

Cases of alkali-aggregate reactions in radioactive waste management in Belgium

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Abstract

Cement-based materials are common materials used for the immobilization of radioactive waste produced by the nuclear activities. The aim of this contribution is to give an overview of how pathologies such as alkali-silica reactions are taken into account in the management of radioactive waste in Belgium through a recent example of non-conform waste packages.

In 2013, a gel formation was observed on several inspected cemented radioactive waste packages, leading to extended inspections and an extensive research program. The waste streams affected by the phenomenon are cemented radioactive effluents with high sodium hydroxide content. Following the evidence of gel formation, preliminary analyses concluded to an Alkali Silica Reactions (ASR) diagnosis and a research program is on-going to develop acceptable long-term scenarios for the disposal of the affected waste packages. A new dedicated temporary storage facility is also under construction to allow a dedicated follow-up of the gel formation.

The results of the research program and the inspection and follow-up program tend to show that the unusually liquid ASR product exudation has stopped or is very slow. Further results are still needed to develop a safe final disposal solution.

Additionally, acceptability and disposability criteria for the future radioactive waste included standardized tests in order to guarantee the absence of AAR in the contaminated concrete structures and the cementitious materials used for the waste immobilization.

Keywords: *alkali-silica reaction; radioactive waste management; cementation of radioactive waste; expansion; ASTM test methods*

1. INTRODUCTION

The radioactive waste management system in Belgium complies with the general rules and guidelines of the IAEA [1, 2, 3] and involves different stakeholders. The producers are the owners of the radioactive waste and are responsible for the operations that led to the radioactive waste production, while the waste management organization, ONDRAF/NIRAS, ensures that the radioactive waste is safely managed from its production to its final disposal.

In Belgium, most of the radioactive waste results from the operations of both nuclear powerplants of Doel and Tihange. The raw radioactive waste has to be conditioned or immobilized using a conditioning matrix or an immobilization matrix in order to confine the radionuclides within a conditioned waste package.

The radiological, physical and chemical properties of the raw radioactive waste have to be characterized before designing a specific conditioning process, possibly including a pre-treatment step. Three waste categories are used in Belgium based on the activity of the waste and its half-life:

- Category A for the short-lived (SL) low-level waste (LLW) and intermediate level waste (ILW),
- Category B for the long-lived (LL) low-level waste (LLW) and intermediate level waste (ILW),
- Category C for the high-level waste (HLW), which represent less than 5% of the total volume of radioactive waste for more than 90% of the total radioactivity.

The industrial and technological strategy behind the conditioning process of one specific radioactive waste stream takes into account the properties of the raw radioactive waste, the adequacy of a conditioning matrix with the waste properties and the requirements for the safe temporary storage and final disposal of the radioactive waste. At each step of the waste management system, there are specific criteria that the waste has to comply with.

