Disposing of Waste Package with Respect to Physico-Chemical Characteristics – 15041

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ABSTRACT

The waste inventory to be taken into account for the design of the Cigeo project [1] is defined in the industrial program for waste management established in cooperation between nuclear operators (EDF, AREVA, CEA) and Andra. This waste inventory has been constituted with a list of different families of radioactive waste packages from High Level Waste (HLW) to Intermediate Level Long-lived Waste (IL-LLW).

IL-LLW include a large variety of items such as structural elements from fuel assemblies (cladding from the fuel rods called "hulls", end pieces called "end caps" and assembly spacer grids, etc.), sludge from effluent treatment or miscellaneous equipment (filters, pumps, etc.). Most ILLW (except solidified sludges) are basically metallic but organic and inorganic components such as plastics (cellulose, PVC…) and salts may also be included.

The rules used for designing Cigeo are based on the separation of waste having different physico-chemical characteristics in different cells. However it could be possible to dispose of waste packages with different physico-chemical characteristics in the same cell if we can demonstrate that a type of waste does not generate significant physico-chemical perturbations on other co-stored families.

The first step of our analysis has been the identification of categories of materials present in waste likely to interact with each other. The second step consisted in analysing qualitatively the physico-chemical interactions between these materials. In the third step, we defined criteria allowing evaluation of the consequences of these interactions on (i) gas production (mainly H_2), (ii) radionuclide release and transfer and (iii) degradation of concrete components of the disposal cell. We built then a table allowing qualitative comparison of the consequences of the interactions between the materials types defined in the first step.

The conclusion of this study shows that it is necessary to pay a very careful attention before disposing of waste packages containing organic components nearby waste packages containing actinides radionuclides. The geochemical phenomena coupling must also be quantitatively evaluated so as to analyse possible interaction processes between the released organic compounds and the cementitious components of the cells, the near-field claystone and the radionuclides migration.

INTRODUCTION

The Industrial Program of Waste Management (PIGD) [2] describes the waste inventory. It schedules the delivery of the different categories of specially Intermediate Level Long-lived Waste (IL-LLW) and defines associated flows. In this context, the issue of co-disposal of different IL-LLW waste families in the same cell invariably arises when it comes to optimize the architecture of the disposal. Several criteria, qualitative or quantitative, can be used to analyse the feasibility of co-storage: criteria related to their physico-chemical behaviour, the size of their disposal package and the delivery flows.

The analysis developed here focuses exclusively on the physico-chemical compatibility of waste considering physico-chemical interactions between two adjacent waste packages and effects of temperature conditions with respect to (i) production of non-radioactive gas, (ii) release and transfer of radionuclides and (iii) degradation of the concrete materials of the pieces constituting the disposal package (matrix, container ...). We consider that a disposal package contains packages of waste belonging to only one IL-LLW family.

Based on the knowledge of the materials variety of waste, the analysis identifies the physico-chemical incompatibilities between these materials. Subsequently, in connection with the composition of the different waste families and the disposal safety, the approach allows to specify the criteria relevance to distinguish and define different categories of waste families potentially co-storable from a physico-chemical compatibility point of view.

METHODOLOGY

The general methodology applied in this analysis can be divided in three successive steps. The first step of our analysis has been the identification of categories of materials present in waste likely to interact with each other. The second step consistes in analysing qualitatively the physic-chemical interactions between these materials. In the third step, we define criteria allowing evaluation of the consequences of these interactions on (i) gas production (mainly H_2), (ii) radionuclide release and transfer and (iii) degradation of concrete components of the disposal cell. We build then a table allowing qualitative comparison of the consequences of the interactions between the materials types defined in the first step.

Definition of Waste Materials

IL-LLW include a large variety of items such as structural elements from fuel assemblies (cladding from the fuel rods called "hulls", end pieces called "end caps" and assembly spacer grids, etc.), sludge from effluent treatment or miscellaneous equipment (filters, pumps, etc.). Most IL-LLW (except solidified sludges) are basically metallic but organic and inorganic components such as plastics (cellulose, PVC…) and salts may also be included.

