Decontamination of Nuclear Liquid Wastes Status of CEA and AREVA R&D: Application to Fukushima Waste Waters - 12312

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ABSTRACT

Liquid wastes decontamination processes are mainly based on two techniques: Bulk processes and the so called Cartridges processes.
The first technique has been developed for the French nuclear fuel reprocessing industry since the 60's in Marcoule and La Hague. It is a proven and mature technology which has been successfully and quickly implemented by AREVA at Fukushima site for the processing of contaminated waters.
The second technique, involving cartridges processes, offers new opportunities for the use of innovative adsorbents.
The AREVA process developed for Fukushima and some results obtained on site will be presented as well as laboratory scale results obtained in CEA laboratories.
Examples of new adsorbents development for liquid wastes decontamination are also given.

INTRODUCTION

Decontamination of nuclear liquid wastes has two main objectives:

1. Processing of liquid wastes during nuclear plant resulting from production operations. In this case the volumes to be processed are known and liquid wastes processing facilities are integrated in the global production scheme of the plant. The need for improvements of the liquid wastes technology have at least two reasons:

- To meet more and more severe regulation regarding the release of liquid wastes in the environment
- To recycle decontaminated liquid in the process in order to minimize liquid volumes consumption of the plant

2. Processing of liquid wastes as a result of a temporary situation like:

- Decontamination or dismantling project generating secondary liquid wastes.
- Incidental or accidental situation like the Fukushima accident.

CEA and AREVA have working for many years on both aspects but Fukushima has been a major challenge in terms of liquid wastes volumes to be processed as well as the short time to set up the process before the start of rainy season in June.

The French strategy regarding the processing of Fukushima contaminated liquid wastes was first to set up quickly a reliable and safe process based on the French experience and secondly, to check alternative adsorbents in order to offer alternative solutions to achieve a better efficiency of the process. It has been also an opportunity to assess new solutions to prevent future liquid wastes contamination.
In the first part, main technologies for the decontamination of liquid wastes studied in France will be introduced. In the second part, the co-precipitation process that has been implemented at Fukushima site will be presented as well as associated R & D studies. In the third part, two examples of prospective studies on new adsorbents will be presented.

TECHNIQUES FOR THE DECONTAMINATION OF LIQUID WASTES

We will focus on chemical techniques addressing solubilized liquid contamination. Electrochemical techniques or solid/liquid separation techniques like filtration or evaporation will not be considered in the following.

Chemical liquid wastes decontamination processes are mainly based on two techniques:

- **Bulk processes**: in this case chemical reaction is achieved in the bulk by addition of chemical reactants. The precipitates produced by the reaction collect the radioelements present in the solution. The precipitates are removed from the solution via a solid-liquid separation operation. This process is called co-precipitation process.
- **Cartridges (or Column) processes**: in this case solid particles bearing complexing agents or ionic exchange sites are inserted in a cartridge and a flux of the liquid to be decontaminated is forced through the cartridge.

The technology based on cartridges is very common and generally consists in ionic exchange resins. Unfortunately, resins are organic products which are difficult to condition and store. Organics are decomposed during storage time thus producing radiolysis gases like hydrogen or enhancing the contamination release to the environment through formation of soluble complexes resulting from polymer decomposition. Therefore, the development of non-organic adsorbents is highly desirable.

The CEA/AREVA R & D addressing liquid decontamination follows three priority axes presented below:

1. The modelling of bulk processes in order to optimize the size and design of precipitation reactors to achieve a better efficiency as well as a minimization of sludge volume
2. Study of non-organic solid adsorbents for cartridge type applications
3. The coupling of solid adsorbents techniques with membrane processes

We will focus on the first two of the above mentioned axes.

BULK PROCESS

Experience for the decontamination of liquid wastes at Marcoule and La Hague plants

Co-precipitation is operated in the French nuclear fuel reprocessing industry since the 60’s in Marcoule and La Hague. It is a proven and mature technology. La Hague facility has been originally designed for a maximal production of 100 000 m^3 of liquid wastes per year. However, by constantly optimizing the reprocessing steps, only a few percent of the initial capacity is used today.