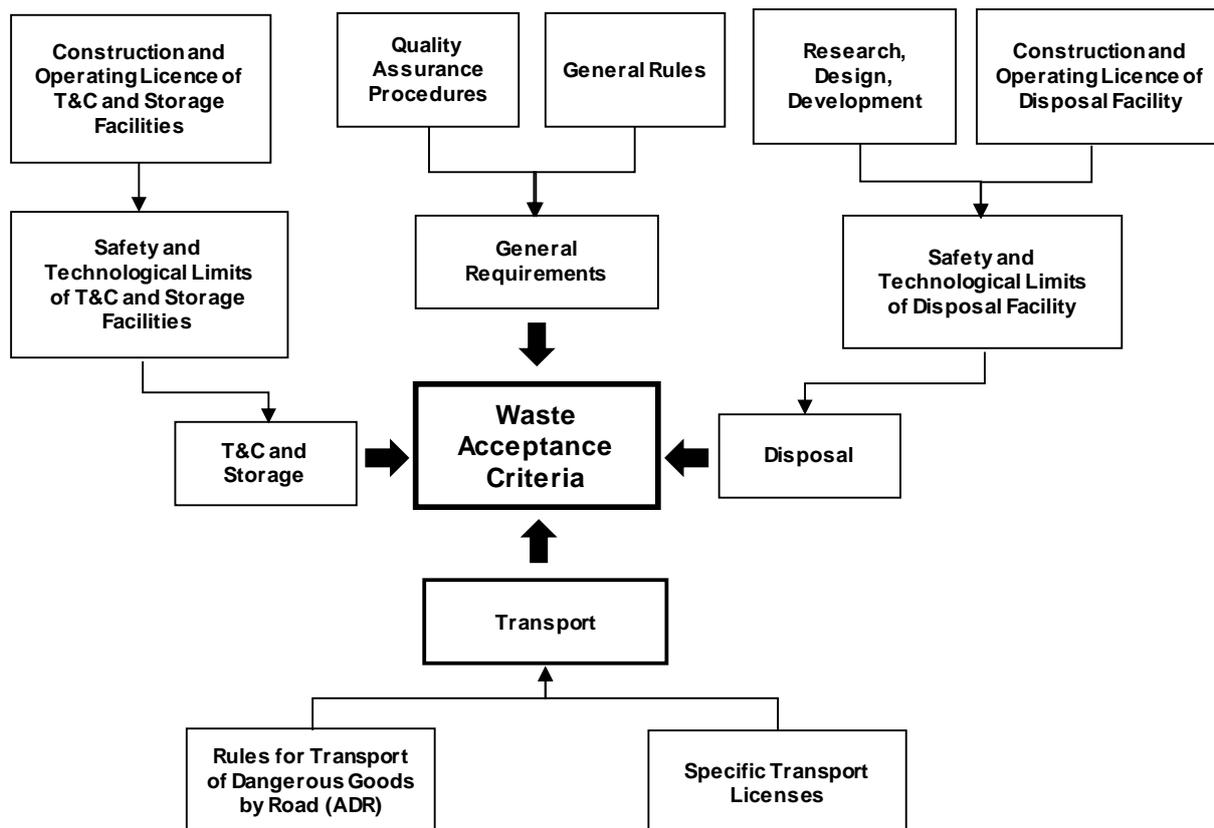


Figure 1.1: The radioactive waste acceptance system in Belgium

For most of the categories A and B waste packages, the used and expected conditioning processes are based on a cementitious immobilization matrix. Solid and bulk radioactive waste is usually encapsulated in a heterogeneous cemented waste package while a liquid or granulated radioactive waste is homogeneously mixed and immobilized with the cementitious matrix. A cement-based conditioning process has to ensure the compatibility of the cementitious immobilization matrix with the waste stream, but also ensure that the industrial process is sufficiently robust to safely produce stable and compliant waste packages.

The long-term behaviour of a sound and compliant conditioned waste package ensures the confinement of the radioactivity and thus an acceptable radiological impact on the environment and the population. However, as waste management organization, ONDRAF/NIRAS faces a number of non-compliant waste packages and has to properly identify the non-conformity, determine its root cause, evaluate its possible consequences on the temporary storage and the final disposal, and design a suitable long-term solution to render the waste package compliant again. Alkali-aggregates reactions are one specific case of a non-conformity that can potentially threaten the safe storage and disposal of the waste packages. The following sections describe one case of non-conformity due to ASR from its initial diagnosis to its consequences on the Belgian waste management system.

2. A SPECIAL CASE OF ALKALI-SILICA REACTIONS: THE “GEL DRUMS”

2.1 Context

After the radioactive waste is conditioned, the waste packages remain temporarily stored in dedicated facilities until their final disposal. In 2013, during routine inspections of conditioned low-level waste packages, a yellow gelatinous substance was found on the lid of several drums. This anomaly triggered an extended inspection of other waste packages produced with similar conditioning lines. More than 8000 drums suspected of showing signs of gel formation were identified. Part of the drums has shown

evidence of a large amount of gel formation. More importantly, the spillage of radioactive material out of the drums and the leakage of radionuclides are concerning safety issues.