A classification of the different materials identified in the ILLW leads to define four main types: organic components, cement, metallic components and salts. In the specific case of the organic materials, we need to refine our analysis by sorting them into five sub-types depending on their complexing power: chloride polymers, cellulose, fluoride polymers ion exchange resin and miscellaneous polymers:

- Organic materials:
    - Chlorinated polymers (PVC ...) have been identified as (i) potential sources of organic complexing agents (especially because of the high amounts of phthalates in PVC) and (ii) producers of corrosive gas that can damage metallic or cement casing;
    - Cellulose was specifically identified as iso-saccharinic acid (ISA) (product of cellulose degradation) has a strong complexing strength;
    - Fluoropolymers have been specifically identified as a result of the production of corrosive gas (HF) which may degrade metallic or cement casing;
    - The ion exchange resins (IER) have been identified as sulphates and ammonium suppliers from degradation by radiolysis / hydrolysis allowing to degrade cementitious materials;
    - Waste characterized by different sub-categories of "Miscellaneous polymers" are all likely to produce gas (H_2, CO, CO_2, CH_4) and release potentially complexing organic acids. It should also be kept in mind that a large number of organic species produced by radiolysis / hydrolysis of different polymers has not been identified and we cannot guarantee that these species will not have a strong complexing strength (di-acids, aromatic ...). The analyses carried out so far have been limited to the identification of the following complexing species: formic, acetic, oxalic, adipic, glutaric, phthalic and iso-saccharinic (for example, this represents approximately 20% of the total organic carbon produced by the degradation of PVC Plastunion).
- Cementitious embedding materials:
  This type includes cement or “cement-bitumen” embedding matrices. The long-term behaviour of “cement-bitumen” matrices is not currently known. In the absence of data, it is considered that the matrix combines both the effects of the presence of hydraulic binders and organic materials.

- Metallic materials:
  Metallic Materials are divided into subtypes based on the corrosion rate and gas production in distinguishing low reactive materials (zirconium alloys, stainless steels, nickel alloys, non-alloy steel) from reactive materials (alloy magnesium, sodium and aluminium alloy).
  Among the reactive metals, three of them have been specifically considered:
  - Aluminium alloys which corrosion could produce hydrogen at very low relative humidity values;
  - Magnesium alloys having very high corrosion rates in the presence of chlorides;
  - Sodium due to the risk of explosion and fire.

  The choice of types has also led to differentiate activated materials and non-activated materials in connection for the former with the significant radiological inventory and non-labile nature of the radionuclides release associated with this inventory. It should be noted that the metallic casing of some waste packages are also classified in the non-activated and with low reactivity metallic type.
- Salts:
  This type considers all kind of salts of the ILLW, based on the inventory of sludge reprocessing. The presence of these salts may alter the hydraulics and transfer (chemical osmosis related to salinity gradient, high ionic strength ...) and redox conditions (presence of nitrates ...) in the ILLW cells and in their near field. The presence of ammonium sulphate can lead to the degradation of cementitious materials and the clay-stone in the near field.

Table 1 summarizes the different types of materials selected to conduct the analysis of interaction and physicochemical compatibility between ILLW families. These types are used to identify the main interactions processes such as gas production, species capable of interacting with radionuclides and / or materials ... The choice of these different categories and sub-categories of materials is based on the expertise of Andra. Thermal conditions intervene as a transverse factor influencing the degradation kinetics of materials and the behaviour of radionuclides.

TABLE 1 : Types of materials considered in this analysis

	Types of materials		**Types of interactions**
Organics	Chlorinated polymers (PVC ...)		• Release of phtalates (complexing organic species) • HCl production
	Cellulose		• Release of iso-saccharinic acid (ISA) (complexing organic species)
	Fluoropolymers		• HF production • Release of complexing organic species
	Ion Exchange Resins (IER)		• Release of ammonium et sulphate • Release of complexing organic species
	Miscellaneous polymers		• Release of complexing organic species
Cementitious materials	Cement		• Alkaline conditions
	Cement-Bitumen		• Gas production (H_2) by radiolysis • Release of complexing organic species
Metallic materials	Metallic materials with low reactivity	Activated waste: zirconium alloys, stainless steels and nickel alloys	• Release of radionuclides by corrosion of the alloy • Hydrogen production in anoxic conditions (slow rate)
		Casing or non-activated waste: stainless alloys, non-alloy steel	• Hydrogen production in anoxic conditions (slow rate)
	Metallic materials with high reactivity	Al alloys	• Hydrogen production with high rate: o in oxic or anoxic conditions o for very low relative humidity values
		Mg alloys	• H_2 production with high rate in oxic or anoxic conditions • Release of radionuclides by corrosion of the alloy
		Na	• H_2 production with high rate • Risk of explosion / fire