This process consists of a cascade of stirred reactors. In each reactor, chemical additives are added to the liquid mixture. These additives have different actions regarding their chemical nature. Three types of decontamination phenomena take place in the bulk:

- Pure co-precipitation phenomenon: soluble contamination is trapped or encapsulated in a solid. The solid is formed in-situ by generally contacting two chemical species. For example, Barium nitrate is added in combination with sulphuric acid to form barium sulfate. This addresses strontium contamination.
- Adsorption: in this case colloidal particles are introduced in the reactor. The particles are produced outside the reactor. The phenomenon involved in the decontamination can be a pure adsorption one or based on an ion exchange reaction. The most striking example is the use of ferrocyanates precipitates that specifically remove Cs.
- A combination of both of the above mentioned phenomena.

One of the main feature of the process is a large concentration difference between the soluble contamination concentration which lays generally around a few 10^{-9} $mol.L^{-1}$ and the chemical species concentration that are added to remove them, which lays around a few 0.1 $mol.L^{-1}$. This high difference is necessary to reach a high efficiency of the process.

As the contaminated solids resulting from the co-precipitation process must be conditioned and stored as a solid waste, one of the main challenges of the R & D is to reduce their amount by optimizing co-precipitation operations. In the past years CEA R & D has focused on the modelling of co-precipitation phenomenon as well as on the optimization of reactor design.

Co-precipitation model

The absence of a model for the elementary decontamination process had immobilized the modeling of the global process until recently. Pacary et al. [1-3] bring a totally new theoretical approach and resolve the problem of modeling of the principal elementary process used to remove the radioactive Sr^{2+}.

The system involving a carrier component ($BaSO_4$) in the presence of a trace element (strontium ions) has been modeled. The theory considers two aspects: first, the modeling of the precipitation process of the carrier component in a continuous stirred tank reactor (CSTR) which is representative of La Hague type reactor, and in a second step, the modeling of the trace element co-precipitation. The co-precipitation model is based on the concept that a crystal is made of a succession of carrier component layers. Before the formation of the succeeding layer, the crystal surface enables to adsorb trace component. When the surface layer is covered up by the succeeding layer, trace component adsorbed is “sequestrated”.

Eq. 1 presents the equation for the CSTR. This expression gives the value of the decontamination factor, DF (ratio of initial activity -or concentration- to residual activity-or concentration- in the bulk) and is obtained from two material balances: a first, at the surrounding layer of a particle of fixed size and a second the CSTR scale. The first balance gives an expression of the trace component concentration in the new solid layer at the particle surface as a function of the mass transfer coefficient. The overall Eq. 1 is finally obtained using the material balance on the CSTR.

$$DF = \frac{T_E}{T} = 1 + \frac{6k\phi_V r_N G^3 \tau^4}{1 + \frac{3\phi_V kG}{k_d \phi_S}} \qquad \text{Eq.1}$$

With:

G linear crystal growth rate, $m.s^{-1}$
k adsorption parameter, dimensionless
k_d mass transfer coefficient, $m.s^{-1}$
r_N nucleation rate, nuclei.$m^{-3}.s^{-1}$
T activity or concentration of trace element in the bulk solution, $mol.m^{-3}$
T_E inlet activity or concentration of trace element in the bulk solution, $mol.m^{-3}$
$\square_S$ surface shape factor, dimensionless
$\square_V$ volumic shape factor, dimensionless
$\square$ mean residence time, s

Eq. 1 provides a new method to estimate the impact of precipitation conditions on decontamination efficiency.
The model has been compared to experimental values. The experimental device is presented in the figure below:

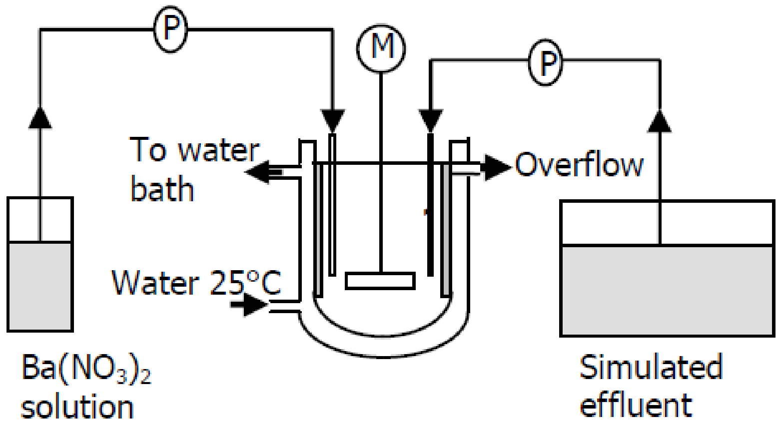

Figure 1: Experimental set up used for experiments in continuous reactor

To test the modeling approach, simulations of the co-precipitation of strontium ions with barium sulfate have been performed in CSTR. The DF values at steady state as a function of mean residence time are reported in Figure 2.

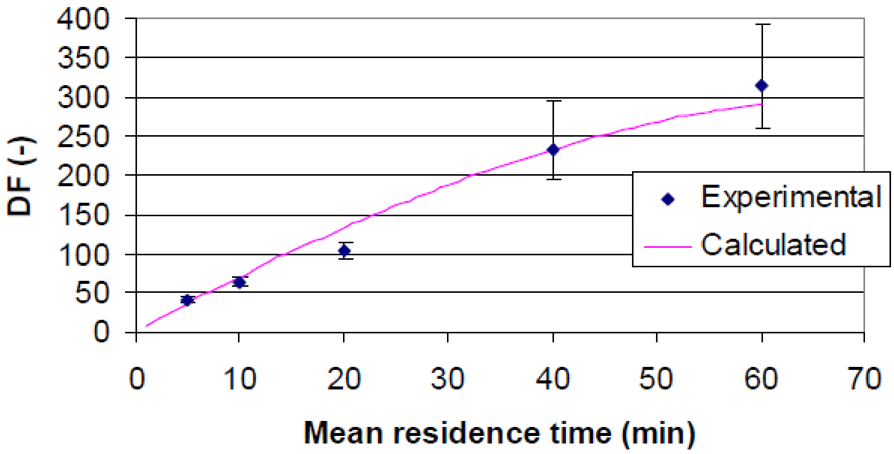

Figure 2: Decontamination factor (DF) as a function of mean residence time. Comparison between experimental data and calculations. Experiments carried out with inactive Sr^{2+} (10mg/L)

Figure 2 shows a good agreement between experiments and the co-precipitation model. This methodology will now be extended to other co-precipitation systems. The final goal is to predict the behavior of complex mixtures at an industrial scale.

Optimization of reactor design

It can be seen from previous paragraph that DF results from a compromise between residence time, hydrodynamic (via the mass transfer parameter) and concentration of the adsorbent. In order to reduce the mass ratio of adsorbent to fixed contamination, and also to reduce the size of the tanks for the same global capacity, we worked out the design of the co-precipitation reactors. An optimized reactor type called the settling tank reactor has been designed to this purpose (see Figure 3).

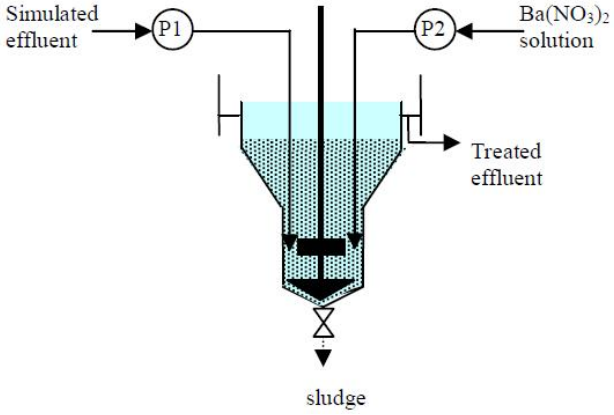

Figure 3: Schematic flow scheme of the reactor-settling tank pilot.

