Non-destructive techniques as a tool for radioactive waste package performance testing to ensure long-term safety

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Abstract
This paper assesses the applicability of the Non-Destructive Techniques based on electromagnetic, ultrasonic and potential corrosion to monitor the corrosion and internal defect of cementitious packages. The evaluation was undertaken on cementitious package with an optimal formulation water/binder (W/B) ratio of 0.5 and after curing at 20 °C for 28 days. The corrosion mapping results show that the first test considered as the reference specimen has maximum and minimum values of -69 mV and -97 mV, respectively. In tests 2 and 3, we observed a decrease in corrosion potentials of -321 mV and -375 mV after adding steel bars and HCL. Regarding the ultrasonic testing, the results show that the average value of the crack depth equal to 0.032m. The obtained results show that the three techniques used have a strong correlation and complementarity. This means that it will be a good approach if we exploit these NDT in order to enhance and strengthen the safety related to radioactive cementitious package.

1. Introduction
The application of nuclear techniques in several field, particularly in research laboratories, medical facilities, industry and nuclear power plant contribute in the generation of a significant amount of radioactive waste, this kind of waste must be treated in accordance with national legislative and regulatory frameworks as well as international standards in order to improve radioactive waste management safety. As a result of the different activities using radioactive materials, several types and classes of waste are generated, requiring different waste management process. The immobilization in a solid and stable form by using cementitious matrices are the most common method for conditioning radioactive waste after treatment [1].

On the one hand, given the extent of the radiological risk from a safety and security point of view presented during the process of radioactive waste management, storage, and disposal, and given that the cement package is the first barrier between the environment and radioactive material. On the other hand, the cementitious packages of radioactive waste are exposed to degradation and stress as a result of the physicochemical storage conditions. Researchers seek to achieve the highest level of radiological safety possible at each stage of the process. This is the context of our research, which is being conducted...
through the implementation of a new process for the inspection and quality control of radioactive waste containment packages [2]. To ensure a high level of safety during transportation, storage, and disposal, we should implement a quality control and inspection monitoring system in the radioactive waste management process. This will provide us with critical information about the integrity of cementitious packages without destroying them, while also preventing any possible contamination of workers and the environment, especially in the case of orphan sources, where the source does not dispose in any data sheet.

Recent work from our laboratories showed that the use of cement binders and geopolymer package can potentially produce a high performance solid and stable form [3-5]. However, the utilization of Non-Destructive Techniques is advanced in the field of civil engineering [6]. To increase the safety of radioactive waste, researchers are seeking to strength the utilization of NDT for some special operations [7-8]. To achieve this purpose, non-destructive testing techniques, specifically X-ray radiography and the electrode potential corrosion diagnostic technique, were used in this present work as a tools for detecting and characterizing defects and alterations in the cement packages. In addition to the aforementioned techniques, the Pundit (Portable Ultrasonic Non-destructive Digital Indicating Tester) was used to measure the depth of cracks [9].

The purpose of this study is to improve the quality of concrete specimens by conducting research on the various defects that may exist and/or develop over time in concrete, such as corrosion, cracking, vacuum, and intruders with densities other than concrete, such as wood, steel, polyster. Which will help to prevent radioactivity from being released into the environment [10].

1. Experimental methodology

1.1 Cement package preparation protocol

The confinement of radioactive waste requires the use of a concrete whose characteristics are well determined and which is subject to long-term storage. The study of the composition of a concrete consists in defining the optimal mixture of the various aggregates available, as well as the dosage of cement and water in order to produce a concrete whose qualities are those sought for the realization of the work in question. The formulation used in this study is the one adopting by the department of radioactive waste management of CNESTEN (table1) [11].