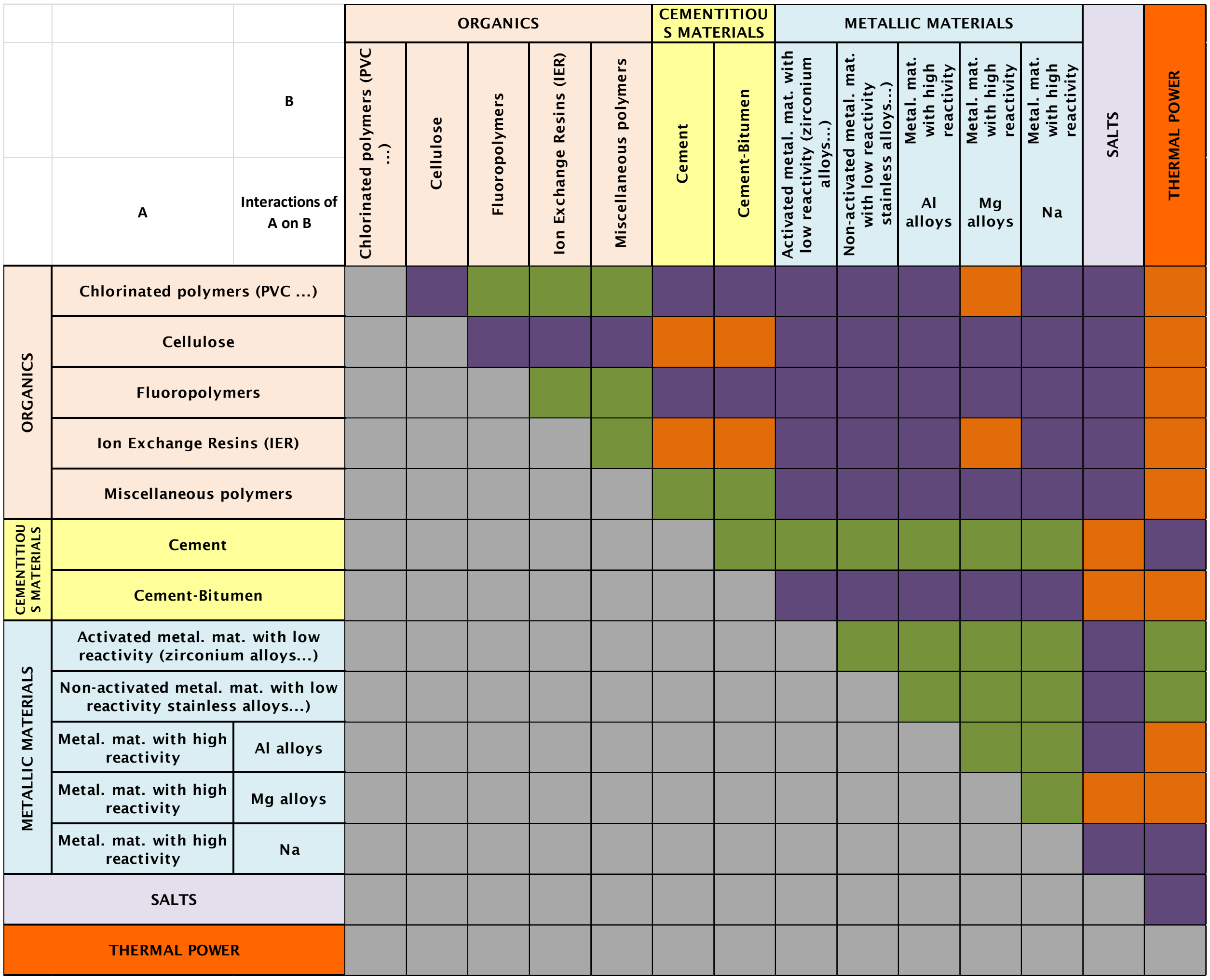
Salts	• Release of large amounts of sulphates, nitrates, carbonates, ammonium chloride ...