Figure 2.1: Evidence of ASR gel formation on cemented radioactive waste packages

2.2 Scope and diagnosis of the anomaly

2.2.1 The radioactive waste and the conditioning process leading to the anomaly

A more thorough inspection of the drums was carried out to determine the scope of the anomaly. This led to the conclusion that the affected drums originated from campaigns ranging from 1980's to 2010's. The identified conditioning process was used for the cementation of low level radioactive effluents resulting from the operations at the Doel nuclear powerplant, more commonly called "concentrates".

The raw radioactive waste has initially a high boric acid concentration and is basified using lye (NaOH) up to a pH around 10-12. The waste is kept at high temperatures around 70-90°C to prevent the crystallization of boric phases. The cementation process introduces then the ingredients in the following order: gravel, sand, lime (Ca(OH)₂), radioactive waste and CEM I cement. A limited amount of water was added in some cementation campaigns to improve the workability of the material. Table 1 gives an overview of the chemical composition and the properties of the radioactive waste (after its basification) and the immobilization matrix recipes that were used between 1980 and 2010.

The mixture is then poured in a 400L standard metallic drum and closed with a non-air-tight lid. The inner surfaces of the drum and lid are also coated with a zinc layer to prevent premature corrosion. Once their conformity is established, the produced waste packages are transported to a storage facility at BelgoProcess. Routine inspections are carried out during the temporary storage phase, during which if an anomaly is observed, a follow-up inspection is carried out leading potentially to a non-conformity.

Table 1.1: Variability of the chemical composition of the cemented concentrates and the used concrete recipes from 1980 to 2010 (only the main species are listed in the table)

Concentration in the radioactive waste	Average value	Standard deviation	Max value	Min value	Median value
Na [mg/kg]	84207.89	25431.91	127915.00	6368.00	84915.00
B [mg/kg]	22248.48	11346.02	50000.00	1910.00	20933.00
Cl [mg/kg]	30333.37	26108.56	121846.00	4729.00	19614.00
(PO ₄) ³⁻ [mg/kg]	9164.76	9892.86	42140.00	7.00	6829.00
(SO ₄) ²⁻ [mg/kg]	28197.19	27126.70	138959.00	5000.00	19067.50
Variables	Average value	Standard deviation	Max value	Min value	Median value
pH	10.97	0.70	12.84	9.70	10.88
Density [g/L]	1299.89	54.47	1376.00	1201.00	1301.00
Ingredients	Average value	Standard deviation	Max value	Min value	Median value
Cement [kg/drum]	300.82	63.60	430.00	172.00	300.00
Gravel [kg/drum]	276.64	44.29	375.00	180.00	270.00
Sand [kg/drum]	102.64	50.88	192.00	0.00	103.00
Concentrate [kg/drum]	170.73	25.21	200.00	116.00	185.00
Lime [kg/drum]	11.86	11.71	30.00	0.00	6.00
Water [kg/drum]	2.14	6.38	25.00	0.00	0.00
W/C	0.41	0.05	0.52	0.30	0.41

2.2.2 Diagnosis of the phenomenon

After the first evidence of gel formation, with a limited number of drums overflowing with gel, a thorough action plan was devised to better understand the root cause of the phenomenon:

- *Analysis of gel samples:* the amount of extruded gel varied from one drum to another with no apparent correlation to the cemented waste packages characteristics or the campaign year. During the following months in 2013, samples of the radioactive gel were collected and analyzed by BelgoProcess (the industrial daughter company of ONDRAF/NIRAS). The results showed high alkali and silicon contents. Other species such as chlorides, sulfates, borates, aluminium and calcium were also found in smaller amounts. Low radioactivity levels were also measured (ranging from 40 to 400 Bq/g of Cs-137). The chemical composition of five different samples was determined using ICP-OES.
- *Characterization of the affected concrete material:* core samples were taken from affected waste packages and the material was then analysed using SEM and microscopy techniques (Figure 3). The interface between the siliceous aggregates and the cement paste clearly indicate signs of alkali-silica reactions.
- *Replicability of the phenomena on non-radioactive samples at lab-scale:* concrete samples were cast at lab-scale using a batching solution similar to the cemented radioactive waste. The concrete samples clearly indicated the presence of alkali-silica reactions and cracking within the concrete samples (Figure 3).