<table>
<thead>
<tr>
<th>Concrete</th>
<th>Cement</th>
<th>Sand</th>
<th>Gravel</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>1m³</td>
<td>300Kg</td>
<td>760 kg</td>
<td>1190 Kg</td>
<td>170 L</td>
</tr>
</tbody>
</table>

Concerning the procedure for preparation of the concrete, we put the sand and gravel in the mixer container first, then pour the cement mix everything manually by pouring water gradually. After 30 seconds of mixing, we add the remaining water regularly and let the whole mix agitate in the mixer for 30 seconds. After mixing we pour the mixture into the 25×25×5 cm and 5×10 cm molds for concrete test specimens. The mussels are kept in the standards at a temperature of 20°C which corresponds to the room temperature. For the test 2 and 3, we followed the same protocol mentioned above, although we changed the pH by adding HCL (pH=5). To have a good homogeneity and the total evacuation of the air bubbles which can be introduced during the filling of the specimens, a vibration operation was made.
1.2 Assessment methods

Corrosion diagnostic technique CANIN+ was carried out to monitor the level of corrosion of reinforcement in concrete by performing corrosion potential measurements. Table 1 summarizes the characteristics of this technique [12]. The interpretation of the results in terms of potential threshold is dependent on the following potential intervals, according to the referenced standard: ASTM C876-91 (Cu/CuSO4) [13]:

- $E > -0.2$ V : No corrosion with 90% probability
- $-0.2$ V > $E > -0.35$ V : Higher probability of uncertain corrosion (50%)
- $-0.35$ V > $E$ : Corrosion with 90% probability

Table 2. CANIN+ technical characteristics

<table>
<thead>
<tr>
<th>Measurement range:</th>
<th>$\pm 999$ mV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Resolution:</td>
<td>1 mV</td>
</tr>
<tr>
<td>Electrodes:</td>
<td>Rod Electrode (copper/coppersulphate)</td>
</tr>
<tr>
<td>Impedance:</td>
<td>10 MΩ</td>
</tr>
<tr>
<td>Temperature range:</td>
<td>From 0°C to 60 °C</td>
</tr>
</tbody>
</table>

Radiography was also used to detect and characterize internal defects. X-rays are electromagnetic radiation with enough energy to pass through objects without being altered. Unlike light, which is absorbed or reflected by solid objects, x-rays pass through bodies that are opaque to light. The attenuation of the X-ray beam as it passes through the object to be radiographed is not always uniform (Figure 1). This is due to the differences in attenuation between the different components of the object. These differences in attenuation are essential to the formation of the radiographic image: The radiographic image results from the difference in attenuation of X-rays in the media crossed [14].

Figure 1. Schematic of radiography set-up

X-rays are generated in a generator by the interaction of electrons with a metallic target (Figure 2). The electrons are emitted by a filament heated by the Joule effect. A potential difference accelerates these electrons and directs them to a metal target (anode or anticathode). The rapid deceleration of
electrons during their impact on the target causes the production of X-ray photons. An X-ray generator with a maximum power of 300 kV and a maximum current of 5 mA was used in the following work. The distance between the source and the film is approximately 700 mm, and the exposure time ranges from a few seconds to a few minutes. The detectors used are sensitivity films with dimensions adapted to the size of the objects studied.

**Figure 2.** Basic X-ray production process

Pundit technique was used to monitor crack depth measurement. The Pundit is an ultrasonic pulse velocity measuring instrument used to assess the quality of concrete and other materials such as rocks, wood, and ceramic. This technique has a wide range of applications, including path length measurement, uniformity analysis, surface velocity measurement, and crack depth measurement. The Pundit should be reset on a regular basis by using the calibration bar, especially if the transducer frequency or cables are changed. **Table 2** shows the Pundit technical specifications [15]. The calibration bar indicates the expected calibration value (μs). Couple the transducers to the calibration bar by pressing firmly on the transducers and both ends of the bar as shown in the **Figure 3**.