The settling tank reactor is a fluidized bed type reactor i.e. there is an accumulation of precipitates into the apparatus. Since the concentration of $BaSO_4$ is quite large in reactor, the sorption capacity of the bed is much higher than in classical CSTR. Moreover, an efficient solid/liquid separation is performed in the apparatus.

In a new study accepted for publication the theoretical approach of this reactor will be presented [4]. One of the key results is that for the same decontamination efficiency, the volume of the new reactor is $\square^{3/4}$ times smaller than the volume of CSTR where □ is a dimensionless liquid-solid separation parameter. This optimized concept enables the reduction of tank volume by a factor of about 10 compared to standard CSTR reactors (□=20 for realistic operating conditions). Thus, in the view of an industrial application a great reduction of the plant volume is expected.
Moreover, the design of mobile co-precipitation units for accidental issues based on such designs would be of great interest.

APPLICATION OF THE CO-PRECIPITATION KNOW HOW TO FUKUSHIMA WATERS

Industrial Context

The co-precipitation technique was proposed by AREVA and VEOLIA to TEPCO in April 2011 to solve the problem of the contaminated waters produced by the cooling of the FUKUSHIMA nuclear reactors after the tsunami of March 11th. Daily, hundreds of cubic meters of water (~500 m^3 per day) were added at that time to cool down the reactors. In a first stage, the added water has been sea water until end of March and then soft water has been sprayed or injected.

CEA has been involved in the project since the beginning to provide a scientific and experimental support to AREVA and VEOLIA teams. CEA has been particularly involved in the following areas:

- Co-precipitation expertise (process and basic knowledge) and radioactive test
- Test of new adsorbents for the Cesium and Strontium removal
- Knowledge of the behavior of radioelements in sea water.

AREVA & VEOLIA designed a co-precipitation unit with a maximum capacity of 400 $m^3.h^{-1}$. The process called ACTIFLO RAD has been implemented at Fukushima site and is a part of the overall process scheme (illustrated in the Figure 4 below). The main objective of TEPCO at the time was to recycle soft water for the cooling of the reactors. Thus a reverse osmosis unit as well as evaporator units have been implemented to remove the salts (mainly NaCl) contained in the water. The ACTIFLO RAD process has been introduced after the so called KURION process which consists in adsorption columns to remove Cs activity.

The ACTIFLO unit as well as the global process had been started by the end of June 2011.

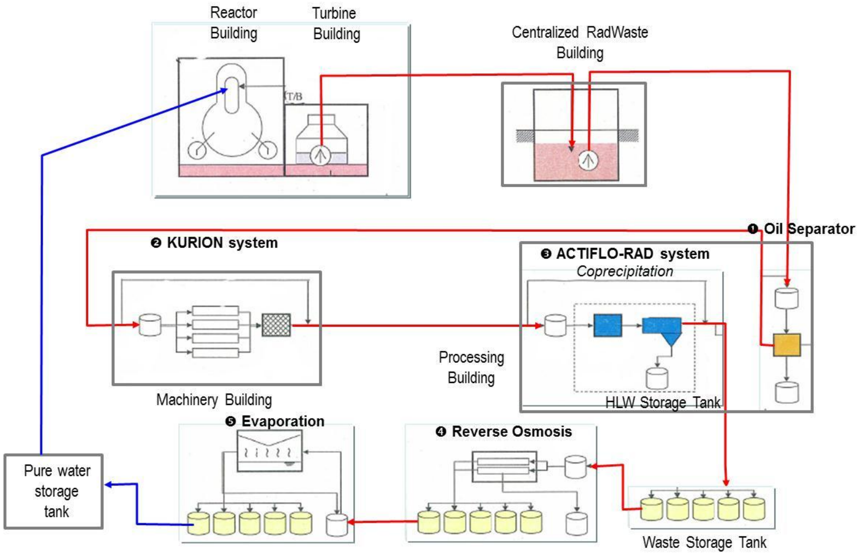

Figure 4: Flowsheet of the liquid wastes treatment unit at Fukushima site

Figure 5 shows the ACTIFLORAD facility.