Lastly, in order to have an independent scientific peer review, an expert panel was gathered and the necessary data was made available to them. Based on the preliminary results and evidence, it was stated that the phenomena were most likely the result of alkali-silica reactions occurring between the highly alkaline waste and the siliceous aggregates that were used in the cementation process.

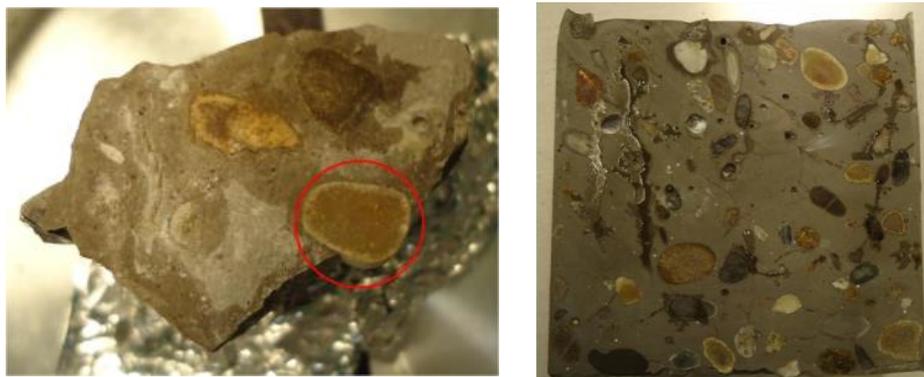


Figure 2.2: Fracture and section from a radioactive core sample from an affected waste package (left) and a non-radioactive concrete sample representative of the waste (right)

3. THE MANAGEMENT STRATEGY FOR THE “GEL DRUMS”

3.1 Short term management of the potentially affected waste packages

3.1.1 Extensive research program

Following the expert panel review, a 4-year research program was devised in order to define an appropriate long-term safe disposal solution for the potentially affected waste packages. The research program started in 2017 and involved different research partners and institutes. The four main objectives of the research program are the following:

- Understanding the mechanisms of the gel formation and exudation: based on extensive analyses and characterization of radioactive gel samples, similar non-radioactive ASR products will be synthesized and produced on representative concrete materials. How the chemical composition, the structure of the amorphous silica and the rheological properties of the ASR products relate to each other will be further investigated.
- Developing monitoring techniques for the observed phenomena: using a combination of non-destructive techniques (acoustic emission, micro-wave analysis and thermography) the observed phenomena will be replicated at lab-scale on non-radioactive samples and controlled environment conditions. The objective is to study the feasibility of such approach before potentially applying a similar method on a full-scale waste package.
- Assessing the consequences on the disposal systems: the consequences on the disposal systems of the waste packages depend on how fast and to what extent the alkali-silica reactions would form such a liquid ASR product and would result in expansion and cracking. The water uptake, water loss and swelling pressures of the ASR products will be investigated using radioactive samples and representative synthesized gels.
- Designing full reprocessing and rehabilitation solutions for the drums and for the removed gel: a side research topic is the investigation of the possible reprocessing of the affected waste packages using thermal techniques (such as a plasma oven). This scenario is still considered viable in case the safe disposal of the affected waste packages in their current state cannot be demonstrated.