Matrix of Interactions

After defining the main types of materials present in the ILLW, we developed a matrix of physic-chemical interactions between these different types of materials. This matrix identifies the specific physico-chemical processes (i) between the various degradation products released by the various types of materials of the ILLW and (ii) resulting from thermal effects produced by some ILLW.
We can notice that mechanical interactions have not been considered because they do not fall within the co-storage question. It is the same for interactions of radiation, assuming the role played as attenuator screen by the storage package. It should also be noted the co-disposal of bitumen waste or the one of vitrified IL-LLW is not addressed, because they are considered as not being co-stored with other types of IL-LLW.

This matrix of interactions (Figure 1) indicates whether or not interactions between the different materials identified in the waste occur and, if there, if these interactions are weak, strong, or if there are difficult to characterize. Thus, the colour code employed means:

- Green: no or weak interactions;
- Orange: strong interactions;
- Purple: uncertainties as to the intensity of interactions.



		B	ORGANICS					CEMENTITIOUS MATERIALS		METALLIC MATERIALS					SALTS	THERMAL POWER
	A	Interactions of A on B	Chlorinated polymers (PVC ...)	Cellulose	Fluoropolymers	Ion Exchange Resins (IER)	Miscellaneous polymers	Cement	Cement-Bitumen	Activated metal. mat. with low reactivity (zirconium alloys...)	Non-activated metal. mat. with low reactivity stainless alloys...)	Metal. mat. with high reactivity Al alloys	Metal. mat. with high reactivity Mg alloys	Metal. mat. with high reactivity Na		
ORGANICS	Chlorinated polymers (PVC ...)			🟪	🟩	🟩	🟩	🟪	🟪	🟪	🟪	🟪	🟧	🟪	🟪	🟧
	Cellulose				🟪	🟪	🟪	🟧	🟧	🟪	🟪	🟪	🟪	🟪	🟪	🟧
	Fluoropolymers					🟩	🟩	🟪	🟪	🟪	🟪	🟪	🟪	🟪	🟪	🟧
	Ion Exchange Resins (IER)						🟩	🟧	🟧	🟪	🟪	🟪	🟧	🟪	🟪	🟧
	Miscellaneous polymers							🟩	🟩	🟪	🟪	🟪	🟪	🟪	🟪	🟧
CEMENTITIOUS MATERIALS	Cement								🟩	🟩	🟩	🟩	🟩	🟩	🟧	🟪
	Cement-Bitumen									🟪	🟪	🟪	🟪	🟪	🟧	🟧
METALLIC MATERIALS	Activated metal. mat. with low reactivity (zirconium alloys...)										🟩	🟩	🟩	🟩	🟪	🟩
	Non-activated metal. mat. with low reactivity stainless alloys...)											🟩	🟩	🟩	🟪	🟩
	Metal. mat. with high reactivity	Al alloys											🟩	🟩	🟪	🟧
	Metal. mat. with high reactivity	Mg alloys												🟩	🟧	🟧
	Metal. mat. with high reactivity	Na													🟪	🟪
SALTS																🟪
THERMAL POWER																

Fig. 1: Matrix of interactions (green: no or weak interactions, orange: strong interactions and purple: uncertainties as to the intensity of the interactions)

Matrix of Consequences

Subsequently, a second matrix is developed to show the analysis of the consequences of potential interactions identified with respect to:

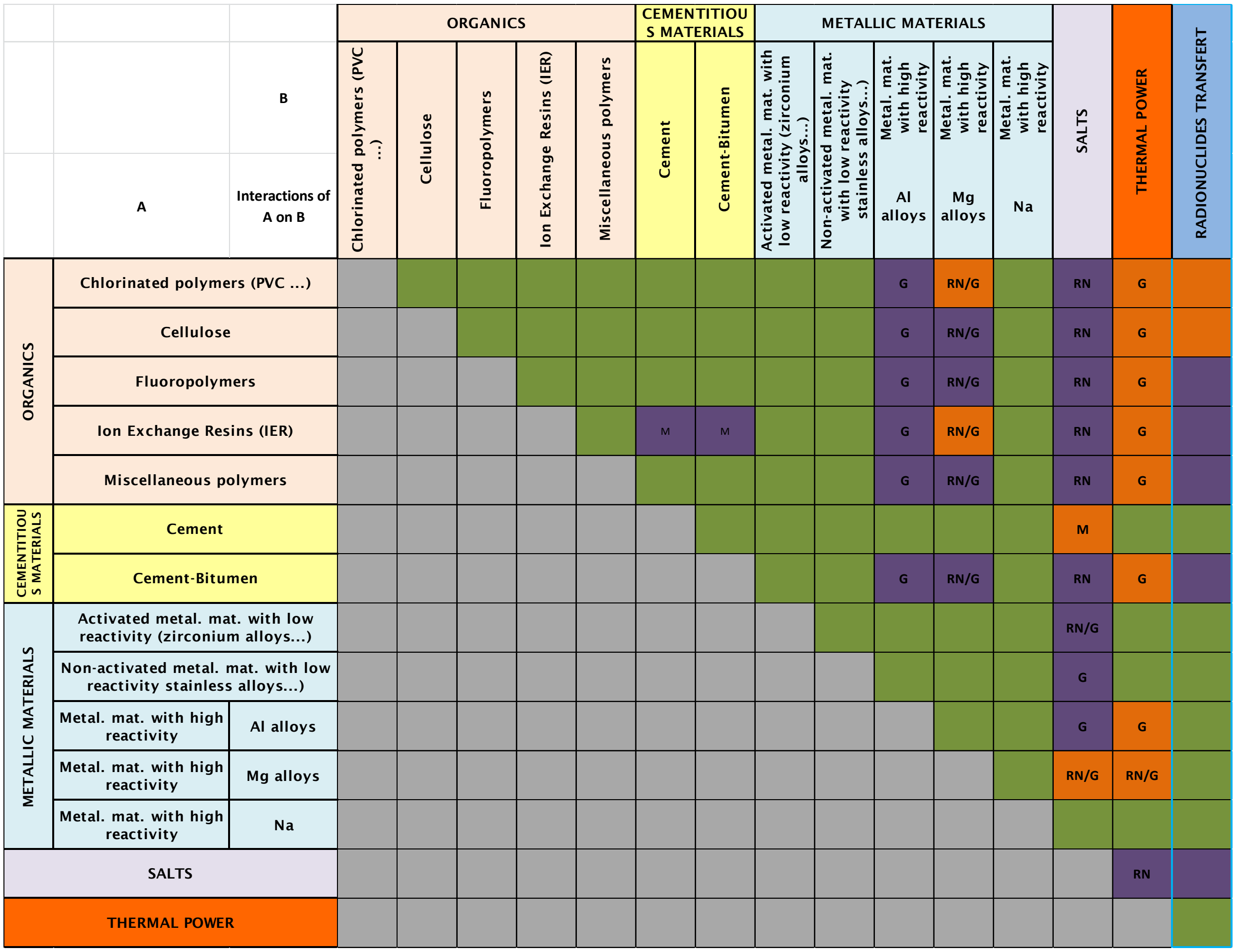
i) production of non-radioactive gas (mainly hydrogen, HCl). The importance of the gas production is controlled by the reaction rate of metallic materials corrosion (mainly in anoxic conditions) and by the rate of radiolysis of the organic compounds. Gas production by radiolysis of the water content in cementitious materials, estimated as being low in disposal conditions, is not considered here. The possible consequences of this production mainly concern the period of operation / reversibility of the ILLW disposal;

ii) release and transfer of radionuclides. The qualitative analysis of the consequences aims to identify the conditions likely to increase the release rate and mobility out of the storage package or can point out uncontrolled uncertainties to date. Particular attention is paid to the effects of organic complexing agents. The possible consequences of this production concern mainly the period of post-closure;

iii) degradation of the materials of the disposal package (matrix storage container ...). It focuses on the degradation of concrete by the degradation compounds released by the ILLW (acids, sulphated species ...)