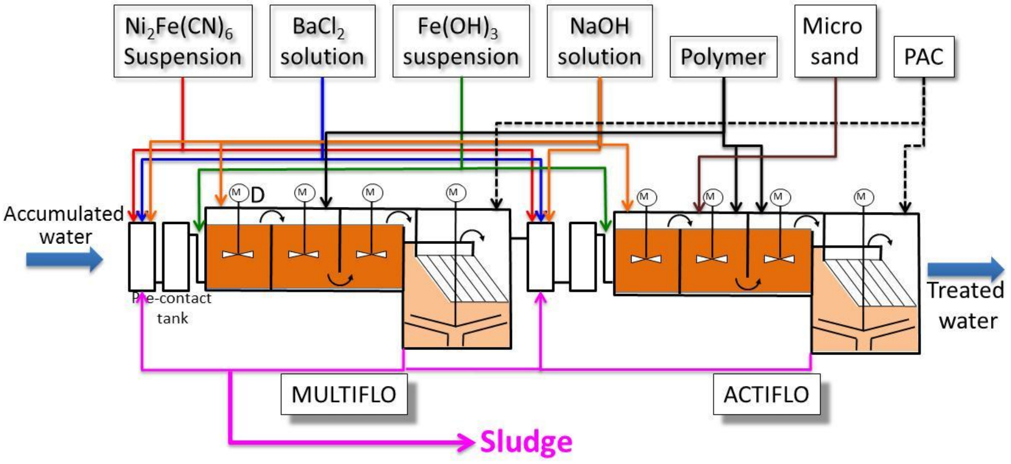

Figure 5: Presentation of the ACTIFLO RAD facility

The ACTIFLO facility (Figure 5) has been originally designed for the cleaning of domestic waters by VEOLIA. It is originally devoted to the co-precipitation of ferric hydroxide and the addition of a polymer mixed with a micrometric sand to enhance agglomeration and settlement of particle thus facilitating the separation process in the last tank. The step with ferric hydroxide has been kept in order to remove alpha emitters.

For the case of Fukushima waters, additional tanks have been added upstream from the ACTIFLO facility. This is the so called MULTIFLO step which has been designed by AREVA with the help of CEA. The combination of MULTIFLO and ACTIFLO process has been called the ACTIFLO RAD facility. In the MULTIFLO facility two main chemical reactions take place:

- To remove ^{90}Sr thanks to the addition of barium and sulfate.
- To remove ^{134}Cs & ^{137}Cs due to the addition of a pre-formed precipitate of ferrocyanates

The sludge produced in the process is partially recycled upstream to increase contact time with contamination. An approaching 3 h contact time has been set for a global 50 $m^3.h^{-1}$ flowrate of processed solution.

Total volumes to be decontaminated have been initially estimated in the order of 70 000 m^3 and around 100 000 m^3 by the end of May for intermediate contamination level (10^5 to 10^9 $Bq.L^{-1}$). The total amount of low to intermediate contaminated water was estimated about 200 000 to 300 000 m^3. Table 1 below gives the initial concentration of radioactivity for each for reactor units.

		Unit 1	Unit 2	Unit 3	Unit 4
^{131}I	Bq/L	3×10^7	2×10^9	1.6×10^8	3.1×10^5
^{134}Cs		1.2×10^8	2.6×10^9	1.4×10^8	2.0×10^4
^{137}Cs		1.6×10^8	2.8×10^9	1.6×10^8	2.2×10^4
^{140}Ba		<560	2.4×10^8	1.5×10^7	$<0.68 \times 10^3$
^{89}Sr		5.7×10^4	7.0×10^8	8.6×10^7	$<0.13 \times 10^3$
^{90}Sr		2.1×10^4	1.4×10^8	1.5×10^7	<0.13

Table 1: Activity of Fukushima intermediate contaminated waters (values as measured April 13th 2011) - Source: http://www.tepco.co.jp/en/nu/fukushima-np/images/handouts_110522_04-e.pdf)

The majority of the activity comes from Cesium (134 & 137) and Iodine 131. No alpha emitters are present. Considering the extremely low period of iodine (8 days), this element has not been considered for the co-precipitation process.

