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Extraction of rubidium from the concentrated brine rejected by integrated nuclear desalination systems

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ABSTRACT

Rubidium is one of the elements present in the concentrated brine rejected by desalination systems. In view of the potentially high price of the pure metal, it is worthwhile to investigate its extraction, even though presently the available Rb resources are adequate enough to meet the current demands.Two methods have been reported. The first makes use of the ion-exchange resins and the second of the complexation of Rb with specific molecules (calixarenes) followed by one or more nanofiltration/reverse osmosis (NF/RO) stages. First results of calculations indicate that the two methods would be technically very attractive but much experimentation would still be required before an industrial scale extraction process can be evolved.

Keywords: Nuclear desalination; Zero desalination plant discharge; Valorisation of the rejected brine

1. Introduction

In a previous paper [1], we indicated the possibilities of extracting various strategic materials (Table 1) from the concentrated brine that would be rejected by integrated nuclear desalination systems.

One of the elements of interest, as shown in Table 1, is Rubidium. It could theoretically be a source of additional revenue, provided there is sufficient demand in the future.

Rubidium is used in the fabrication of photo-electric cells and fuel cells, as absorber of gases in vacuum tubes and for the fabrication of monolithic lasers. Rb isotopes (e.g. Rb^{86}) are frequently used in various medical examinations, especially for myocardial ones.

Rubidium is mostly present on earth in the form of lepidolite ($K(Li,Al)_3(Si,Al)_4O_{10}$), which may contain up to 3.5% of Rb_2O. It is in fact extracted from lepidolite as a by product of lithium extraction.

The world production of rubidium (and its compounds) is about 5000 kg/year. Rubidium production from lepidolite satisfies the present world needs. However, many other strategic and military applications are being considered and it is judicious to look for alternate Rb production.

In a typical integrated desalination plant (Fig. 1), Rb is found in the second brine stream, corresponding to the reject stream after the desalination stage.

The composition of brine to be treated (brine 2) is: $[Rb] = 5.6 \times 10^{-6}$ mol/L, $[Na] = 1.8$ mol/L.

2. Extraction of rubidium

Rubidium is present in the seawater in the form of cationic monovalent Rb^+ ions. Dissolved in water, it behaves exactly as other alkali cations. We outlined

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Table 1
List of valuable elements, which could be extracted from the brine, rejected by a RO plant, with a desalting capacity of 294,000 m^3/day

Element	Seawater content (mg/L)	Available quantity (t/year)	Major use	Selling price ($/kg)	Value (M$/year)
Na	1.05×10^4	1.5×10^6	Fertilizers	0.13	180
Mg	1.35×10^3	1.9×10^5	Alloys	2.8	525
K	3.8×10^2	5.3×10^4	Fertilizers	0.15	8
Rb	1.2×10^{-1}	17	Laser etc.	11,180	190[a]
P	7.0×10^{-2}	10	Fertilizers	0.02	0.0
In	2.0×10^{-2}	3	Metallic protection	300	0.9
Cs	5.0×10^{-4}	0.07	Aeronautics	63,000	4
Ge	7.0×10^{-5}	0.01	Electronics	1700	0.02

[a] No market price is available as Rb is not traded. The price cited is of 99.75% pure Rb, as offered by one company [7].

in [1] that the chemistry of rubidium is very close to that of Cs.

It is for this reason that we adopted the extraction protocols for caesium to extract rubidium.

RbCl is in particular more soluble in water than KCl but significantly less than CsCl. Thus at 100 °C, the solubility of RbCl is 138.9 g in 100 mL of water.

As for caesium, two approaches for rubidium extraction are thus retained:

- Utilisation of ion-exchange resins.
- Utilisation of complexing molecules (e.g. calixarenes) for the extraction of heavy cations.

2.1. Extraction by ion-exchange resins

The most efficient and economic method that we have examined is the use of cation-exchange resins.