3.1.2 Temporary storage and follow-up program of the affected packages

In addition to the on-going research program, the unpredictable gel formation kinetics and behaviour required an extended inspection and follow-up program of the potentially affected waste packages. The following actions were carried out and/or are still going-on:

- More than 8000 waste packages were identified and externally inspected. The priority was indeed to determine whether more packages showed signs of gel overflow and possible contamination of neighbouring packages. Opening of the lids requires mechanical devices and possible exposition of the operators and is limited to a few packages per campaign. There has been no specific expansion or deformation of the waste packages that could point out the occurrence of an expansive reaction.

- 16 waste packages have had their lids replaced by a Plexiglas lid to easily follow up the gel formation on the surface of the immobilization matrix (see Figure 4). No visible evolution of the gel spots on the surface of the waste packages was observed between 2015 and 2019. There are no visible cracks on the surface neither.

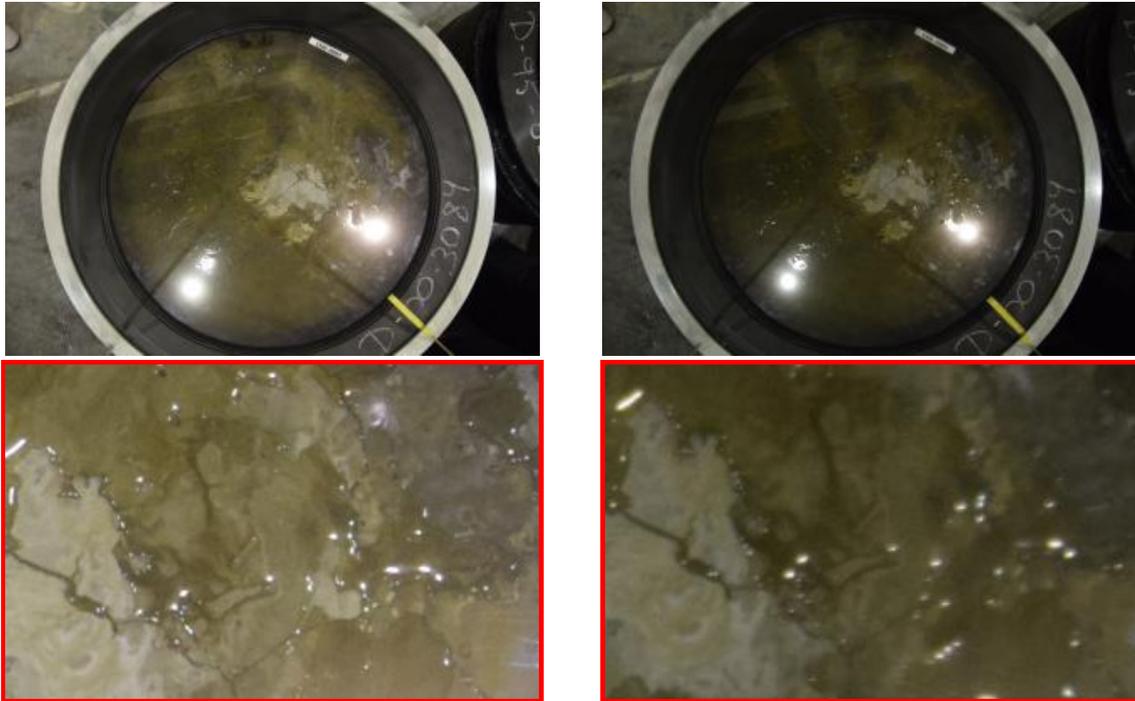


Figure 3.1: Follow-up of the gel formation on the surface of a waste package, picture of the surface in December 2015 (left), picture of the same surface in November 2019 (right). The bottom pictures show zoom-in on the same gel spot on the surface in 2015 and in 2019.