The industrial objective was initially to reach a decontamination factor over 100000 for Cesium and 10 for Strontium 90. The flowrate has been set to 50 $m^3.h^{-1}$ in order to match the ACTIFLO RAD facility with the kinetic of other decontamination processes involved in the global decontamination scheme.

Optimization of co-precipitation operation

Water composition

As water to be treated was in a first step very saline and expected to be less and less saline due to the use of soft water for the cooling. The issue has been to design a process able to remove traces of Cs and Sr in a very saline medium. Seawater has been considered as a conservative approach for process design.

Near La Hague Seawater has been taken as representative of Fukushima seawater according to the urgent need at developing a solution for Fukushima. Table 2 below presents the composition of La Hague seawater.

Cations	Sr (mg/l)	Cs (mg/l)	Na (g/l)	Mg(g/l)	Ca(mg/l)	K(mg/l)
	7,78	< 10µg/L	9,58	1,28	405	529
Anions	Cl (g/L)	F (mg/L)	Br (mg/L)	SO_4 (g/L)	PO_4 (mg/L)	NO_3 (mg/L)
	19	0,56	66,3	2,7	<1	<1

Table 2: Composition of La Hague seawater

Strontium decontamination

One can see from Table 2 that the non-radioactive Strontium concentration in natural seawater is not negligible.
Not only inactive Strontium will adsorb on to $BaSO_4$ particles but also Ca^{2+}, Mg^{2+} thus decreasing the co-precipitation efficiency. To avoid this it has been decided to increase the ratio of sulfate to sodium in the solution. To achieve a decontamination factor greater than 10 in the bulk, experimental tests made on doped seawater with representative activity of strontium 90 it has been shown that a ratio $R = SO_4/Ba$ of 7 was necessary. Experimental studies addressing barium sulfate precipitation as illustrated in the previous paragraph have shown that the crystal size of $BaSO_4$ decreases with the increase of sulfate excess. It has been observed that the mean crystal size decreases from 3.6 µm to 1 µm when R increases from 2 to 5. Thus the exchange surface able to adsorb Strontium ions increases.

Of course, the sulfate already present in the seawater (concentration about 2.7 g/L) has been taken into account for the process design. The interest of introducing $BaCl_2$ instead of $Ba(NO_3)_2$ has also been assessed regarding the high concentration of Chlorine in seawater.

Cesium decontamination

Addressing the Cesium case, hexaferrocyanates are known for their efficiency and selectivity toward Sodium. Hexaferrocyanates are used in both Marcoule and La Hague liquid wastes decontamination facilities. Cs^+ is precipitated as $Cs_2NiFe(CN)_6$ in the bulk. In the case of Fukushima, the concentration value of 300 mg.L^{-1} of $Fe(CN)_6$ was enough to reach a decontamination factor greater than 1000.

Efficiency of the decontamination process

By mid of September, the amount of water processed through the ACTIFLO RAD unit was 77 400 m^3.
The overall observed decontamination factor on both ^{134}Cs and ^{137}Cs was ranging from 10^5 to 10^6 for the 2 steps of the process (Kurion and ACTIFLO). The DF of the ACTIFLO unit processing downstream of the Kurion adsorption columns ranges from 10^3 to 10^4. An estimated value of 100 000 m^3 contaminated water needs to be processed.

Co-precipitation has proven to be a robust and efficient technology. The amount of generated sludge is estimated around a few 0.1% in volume of the total processed liquid wastes. A conditioning step of the sludge is now necessary. Both cementation and vitrification techniques can be studied for the future conditioning of Fukushima sludges.

CEA has already developed cementitious formulations for the conditioning of sludges for the liquid waste plant in Marcoule. The process will be implemented in 2015. Furthermore, CEA has gained experience in the field of conditioning evaporators concentrates in a cementitious matrix [5,6].