Ion-exchange is essentially a stoichiometric process. Consider an insoluble material in the form of beads (resin), containing ions of type B and placed in a well stirred solution, containing ions of type A.

According to the affinity of the resin for A, as equilibrium is established, ions B diffuse out of the beads into the solution and ions A diffuse from the solution to the beads. The exchange process, which obeys the law of mass action, can be represented by:

$$\underset{\text{resin}}{Z_A B^{zB}} + \underset{\text{solution}}{Z_B A^{zA}} \longleftrightarrow \underset{\text{resin}}{Z_B A^{zA}} + \underset{\text{solution}}{Z_A B^{zB}}$$

where zA and zB are the respective valencies of A and B.

In fact, rubidium is poorly concentrated in seawater. The use of the ion-exchange resins is therefore well adapted. In this field, the best resins are based on a framework of titanium silicates whose generic formula is of the type $M_2Ti_2O_3SiO_4{\cdot}nH_2O$, where M designates the ion that is exchanged (H^+ or Na^+).

In acidic work conditions, which result from the first steps of the protocol presented in [1], the framework exhibits a high affinity for heavy alkali metals: rubidium, caesium and potassium. The exchange cation is usually sodium. The resulting selectivity of this resin is then in the order $Rb^+ > Cs^+ > K^+ >> Na^+ > Li^+$.

In [2,3], a new ion-exchange resin, marketed by UOP as IONSIV IE-911, has been used in experiments aiming to remove radioactive Cs^+ from aqueous nuclear wastes.

The resin is stable to radiation, highly selective for Cs (and Rb) relative to Na, K and H and performs well in acid, basic or neutral solutions. It has been shown to be particularly adapted for Cs and Rb extraction in solutions with high concentrations of Na. Thus in [2,3], solutions had concentrations of Na^+ of the order of 3 M, as compared to 1.8 M in the rejected brine.

The equilibrium exchange between the solution and the resin (in which silico-titanate is represented by ST) is given as:

$$\text{ST} - (\text{M}^+)_n + \text{A}_n^+ \Leftrightarrow \text{ST} - \text{A}_n^+ + n\text{M}^+$$

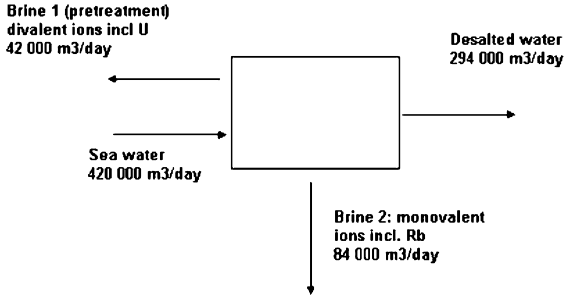

Fig. 1. Schematic flow of input and output streams in an integrated desalination system.

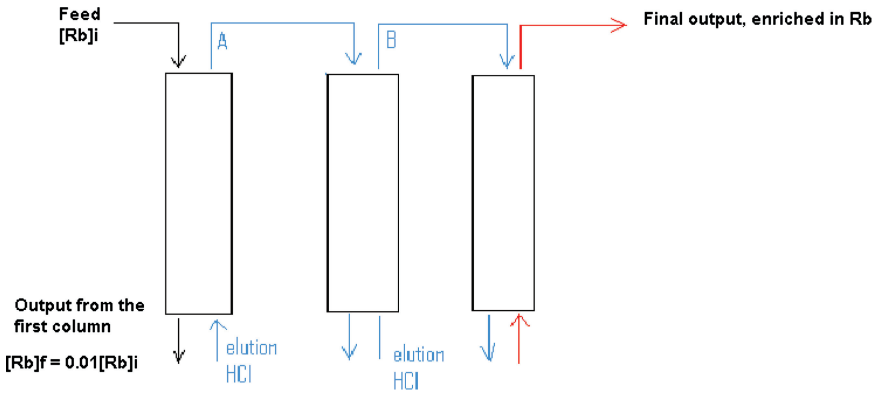

Fig. 2. Principle of the successive adsorption/elution process using ion-exchange resins.