This matrix indicates whether or not there is an impact of the contact between the different types of materials identified in the preceding paragraph and, if there, if these effects are weak, strong, or if it is difficult to characterize. A dual code has been used, on the one hand the same color code as that used for the matrix of interactions and, on the other hand, letters identifying what are these consequences: G for gas production, RN for the transfer of radionuclides and M for the degradation of cementitious component. As for the matrix of interactions, the following color code was used (Figure 2):

- Green: little or no consequences;
- Orange: significant consequences;
- Purple: uncertainties as to the intensity of the consequences.



			ORGANICS					CEMENTITIOUS MATERIALS		METALLIC MATERIALS					SALTS	THERMAL POWER	RADIONUCLIDES TRANSFERT
		B	Chlorinated polymers (PVC ...)	Cellulose	Fluoropolymers	Ion Exchange Resins (IER)	Miscellaneous polymers	Cement	Cement-Bitumen	Activated metal. mat. with low reactivity (zirconium alloys...)	Non-activated metal. mat. with low reactivity stainless alloys...)	Metal. mat. with high reactivity Al alloys	Metal. mat. with high reactivity Mg alloys	Metal. mat. with high reactivity Na			
	A	Interactions of A on B															
ORGANICS	Chlorinated polymers (PVC ...)			🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟪 G	🟧 RN/G	🟩	🟪 RN	🟧 G	🟧
	Cellulose				🟩	🟩	🟩	🟩	🟩	🟩	🟩	🟪 G	🟪 RN/G	🟩	🟪 RN	🟧 G	🟧
	Fluoropolymers					🟩	🟩	🟩	🟩	🟩	🟩	🟪 G	🟪 RN/G	🟩	🟪 RN	🟧 G	🟪
	Ion Exchange Resins (IER)						🟩	🟪 M	🟪 M	🟩	🟩	🟪 G	🟧 RN/G	🟩	🟪 RN	🟧 G	🟪
	Miscellaneous polymers							🟩	🟩	🟩	🟩	🟪 G	🟪 RN/G	🟩	🟪 RN	🟧 G	🟪
CEMENTITIOUS MATERIALS	Cement								🟩	🟩	🟩	🟩	🟩	🟩	🟧 M	🟩	🟩
	Cement-Bitumen									🟩	🟩	🟪 G	🟪 RN/G	🟩	🟪 RN	🟧 G	🟪
METALLIC MATERIALS	Activated metal. mat. with low reactivity (zirconium alloys...)										🟩	🟩	🟩	🟩	🟪 RN/G	🟩	🟩
	Non-activated metal. mat. with low reactivity stainless alloys...)											🟩	🟩	🟩	🟪 G	🟩	🟩
	Metal. mat. with high reactivity	Al alloys											🟩	🟩	🟪 G	🟧 G	🟩
	Metal. mat. with high reactivity	Mg alloys												🟩	🟧 RN/G	🟧 RN/G	🟩
	Metal. mat. with high reactivity	Na													🟩	🟩	🟩
SALTS																🟪 RN	🟪
THERMAL POWER																	🟩

Fig; 2: Matrix of consequences. The consequences include the production of gas (G), the transfer of radionuclides (RN) and the degradation of component materials (M). The last column analyses the consequences with respect to the transfer of radionuclides in the storage cell and the near field ((green: no or weak consequences, orange: strong consequences and purple: uncertainties as to the intensity of the consequences)

It should be noted that there may be interactions inherently "strong" between different types of materials without any consequences with respect to the three criteria at the scale of the cell (e.g. high calcium complexation of cementitious materials by degradation products of cellulose without having a significant effect on the degradation of cementitious material of the container).