PROSPECTIVE R & D USING NEW ADSORBENTS

The counterpart of co-precipitation process is the production of sludge.
In this paragraph two prospective techniques developed in CEA and belonging to the family of cartridges process will be presented as promising for future applications dealing with liquid wastes decontamination. These techniques aim at:

- Reducing the volume of solid wastes generated by the liquid wastes treatment
- Producing solid wastes that are easily and efficiently conditioned
- Efficiently control the hydrodynamic of the process

The first technique consists in grafted ferrocyanates on glass particles that are totally compatible with the vitrification process.
The second technique deals with a new generation of reversible adsorbents. In this case adsorbent can be recycled. Thus the production of solid wastes is greatly reduced.
Both techniques enable an efficient control of the hydrodynamic in the cartridge by controlling the size of the particles or solid substrate porosity on which the adsorbents are grafted.

Development of grafted ferrocyanates on glass material

Development of grafted ferrocyanates on glass material is discussed in Reference [7]. Commercial Cs or Sr selective inorganic materials are in most cases available in the form of a fine powder. They are therefore difficult to use in a continuous process.

Glass material used as a substrate for the grafting of ferrocyanates has two major advantages: the compatibility of the material with the vitrification process and the tunable geometry of the glass particles. A cartridge type application is foreseen. The pressure loss across the cartridge is thus controlled by the size of the glass particles. The process can work in a continuous mode.

CEA & ICSM launched a common study in 2010 focused on grafted glass. A patent is pending for the synthesis and grafting of ferrocyanates in the glass porosities. The grafting process involves a pyridine function. Every kind of metal hexaferrocyanate can be grafted on the pyridine group as illustrated in the figure below. Slightly different synthesis ways based on this principle are also studied.

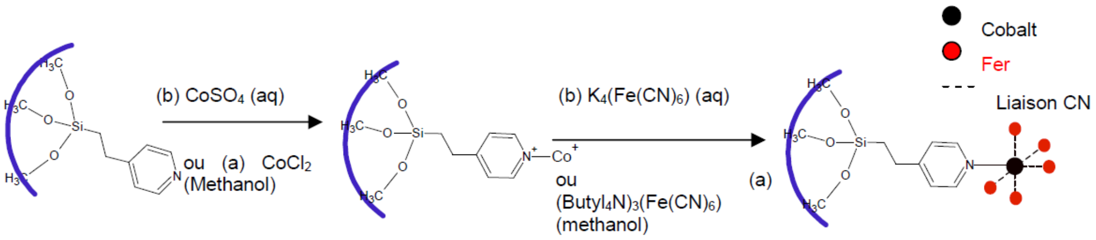

Figure 6: Grafting of hexaferrocyanates on glass particles

Table 3 presents typical values of K_d obtained with different adsorbents including grafted glass in a doped seawater [^{137}Cs] ~30 000 Bq.L^{-1}. This last value is close to activities values at the inlet of ACTIFLO RAD unit.

K_d (in L.mg^{-1}) is the equilibrium value of the ration of adsorbed cesium to residual cesium in the solution.

$$K_d = \frac{Q}{C} = \frac{C_0 - C}{C} . \frac{V}{m} \quad \text{Eq.2}$$

With:

Q = equilibrium concentration of cation in the adsorbent (mg.g^{-1})
C_0 =initial concentration of cation in the solution (mg.L^{-1})
C = equilibrium concentration of cation in the solution (mg.L^{-1})
V = volume de solution (L)
m = mass of adsorbent (g)

	Hexaferrocyanates (colloidal) As described in Co-precipitation section	**Grafted particles (this study)**	**Commercial grafted hexaferrocyanates**
K_d (L.mg^{-1})	**$4x10^6$**	**$8x10^5$**	**$9.6x10^5$**

Table 3: Typical values of K_d obtained with different adsorbents

Grafted glass particles are already exhibiting very high K_d values. These values are close to representative commercial products. The grafting method is now entering in an optimization phase. We hope to increase the K_d value. The increase of grafting density is one of the ways to reach this objective.

Because the manufacturing of grafted glass particles is easy to transfer at an industrial scale, pre industrial scale quantities have already been elaborated.

It is also expected in a near future to work out grafted glass particles with other adsorbents.