The equilibrium constant for this process applied to the extraction of Rb is then given by

$$K_{\mathrm{Na}}^{\mathrm{Rb}} = \frac{m_{\mathrm{Na}} \times X_{\mathrm{Rb}}}{m_{\mathrm{Rb}} \times X_{\mathrm{Na}}},$$

where m_{Na} and m_{Rb} are respectively the molalities of Na and Rb cations in the solution and X_{Na} and X_{Rb} are the molar fractions in the resin.

The effective equilibrium constants for different processes can be derived from the curves presented in [2]. They are as follows:

$$K_{\mathrm{Na}}^{\mathrm{K}} = 40, \quad K_{\mathrm{Na}}^{\mathrm{Rb}} = 1995, \quad K_{\mathrm{Na}}^{\mathrm{Cs}} = 100,000.$$

It is thus clear that Rb cannot be extracted in only one step:

- First, the equilibrium constant for Cs is about 100 times higher.
- Second, the ratio between Na and Rb concentrations is more than 10^7. Thus the ratio $K_{\mathrm{Na}}^{\mathrm{Rb}}/K_{\mathrm{Na}}^{\mathrm{K}}$ ($\sim$50) cannot allow a one step separation.

One would thus need a series of columns as shown in Fig. 2.

The solution obtained in stage A is still quite rich in Na but the ratio of Rb/Na concentrations is higher than that in the feed because of the preferential adsorption of Rb.

This process is repeated in B and in successive columns until the concentration of Rb is sufficiently increased.

2.2. Modelling of rubidium adsorption

The dimensioning of a fixed bed adsorption column requires the breakthrough curve, characterized by the general Langmuir equation:

$$q = \frac{q_t KC}{1 + KC}$$

where q is the equilibrium concentration in the resin (mmol/g), C is the molar concentration in the solution and K and q_t are Langmuir parameters representing the characteristic affinity of the ion (in our case Rb) for the resin.

The breakthrough curve can be obtained experimentally and/or through modelling. The modelling scheme that we have adopted, following [4], is briefly described below:

The performance of a fixed bed is generally expressed in terms of the solute concentration in the exit stream as a function of time for a given bed length, liquid velocity, physical chemical properties, etc.

One thus obtains the set of differential equations of the form:

$$e\frac{\partial C}{\partial t} + U_L.\frac{\partial C}{\partial z} - e.E_L\frac{\partial^2 C}{\partial^2 z} + J.a = 0, \qquad (1)$$

where C, is the concentration in the solution at a column height z and at an instant t, e is the bed voidage (= 0.34), U_L is the superficial liquid velocity, E_L is the axial dispersion coefficient, J is the flux of the solution diffusing towards the resin through the boundary layer of the bead surface, and a is a constant representing the geometrical characteristics of the bed ($= 6(1 - e)/d$, where d is the resin bead diameter).

and,

$$(1 - e)\frac{\partial q}{\partial t} - J.a = 0 \qquad (2)$$

where q is the concentration in the resin.

Eqs. (1) and (2) are coupled by

$$J = k(C - C_s) \qquad (3)$$

and,

$$q = \frac{QKC_s}{1 + KC_s}, \tag{4}$$

where C_s is the solute concentration at the surface of the resin and in Langmuir equilibrium at all instants t, k is the mass transfer coefficient, dependent on hydrodynamic conditions, Q and K are Langmuir constants specific to the ion concerned and the resin.

