse-echo mode and the influence of temperature on ultrasonic investigations has been discussed.

Moreover, the ultrasonic data are compared with the "true profile" by using an optical inspection of a FSW section of a flange.

The integration of the identification and authentication methods is proposed with an innovative and simple scanning system designed to fit to the copper canister lid dimension and shape. Moreover, a patent is already filed covering the identification and authentication methods described in this paper [26]. In order to validate the concept of identification and authentication methods, future developments of this research will include the ultrasonic investigation of full-scale canisters. In particular, a reading system prototype will be designed for the acquisition of the ultrasonic echo of the internal gap, to be collected on a database. This collection will be used to fulfil a statistical analysis to verify the uniqueness of copper canisters fingerprints.

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REFERENCES

[1] M. Chiaramello, M. Sironi, F. Littmann, P. Schwalbach, and V. Kravtchenko, "JRC Candu sealing systems for Cernavoda (Romania) and upcoming developments," *ESARDA Bull.*, vol. 44, pp. 29–39, Jun. 2010. [Online]. Available: https://esarda.jrc.ec.europa.eu/images/Bulletin/Files/B_2010_044.pdf

[2] Y. Lahogue, F. Littmann, S. Synetos, J. Lupo, V. Piron, and M. Sironi, "Developments in the deployment of ultrasonic bolt seals at the storage ponds of a large reprocessing plant," in *Proc. Symp. Int. Safeguards, Linking Strategy, Implement. People*, Vienna, Austria, Oct. 2014, p. 120. [Online]. Available: https://www.iaea.org/safeguards/symposium/2014/home/eproceedings/sg2014-papers/000155.pdf.

[3] J. Goncalves *et al.*, "EU—ABACC cooperation: Strengthening safeguards capabilities," in *Proc. Conf. 37th ESARDA Symp. Safeguards Nucl. Non-Proliferation*, Manchester, U.K., May 2015, pp. 91–98.

[4] F. Littmann, M. Sironi, V. Kravtchenko, B. Wishard, P. Schwalbach, and L. Matloch, "New ultrasonic optical sealing bolts for dry storage containers," in *Proc. Conf. 37th ESARDA Symp. Safeguards Nucl. Non-Proliferation*, Manchester, U.K., May 2015, pp. 799–803.

[5] L. Hildingsson, C. Andersson, and R. Fagerholm, "Safeguards aspects regarding a geological repository in Sweden," Swedish Radiat. Safety Authority, Stockholm, Sweden, IAEA Safeguards Symposium, 2014, paper CN-220-166.

[6] L. Cederqvist, M. Johansson, N. Leskinen, and U. Ronneteg, "Design, production and initial state of the canister," SKB, Stockholm, Sweden, SKB Tech. Rep. TR-10-14, Dec. 2010.

[7] *Final Repository*, accessed on Oct. 20, 2016. [Online]. Available: http://www.stralsakerhetsmyndigheten.se/In-English/About-the-Swedish-Radiation-Safety-Authority1/Spent-nuclear-fuel-repository/Final-Repository/

[8] A. Hedin, J. Andersson, JA Streamflow AB, K. Skagius, and K. Konsult AB, "Long-term safety for KBS-3 repositories at Forsmark and Laxemar—A first evaluation. Main report of the SR-Can project," SKB, Stockholm, Sweden, SKB Rep. TR-06-09, 2006.

[9] J. C. Bennett, D. M. Day, and S. A. Mitchell, "Summary of the CSRI workshop on combinatorial algebraic topology (CAT): Software, applications and algorithms," Tech. Rep., 2009.

[10] L. O. Brown, S. K. Doorn, and P. B. Merkle, "SERS-active nanoparticles as a barcoding technology for tags and seals," in *Proc. INMM 51st Annu. Meeting*, Baltimore, MD, USA, 2010, p. 1.

[11] D. Chernikova, K. Axell, and A. Nordlund, "A new approach to environmentally safe unique identification of long-term stored copper canisters," in *Proc. Symp. Int. Safeguards*, Vienna, Austria, Oct. 2014, pp. 2–5, paper CN-220-104.

[12] D. Demyanuk, M. Kroening, A. Lider, D. Chumak, and D. Sednev, "Intrinsic fingerprints inspection for identification of dry fuel storage casks," *ESARDA Bull.*, vol. 50, pp. 79–86, Dec. 2013.

[13] D. Chernikova, K. Axell, A. Nordlund, and H. Wirdelius, "Novel passive and active tungsten-based identifiers for maintaining the continuity of knowledge of spent nuclear fuel copper canisters," *Ann. Nucl. Energy*, vol. 75, pp. 219–227, Jan. 2015.

[14] L. Capineri, "Seal fingerprint acquisition device," Dept. Inf. Eng., Univ. Florence, Florence, Italy, Final Rep., 2016.

[15] C. Clementi, M. Calzolai, L. Capineri, and F. Littmann, "Ultrasonic identification of copper canisters to be used for long term geological repository," in *Proc. IEEE Int. Ultrason. Symp.*, Tours, France, Sep. 2016, pp. 1–3.

[16] W. M. Thomas, K. I. Johnson, and C. S. Wiesner, "Friction stir welding—Recent developments in tool and process technologies," *Adv. Eng. Mater.*, vol. 5, no. 7, pp. 485–490, 2003.

[17] J. Krautkrämer and H. Krautkrämer, *Ultrasonic Testing of Materials*, 4th ed. Heidelberg, Germany: Springer-Verlag, 1990.

[18] IAEA, Vienna, Austria. (1988). *Ultrasonic Testing of Materials at Level 2*. [Online]. Available: http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/19/100/19100874.pdf

[19] *State of the Art of the Welding Method for Sealing Spent Nuclear Fuel Canister Made of Copper. Part 1—FSW*, Posiva Oy, Eurajoki, Finland, May 2014.

[20] Lecoeur Electronique, Chuelles, France. (Jun. 2004). *US.Box, User Manual*. [Online]. Available: http://www.lecoeur-electronique.net/

[21] M. Engholm and T. Olofsson, "Inspection of copper canister for spent nuclear fuel by means of ultrasound. Copper characterization, FSW monitoring with acoustic emission and ultrasonic imaging," Dept. Tech. Sci., Uppsala Univ., Uppsala, Sweden, Tech. Rep. TR-09-28, Aug. 2009.

[22] *Design of Fuel Handling and Storage Systems for Nuclear Power Plants, IAEA Safety Standards Series*, document NS-G-1.4, 2003.

[23] *Speed of Sound in Water*, accessed on Oct. 28, 2016. [Online]. Available: http://www.engineeringtoolbox.com/sound-speed-water-d_598.html

[24] R. P. Reed and R. P. Mikesell, "Low temperature mechanical properties of copper and selected copper alloys: A compilation from the literature," Inst. Mater. Res., Nat. Bureau Standards, Boulder, CO, USA, Monograph 101, 1959.

[25] F. Tesfaye and P. Taskinen, "Densities of molten and solid alloys of (Fe, Cu, Ni, Co)-S at elevated temperatures—Literature review and analysis," Dept. Mater. Sci. Eng., Aalto Univ., Espoo, Finland, Tech. Rep. TKK-MT-215, 2010.

[26] F. Littmann, "Ultrasonic identification and authentication of containers for hazardous materials," European Patent 16 166 465, Apr. 21, 2016.

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