Development of reversible adsorbents

In this case, the aim is not only to graft a complexing molecule onto a solid substrate as in the previous example but two challenging innovations are at stake:

- To enlarge the spectrum of complexing molecules that can be used for decontamination including organic molecules
- To easily regenerate the substrate

Since only a thin thickness (few nm) of complexing organics will be grafted at the surface this is not considered as a problematic issue regarding the production of radiolysis gases. Only very small gases volumes will be produced during radiolysis molecule decomposition.

Due to the poor solubility of organic molecules in aqueous medium, these are generally applied in a liquid/liquid extraction processes. Organic molecules are solubilized in an organic solvent and the process consists in a complexing step followed by a decomplexing step.

This type of process generates organic secondary wastes and is not as simple as ion exchanging columns or cartridges to foresee an industrial application especially in the case of accidental issues.

Therefore, CEA is studying an alternative solution. Macromolecules are grafted on an inorganic conducting material such as metal or carbon fabrics. The reversibility of the reaction is obtained by applying an electrical current to release the complexed contamination (Sr^{2+}, Cs^{+}, Co^{2+},…). This technology has been proven feasible for standard complexing molecules like polyacrylic acid films grafted on gold surfaces [8]. The technique is illustrated in Figure 7.

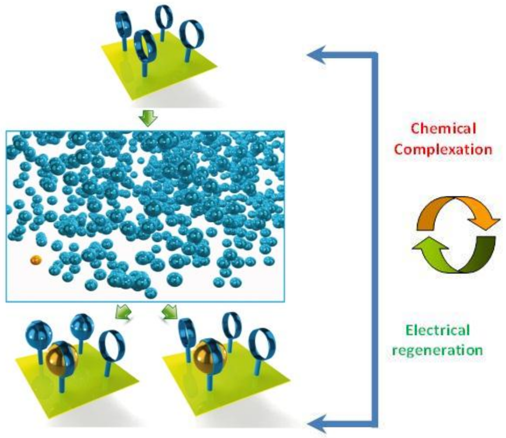

Figure 7: Presentation of reversible adsorption macromolecules grafted adsorbents
-The molecule to be removed (yellow, ex: Cs) in an aqueous system containing high concentration of competitive ion (blue, ex: Na)-

The steps toward the development of a fully reversible process based on the use of grafted macromolecules will need to go through the following stages:

1. Demonstrate the feasibility of grafting macromolecules
2. Demonstrate the high selectivity toward solubilized contamination is not affected by grafting
3. Demonstrate the reversibility of the reaction applying electrical current
4. Increase the density of grafting
5. Produce the grafted substrate at an industrial scale
6. Implement cartridges fitted with electrical reversibility option

Macromolecules open a wide range of complexing opportunities with a high efficiency and selectivity.
This is a major challenge since the classical inorganic adsorbents do work well for Cs as previously seen (for co-precipitation or grafted on a glass matrix) but their selectivity toward Sr in such a complex aqueous system as seawater is generally limited.

CONCLUSIONS

A chemical process unit based on co-precipitation technique has been successfully and quickly implemented by AREVA at Fukushima site for the processing of contaminated waters. The asset of this technique is its ability to process large volumes in a continuous mode. Several chemical products can be used to address specific radioelements such as: Cs, Sr, Ru. Its drawback is the production of sludge (about 1% in volume of initial liquid volume).

CEA developed strategies to model the co-precipitation phenomena in order to firstly minimize the quantity of added chemical reactants and secondly, minimize the size of co-precipitation units. We are on the way to design compact units that could be mobilized very quickly and efficiently in case of an accidental situation.

Addressing the problem of sludge conditioning, cementation appears to be a very attractive solution.

Fukushima accident has focused attention on optimizations that should be taken into account in future studies:

- To better take account for non-typical aqueous matrixes like seawater
- To enlarge the spectrum of radioelements that can be efficiently processed and especially short lives radioelements that are usually less present in standard effluents resulting from nuclear activities
- To develop reversible solid adsorbents for cartridge-type applications in order to minimize wastes.

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