**Table 3.** Pundit technical specifications

<table>
<thead>
<tr>
<th>Tx / Rx Frequency (kHz)</th>
<th>24, 37, 54, 82, 150, 200, 220, 250, 500</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pulse width</td>
<td>1-100 μs</td>
</tr>
<tr>
<td>Excitation voltage (V)</td>
<td>125, 250, 350, 500, AUTO</td>
</tr>
<tr>
<td>Rx Gain</td>
<td>1x, 10x, 100x, AUTO</td>
</tr>
</tbody>
</table>

**Figure 5.** Resetting the Pundit
1.3 Summary table of tests performed

Table 4 summarizes the various analyses performed on prepared specimens with various defects, such as corrosion diagnostics, crack depth measurement, and radiography:

<table>
<thead>
<tr>
<th>Tests</th>
<th>Analyzes</th>
<th>Dimensions</th>
<th>Defect</th>
<th>Photo</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test: 1</td>
<td>Corrosion diagnostic</td>
<td>25 * 25 *5</td>
<td>without defect</td>
<td></td>
</tr>
<tr>
<td>Test: 2</td>
<td>Corrosion diagnostic</td>
<td>25 * 25 *5</td>
<td>2 clean steel bars</td>
<td></td>
</tr>
<tr>
<td>Test: 3</td>
<td>Corrosion diagnostic +Crack depth</td>
<td>25 * 25 *5</td>
<td>2 corroded steel bars</td>
<td></td>
</tr>
<tr>
<td>Test: 4</td>
<td>Radiography</td>
<td>D=5, H=10</td>
<td>only concrete without any defect</td>
<td></td>
</tr>
<tr>
<td>Test: 5</td>
<td>Radiography</td>
<td>D=5, H=10</td>
<td>Crack</td>
<td></td>
</tr>
<tr>
<td>Test: 6</td>
<td>Radiography</td>
<td>D=5, H=10</td>
<td>Plastic pipe</td>
<td></td>
</tr>
<tr>
<td>Test: 7</td>
<td>Radiography</td>
<td>D=5, H=10</td>
<td>Steel bar</td>
<td></td>
</tr>
<tr>
<td>Test: 8</td>
<td>Radiography</td>
<td>D=5, H=10</td>
<td>Steel bar on pipe of plastic</td>
<td></td>
</tr>
<tr>
<td>Test: 9</td>
<td>Radiography</td>
<td>D=5, H=10</td>
<td>Wood</td>
<td></td>
</tr>
</tbody>
</table>

2. Results and discussions

2.1 Test: 1 concrete only

A concrete reference specimen is prepared without introducing any defect. The measurement results obtained by the corrosion diagnostic technique are as follows: The measured values are well above -200 mV, which explains the absence of corrosion in our reference test. The maximum value measured is -69 mV and the minimum value is-97 mV. Therefore, the potential results collected in this case will be considered as reference values for others test.
2.2 test: 2- two uncorroded steel bars

A concrete specimen that contains two clean steel reinforcing bars. The measurement results obtained by the corrosion diagnostic technique are as follows:

![Figure 4. relative frequency of corrosion potentials](image)

From the measurement results obtained, it can be seen that there is a remarkable increase in the potential values compared to the measurement results of the previous test, especially in the area where the concrete reinforcements exist, despite the rebars are visually clean. 84% of the collected potentials say that the corrosion is uncertain according to the standard "ASTM C876-91", the remarkable increase of potentials can be explained by depassivation of the bars.

2.3 test: 3 - concrete and 2 corroded bars

A concrete specimen, which contains two very corroded steel reinforcement bars.

a. Visual examination

The figure below shows a crack in the concrete reinforcement:

![Figure 5. Specimen n°3 which presents a crack](image)
b. Measurement of the depth of the crack

Below are the six measurements obtained by the ultrasonic testing when measuring the depth of the crack after 125 days preparing concrete. In order to minimize the relative error, the transducer distance is 7.5 cm, in which the relative error (% error) of concrete crack depth measurements is between 1.54% to 20% and an average% error of 9.74% [16].