The above equations can be further simplified by making the following assumptions:

- There is no axial dispersion, thus $E_L = 0$.
- C_s will never exceed C, the concentration in the solution. Because for Rb, C is lower than 5.6E^{-6} mol/L, concentration in the brine, the product KC_s is negligible, compared to 1 and thus:

$$q = QKC_s \text{ and } J = k * \left(C - \frac{q}{Q * K}\right) \tag{5}$$

- The diffusion kinetics of Rb towards the resin is mainly governed by two phenomena: by the transfer in the diffusion boundary layer or by internal transfer within the bead. When the concentrations are very low (as is the case here), the transfer is predominantly governed by the boundary layer. This can be verified by the Helfferich number [5], calculated as:

$$\text{Helff} = (0.167 + 0.0064\ K^{\text{Na}}_{\text{Rb}}).\frac{\delta.Q.\bar{D}}{0.015D.C_0.d}),$$

where the thickness δ of the boundary layer is given by:

$$\delta = 0.05.d(\rho d U_L/\mu)^{-0.84},$$

where C_0 is the total solution molar concentration, d is the average resin bead diameter (m), μ is the liquid viscosity (kg/m s), ρ is the liquid density (kg/m^3), $\bar{D}$ is the diffusivity in resin (m^2/s) and D is the diffusivity in solution (m^2/s).

In our case, the value of *Helff* is very high compared to 1. Thus, diffusion is controlled by the transfer in the diffusion boundary layer.

Incorporating these assumptions in Eq. (1) and (2) leads to:

$$e\frac{\partial C}{\partial t} + U_L.\frac{\partial C}{\partial z} - + ak\left(C - \frac{q}{QK}\right) = 0 \tag{6}$$

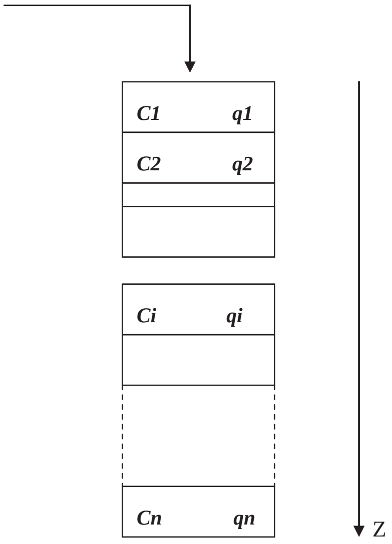

Fig. 3. Discretisation of the adsorption column.

and

$$(1 - e)\frac{\partial q}{\partial t} - ak\left(C - \frac{q}{QK}\right) = 0 \tag{7}$$

where C and q are space and time dependent variables.

2.2.1. Space-time discretisation

The systems of Eqs. (6) and (7) were resolved by discretisation in space (step of length, h along the Z direction) and in time, as shown in Fig. 3.

The initial conditions are:

- Whatever the value of Z, at $t = \underline{0}$, $qi = 0$, since the resin is not supposed to contain any Rb initially.
- Whatever the value of Z, at $t = 0$, $C_i = 0$, since the liquid inside the column is not supposed to contain any Rb initially.

The other associated boundary conditions are:

- Whatever the value of t, $C_i = 5.6 \times 10^{-6}$ mol/L, since the brine entering the column is supposed to have a constant Rb concentration.

The system of Eqs. (6) and (7), based on partial derivatives, can be transformed into a system of ordinary differential equations by successive finite differentiations, leading to $2n - 1$ equations for an nth order mesh:

$$e\frac{\partial C_2}{\partial t} = -U_L\frac{C_2 - C_1}{h} - ak\left(C_2 - \frac{q_2}{QK}\right)$$

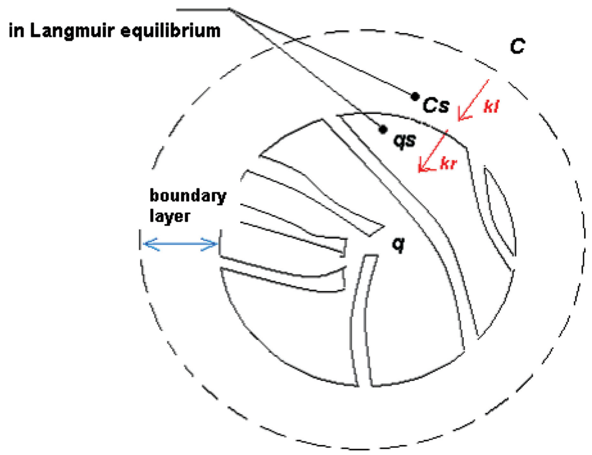

Fig. 4. Radial view of the resin bead.