- 11 waste packages were tilted and placed horizontally to evaluate the gel formation, exudation and overflow. However, the mass of gel coming out of the packages is very limited (in average over the 4 years around 0.3g per day and mostly nothing in 2019)

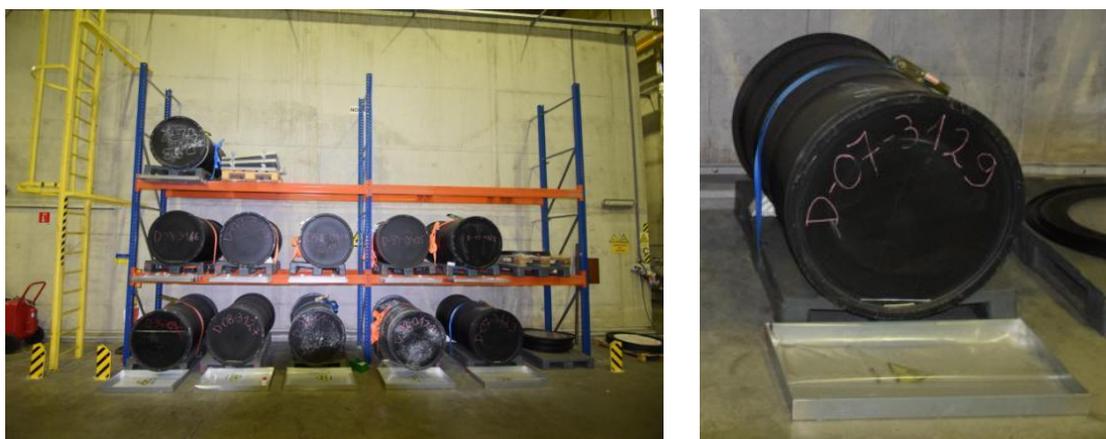


Figure 3.2: Affected waste packages placed horizontally for the gel formation and exudation follow-up

Due to the uncertainties remaining on how the waste packages will behave and how they will be disposed of, a new temporary storage building is foreseen and will be solely dedicated to the around 8000 potentially affected packages. Their temporary storage will allow more time for research, inspection and follow-up until a safe disposal solution will be agreed on.

3.2 Long term management of the potentially affected waste packages

3.2.1 Disposability criteria and surface repository

As shown in Figure 1, the final destination of the conditioned radioactive waste package is a disposal repository. In Belgium, the category A waste is expected to be disposed of in a near-surface repository in Dessel. A first draft of the safety case was submitted by ONDRAF/NIRAS to the Belgian regulatory body AFCN-FANC. It describes the expected behaviour of the radioactive waste on the long term over more than a thousand years and shows that it can be safely disposed of provided that a number of criteria are complied with [4].

Among these criteria, two are of importance when considering the above described case of the “gel drums”: the presence of a free liquid and the risk of expansive reactions.

3.2.2 Formation of liquid ASR products

The exudation of a sticky radioactive gel phase from the cementitious materials could be regarded as a non-conformity. The gel formation had two different implications:

- The presence of a free liquid phase promotes the release of radionuclides and cannot be allowed for the short and long term management;
- The liquid gel formation was on-going and the unpredictable phenomenon compromised the stability of the conditioned waste.

Based on a review of the scientific literature on ASR products in field and in lab materials and using the results provided so far by the research program, the unusually liquid ASR products in the case of the “gel drums” were compared to more conventional ASR products (see Table 2) . The surrogate samples referred to in Table 2 are non-radioactive samples synthesized and studied in the framework of the research program. Some of the XRD and NMR results are available in the submitted paper by Nicolas Courtois for the present 16th ICAAR conference [22].

Table 3.1: Comparison of the properties of the liquid ASR products formed in the cemented concentrates of Doel and the classically encountered ASR products in civil engineering structures