QUALITATIVE ANALYSIS OF PHYSICO-CHEMICAL INTERACTIONS AND THEIR CONSEQUENCES

Consequences Due to the Types of Materials in the Waste Packages

The qualitative analysis of physico-chemical interactions between materials constituting the ILLW, as described in Table 1, and of their consequences result in identifying interactions which consequences (i) are severe or (ii) present a high level of uncertainty.

Major consequences are (without considering the thermal effects, see below):

- Disposal packages containing phthalates, additives abundant in chlorinated polymers, or cellulose: strong interactions with any other type of waste with respect to the transfer of radionuclides (complexation in solution by organic compounds);
- Disposal packages containing Mg alloys: only strong interaction with (i) the Cl-bearing organic waste with respect to gas production and release of radionuclides (effects of chlorides on corrosion rate) and (ii) IER with respect to the same indicators (effects of the sulphates on corrosion rate);
- Disposal packages containing salts: high reactivity with Mg alloys (effects of chlorides and sulphates on corrosion rate);
- Disposal packages with cement matrices: high deterioration by salts or by IER (high uncertainty knowledge in this case).

Consequences with uncertainties (without considering the thermal effects, see below):

- Disposal packages containing Al alloys: reactivity with organic materials with respect to the production of gas (effects of organic compounds on corrosion rate);
- Disposal packages containing Mg alloys: reactivity with organic compounds except chlorinated and IER and with respect to (i) production of gas and (ii) release of radionuclides (effects of organic compounds on corrosion rate);
- Disposal packages containing fluorinated polymers, IER, miscellaneous polymers (excluding chlorinated polymers and cellulose) or salts: reactivity with any other type of waste with respect to the radionuclides transfer (effects of complexing organic species or salts on the radionuclides transfer);
- Disposal packages containing cement-bitumen embedding matrices: uncertainties about the gas release in the presence of Al alloy and Mg alloy as well as about the radionuclides transfer in the presence of Mg alloy (effects of organic compounds on corrosion rate) or of salts (organics compounds and cumulative effects of salts);
- Disposal packages containing cement or cement-bitumen embedding matrices: uncertainties concerning the degradation of the embedding matrices by IER (high uncertainty of knowledge);
- Disposal packages containing salts: reactivity with all types of organic waste with respect to the radionuclides transfer (combined organic compounds and salts effects) and with metallic materials either with respect to the production of gas (all types of metal) or with respect to the radionuclides release (activated materials with low reactivity) (salt effects on the corrosion rate).

It should be noted that the question of metallic sodium is not addressed here since its high reactivity is independent of the presence of materials in adjacent storage package.

The consequences on the interactions of materials with respect to radionuclides transfer are summarized in Table 2. In Tables 2 to 4, the effects considered as "high" are indicated in orange and those for which there are uncertainties are shown in purple.

The analysis of the consequences with respect to the transfer of radionuclides in the cell and near field claystone is detailed in Table 2. This analysis was performed separately because the consequences of co-storage concern all other types of materials. For example, the co-storage of waste “rich” in cellulose causes a change in the transfer of radionuclides from all other type of waste (chlorinated polymers (PVC), fluorinated polymers, IER, miscellaneous polymers, embedded waste, activated metal with low reactivity, non-activated metal with low reactivity, Al, Mg, Na and salts).

The results described in Table 2 lead to avoid co-storing families of wastes containing organic materials with families of waste containing no organic, as well as families of waste containing salts with families which do not contain salts.

TABLE 2 : Comments on the analysis of the matrix of consequences with respect to the transfer of radionuclides in the storage cell and its near field

Types of materials	Consequences with respect to the transfer of radionuclides
Chlorinated polymers (PVC ...)	Changing the transfer of radionuclides by complexing organic species
Cellulose	Changing the transfer of radionuclides by complexing organic species
Fluoropolymers	Changing the transfer of radionuclides by complexing organic species
Ion Exchange Resins (IER)	Changing the transfer of radionuclides by complexing organic species Changing the transfer of radionuclides by possible degradation of cementitious materials due to the degradation of IER (ammonium ...)
Miscellaneous polymers	Changing the transfer of radionuclides by complexing organic species
Cement-Bitumen embedding matrix	Changing the transfer of radionuclides by complexing organic species
Salts	Changing the transfer of radionuclides by salts Changing the transfer of radionuclides by possible degradation of cementitious materials (sulphate…)

Table 3 shows all the consequences of the interactions of materials listed in the first column on those listed in the second column with respect to the three criteria gas production (G), release of radionuclides (RN) and degradation of component materials of the disposal package (matrix storage container (M)).