<table>
<thead>
<tr>
<th>Measures</th>
<th>Crack depth</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.034 m</td>
<td>23.2 °C</td>
</tr>
<tr>
<td>2</td>
<td>0.031 m</td>
<td>23.5 °C</td>
</tr>
<tr>
<td>3</td>
<td>0.034 m</td>
<td>23.5 °C</td>
</tr>
<tr>
<td>4</td>
<td>0.030 m</td>
<td>23.5 °C</td>
</tr>
<tr>
<td>5</td>
<td>0.032 m</td>
<td>23.7 °C</td>
</tr>
<tr>
<td>6</td>
<td>0.035 m</td>
<td>23.2 °C</td>
</tr>
</tbody>
</table>

It can be seen that all six crack depth measurements are in the same order. Once cracks are found in the radioactive waste packages, radiological controls must be carried out to ensure that radioactive materials are not dispersed into the environment, and containment is recommended.

c. Corrosion diagnostic mapping results

The measurement results of corrosion diagnosis technique are as follows:

According to those results:
- The potentials collected in specimen 3 are lower than those in specimen 2;
- The potential values are lower than -350 mV in the area containing the steel bars, which explains the existence of corrosion according to the "ASTM C876-91" standard. Once considerable levels of corrosion are detected, the radioactive waste packaging packages must be systematically checked from a radiological point of view, in order to prevent any contamination dispersion.

2.4 Radiography of samples 4 to 9

![Image](7-a Wood)

![Image](7-b Crack)

![Image](7-c without defect)

Figure 7. Image obtained by the radiography of samples n° 4, 5 and 9

![Image](8-a Plastic pipe (a))

![Image](8-b Steel bar(b))

![Image](8-C Steel bar in plastic pipe(c))

Figure 8. Image obtained by the radiography of samples n° 6, 7 and 8

According to Figure 7-c, it can be clearly seen that the contrast of the image is uniform, which means that the test 4 is without defect and does not contain any inclusions. Therefore, we considered it as a reference specimen.
For the test 5 on the picture, we can clearly see the presence of an area with a very high contrast, which confirms the presence of a transverse crack on the image 7-b. Concerning tests 6 and 9, we notice respectively in the center of images 8-a and 7-a a remarkable blackening, which explains the presence of less dense objects (plastic pipe and wood). However, for test 7 we notice the presence of a white area in the middle of the picture, this can be interpreted by the existence of a more attenuating body (steel bar). For test 7, the iron density is high compared to that of the concrete, we can see that the concrete reinforcement is clearly apparent in image 8-b. Based on image 8-c related to test 8, we can clearly see the presence of a whitening surrounded by a black contrast, which confirms our experimental approach.

3. Conclusion

The present work is part of the general framework of the studies concerning the reinforcement of the safety relative to the treatment of the radioactive or nuclear waste confined in the concrete packages. This study concluded that these three techniques used represent an important tool. For a complete diagnosis of radioactive waste packages, the implementation of these three techniques is essential:

The result obtained for the second test by our technique CANIN+ showed that 84% of relative frequency of corrosion potentials, which means that the corrosion is uncertain, however the third test represent 68% of uncertain corrosion and 32% is corroded with 90% probability.

The corrosion in the third test induced a visual crack that is confirmed by radiography testing, using ultrasonic testing, the average crack depth value equal to 0.032 m.

This study would be a preliminary basis. It will lead to the development of new procedures and guidelines to better control the management and monitoring of radioactive waste by optimizing the level of safety in question like as providing critical information about the integrity of cementitious packages and prevent any possible contamination of workers and environment.

Acknowledgement

The technical inputs of Mr BOUIH Abderrahim the head of Radioactive Waste Management Department at CNESTEN.

Disclosure statement: Conflict of Interest: The authors declare that there are no conflicts of interest. Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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