Properties of the ASR products	Classic ASR products encountered in civil engineering materials	Liquid ASR products formed in cemented concentrates of KCD	
		Radioactive samples	Surrogate samples
XRD patterns [11, 14, 17]	<i>Mostly amorphous. For high Ca/Si ASR products, similarities with tobermorite and shyklovites</i>	<i>Amorphous, no crystalline phases detected</i>	<i>Amorphous, no crystalline phases detected. Small peak of Na-trona on one of the synth. gel samples.</i>
Chemical Composition [7, 8, 9, 10, 11, 12]	<i>Ca/Si = 0.05-0.5 Alkali/Si = 0.1-1.3 Water content = variable</i>	<i>Ca/Si ~ 10⁻⁴ Alkali/Si ~ 1.0-1.1 Water content ~ 35%</i>	<i>Ca/Si ~ 10⁻⁴ Alkali/Si ~ 0.9-1.1 Water content ~ 40%</i>
Structure of the silica [14, 15, 16, 17, 18, 19]	<i>Mostly Q₃ and Q₄ Si tetrahedra</i>	<i>Mostly Q₁ and Q₂ tetrahedra, no Q₄</i>	<i>Mostly Q₁ and Q₂ tetrahedra, very limited Q₄ sites.</i>
Morphology of the phase [20, 21]	<i>Two morphologies are reported: one rosette-like and one granular morph.</i>	<i>Not observed</i>	<i>Liquid and smooth morphology, dried honey-comb pattern upon drying</i>
Rheological Mechanical properties [8, 10, 13]	<i>Solid aspect, measurable non zero yield stress, elastic moduli > 5 GPa</i>	<i>Not measured</i>	<i>Viscous liquid aspect, no yield stress, elastic moduli < 500 MPa</i>

The specific liquid ASR products that were observed at the surface of the cemented concentrates of Doel had distinctive structure, chemical composition and rheological behaviour. These were the result of the large amounts of alkali and hydroxyl ions provided by the waste. In average, the KCD waste packages produced between 1990 and 2011 contained 40 kg of sodium per m³ of concrete. In the case of classic cementitious materials for which only conventional ingredients (cement, water, appropriate aggregates for concrete and admixtures) were used, if the material contains less than 5 kg of Na₂O_{eq} per m³ of concrete, only classic and solid ASR products are expected to form.

Consequently, two conclusions can be drawn regarding the formation of a free liquid phase due to alkali-silica reactions:

1. Such a free liquid phase cannot be formed in regular ASR-affected materials and structure when the available alkali and hydroxyl ions are at a normal to high level of concentrations. The formation of a liquid ASR product is restricted to the specific systems similar to the “gel drums” with unusually high hydroxyl and alkali content and siliceous aggregates;
2. As shown by Figure 4 and in Figure 5, it seems that the amount of visible formed gel on the surface remained the same over the last five years. Further results from on-going experiments will need to confirm that the gel formation is stable and will not lead to a significant new gel overflow episode.

The disposability of the “gel drums” with regard to the criterium of “ absence of free liquid phases” requires further guaranties that the gel formation has indeed stopped and would not start again when the packages will be placed in the surface repository.

3.2.3 Risk of expansive ASR reactions

Following the ASR diagnosis and based on the state of the art, expansion of the cementitious materials is expected. Such expansive reactions threatens the integrity of the engineered barrier systems (EBS) in the disposal repositories.

Due to the nature of the liquid ASR products and because no visible expansion or cracking were observed so far on the inspected affected packages, it still has to be confirmed whether a possible expansive ASR would happen on the long-term and would damage the EBS. In the case of the near-surface repository for the low level radioactive waste packages, four 400L drums are expected to be placed inside a reinforced concrete caisson. A backfilling mortar will then be poured inside the caisson, before placing a concrete lid on top. The current safety model only guaranties a safe disposal if the integrity of the caissons is ensured.



Figure 3.3: Design of the concrete caisson for the disposal of the category A waste packages

There are two on-going research topics currently under investigation to determine the extent of the possible damage due to expansion of the waste packages:

- Specific home-made cells were designed to measure the ASR swelling pressures of confined reactive concrete samples. An alkaline solution is continuously flushed in the cell.
- A numerical model will determine the maximal admissible swelling pressure exerted by the reactive waste to induce a trough cracking pattern in the caisson, under different hypotheses regarding the system, its properties and its geometry.