TABLE 3 : Consequences of interactions of materials with respect to gas production (G), degradation of components (M) and radionuclides release (RN) (excluding the effects of temperature)

Types of materials		Consequences	
Chlorinated polymers (PVC ...)	Mg	Significant increase of corrosion rate of magnesium alloys due to the chlorides, uncertainty about the effects of organic species on the corrosion rate of magnesium alloys	RN, G
IER	Mg	Significant increase of corrosion rate of magnesium alloys by sulphates. Uncertainty about the effects of organic species on the corrosion rate of magnesium alloys	RN, G
Salts	Mg	Significant increase of corrosion rate of magnesium alloys (chlorides and sulphates to a lesser extent)	RN, G
Salts	Cement	Significant degradation of the cementitious embedding matrix by salts (sulfates and ettringite formation ...)	M
Chlorinated polymers (PVC ...), cellulose, fluoropolymers, IER, miscellaneous polymers, cement-bitumen embedding matrix	Al	Uncertainties about the effects of organic species on the corrosion rate of aluminium alloys	G
Cellulose, miscellaneous polymers, cement-bitumen embedding matrix	Mg	Uncertainties about the effects of organic species on the corrosion rate of magnesium alloys	RN, G
Fluoropolymers	Mg	Uncertainties about the effects of fluoride on the corrosion rate of magnesium alloys	RN, G
Chlorinated polymers (PVC ...), cellulose, fluoropolymers, IER,	Salts	Uncertainties cumulative effects of organic species and salts	RN

miscellaneous polymers, cement-bitumen embedding matrix			
IER	Cement, cement-bitumen embedding matrix	Possible degradation of cementitious embedding matrices by IER	M
Salts	Activated metallic materials with low reactivity	Uncertainties of salts effects on the corrosion rate of activated metals with low reactivity	RN, G
Salts	Non-activated metallic materials with low reactivity, Al	Uncertainties of salts effects on the corrosion rate of non-activated metals with low reactivity and of aluminum alloys	G

(i): In general, the radiological inventory of aluminum alloys is low, degradation does not lead to a significant change in the release of the radiological inventory of the waste package.

Consequences Due to Thermal Conditions

The consequences of an increase in temperature due to the co-storage of ILLW package having different thermal powers concern mainly the gas production by radiolysis or corrosion. These effects are important in the case of:

- Organic materials (waste or as cement-bitumen embedding matrix) due to the increase in radiolysis rate;
- Al alloys and Mg alloys because of the increase of their corrosion rate.

A significant increase in temperature has also a significant impact with respect to the release of radionuclides in the case of Mg alloys (higher corrosion rates).
Table 4 gives the effects of temperature on materials/matrix in the first column with respect to (i) the production of gas (G) and (ii) the release of radionuclides (RN).

TABLE 4 : Consequences of temperature with respect to gas production (G) and release of radionuclides (RN)

Types of materials	Consequences	
Chlorinated polymers (PVC ...), cellulose, fluoropolymers, IER, miscellaneous polymers, cement-bitumen embedding matrix	Increase of the rate of radiolysis	G
Al	Increase of corrosion rate	G
Mg	Increase of corrosion rate	RN, G
Sels	Uncertainty of cumulative effects of temperature and salts	RN

CONCLUSION

The analysis conducted here by Andra helps to highlight the complexity of physico-chemical interactions likely to occur in a IL-LLW storage. For optimizing storage concepts it is necessary to pay a very careful attention before disposing of waste packages containing organic components closed to waste packages containing actinides radionuclides. The geochemical phenomena coupling must also be quantitatively evaluated so as to analyse possible interaction processes between released organic compounds and cementitious components of the cells, the near-field claystone and the radionuclides migration.

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