The disposability of the “gel drums” regarding the criterium related to the “absence of expansive reactions” depends on whether it can be clearly shown that the caissons will sustain the ASR swelling pressures within the affected waste packages.

4. IMPACT ON THE ON-GOING AND FUTURE CONDITIONING PROCESSES OF RADIOACTIVE WASTE

The occurrence of the gel formation anomaly in 2013 triggered a number of actions to prevent any future cases of similar anomalies. First, all of the currently used conditioning processes were screened:

- If no siliceous aggregates are used in the immobilization matrix, the risk of ASR was ruled out.
- If siliceous aggregates were indeed used, the alkali content of the immobilization matrix was estimated and the ASR risk evaluated. If the $\text{Na}_2\text{O}_{\text{equ}}$ provided by the cement is lower than 2w% (and if the radioactive waste does not contribute to the alkali content in the waste package), the ASR risk may be neglected.
- If alkali and siliceous aggregates are present in the immobilization matrix, the ASR risk cannot be neglected, the conditioning process has to be stopped and a characterization test is required to further investigate the reactivity of the aggregates.

Additionally to the screening of the operating conditioning lines, criteria specific to ASR were requested for all future conditioning processes:

- In the case of heterogeneously cemented waste, all aggregates used in the immobilization matrix have to be tested using the ASTM C 1260 test method [5]:
 - If the relative expansion is lower than 0.1%, the ASR risk is ruled out.
 - If the relative expansion is higher than 0.2%, the aggregate is reactive and cannot be used for the immobilization of the radioactive waste;
 - If the relative expansion is higher than 0.1% but lower than 0.2%, a second test based on the ASTM C 1293 is required [6]. If the relative expansion obtained with this second test is higher than 0.04%, the ASR risk cannot be ruled out, the aggregate is reactive and can't be used for the immobilization of the radioactive waste.
- In the case of homogeneously cemented waste, a durability test is required from the producers. Surrogate samples representative of the expected immobilized waste are made and followed-up at controlled temperature and relative humidity conditions for 18 months. The producers have to show that the weight and dimensions of the samples are stable over time. Such test can conservatively rule out the occurrence of ASR in case of siliceous aggregates and alkalis provided by the waste.

5. CONCLUSIONS

The occurrence in 2013 of alkali-silica reactions in cemented radioactive waste packages led to the formation of an unusually liquid ASR product potentially compromising the safe management of the waste packages.

Following an extensive inspection program and preliminary analyses of the formed gel, the scope of the anomaly was eventually limited to cemented radioactive effluents that contained a high concentration of NaOH and used reactive siliceous aggregates. The ASR diagnosis was confirmed and a research program was built up to better understand the phenomena and its consequences on the storage and the disposal of the potentially affected waste packages. Meanwhile, a dedicated temporary storage building is under construction and a follow-up program of the anomaly on the potentially affected packages is on-going.

On the long term, the potentially affected waste packages will need to address two disposability criteria: the absence of free liquid phases and the absence of expansive reactions that could threaten the integrity of the engineered barriers system. In 2021, a final integrative review of the results provided by the research program will help building up a suitable long-term solution for the disposal of the affected waste packages. Four different scenarios are still considered by ONDRAF/NIRAS:

1. The risk of ASR can be completely ruled out of the waste package,

2. The risk of ASR cannot be ruled out however its evolution during the temporary storage in controlled conditions shows that the waste package is stable and will be stable,
3. The risk of ASR cannot be ruled out however the consequences of ASR on the disposal systems can be bound and limited, with an adaption of the disposal system design if necessary,
4. The risk of ASR cannot be ruled out, its consequences might affect the safe disposal and a full reprocessing is necessary.

Which one of these scenarios would be suitable for which waste packages will be evaluated based on the results of the on-going research program. A second research program may be necessary to evaluate the implementation of the actual solutions on the scale of the waste packages.

Finally, the waste acceptance system has included specific ASR criteria based on the ASTM C 1260 and 1293 test methods to prevent future similar anomalies,

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