$$e\frac{\partial C_3}{\partial t} = -U_L\frac{C_3 - C_2}{h} - ak\left(C_3 - \frac{q_3}{QK}\right)$$

$$\vdots \qquad\qquad\qquad \vdots$$

$$e\frac{\partial C_n}{\partial t} = -U_L\frac{C_n - C_{n-1}}{h} - ak\left(C_n - \frac{q_n}{QK}\right)$$

and for the resin

$$(1-e)\frac{\partial q_1}{\partial t} - ak\left(C_1 - \frac{q_1}{QK}\right) \;=\; 0-$$

$$\vdots \qquad\qquad\qquad \vdots$$

$$(1-e)\frac{\partial q_n}{\partial t} - ak\left(C_n - \frac{q_n}{QK}\right) \;=\; 0$$

This is analogous to the matrix representation:

$$Y(t+dt) = dt.M.Y(t) + Y(t),$$

where $Y(t)$ is the vector representing the variables $C_1(t)...C_n(t)$ and $q_1(t)...q_n(t)$.

The above system then can be resolved by using the ‘‘ode45’’ function of MATLAB.

The breakthrough curve will then represent the variation with time of the concentrations, Cn(t), coming out from the column ($C_n(t)$) is the concentration for the exit stream.

The system can be further improved by taking into account the internal transfer within the particle (Fig. 4).

The flux J can be defined as $J.a_p = \frac{dq}{dt}$, where a_p is the bead specific surface.

Since there is a perfect equality between the input and output fluxes, we obtain,

$$J = k_L(C - C_s) = k_R(q_s - q). \tag{8}$$

Here q_s and q are, respectively, the concentration at the bead surface and the average concentration within the bead, k_L is the liquid film mass transfer coefficient and k_R is the resin bead mass transfer coefficient. k_R can be written as $k_R = 10\frac{\overline{D}}{d}$, with $\overline{D}$ as the diffusion coefficient in the resin. For Rb, the value of this coefficient is of the order of 3.11×10^{-11} m^2/s, which leads to $k_R = 7.5 \times 10^{-7}$ m/s. C_s and q_s respect the Langmuir equilibrium condition.

$$q_s = KQC_s \tag{9}$$

Hence, from (6), we obtain

$$q_s = q + \frac{k_L}{k_R}(C - C_s) \tag{10}$$

And combining (7–9), we obtain

$$J = k_L + \frac{KQ}{KQ + {k_L}/{k_R}}\left(C - \frac{q}{KQ}\right) \tag{11}$$

Instead of the expression for J in (3) and (5).

2.3. Results and discussion

The above equations were resolved by using the MATLAB software. After initial checking up of internal consistency of the programme thus developed, we first calculated the dimensions of the column(s).

The MATLAB simulation allowed us to obtain the break-through curve as shown in Fig. 5, based on the low liquid velocity of 1.08 m/h (0.0003 m/s). This is usually the value retained in ion-exchange technology since the low velocity favours a better exchange between the solution and the resin.

However, in view of the sea-water capacity of 84,000 m^3/day, and for a column height (L) of 3 m, we obtain the resin volume of 6400 m^3, which is evidently too high.

It is for this reason that we adopted another break-through curve (Fig. 6) based on the liquid velocity of 14.4 m/h (0.004 m/s) and a column height of 1.5 m.

Naturally, the choice of such high velocity is not ideal but this solution is a necessity in view of the very high seawater flow rate.

Results obtained from these two choices are presented in Table 2.

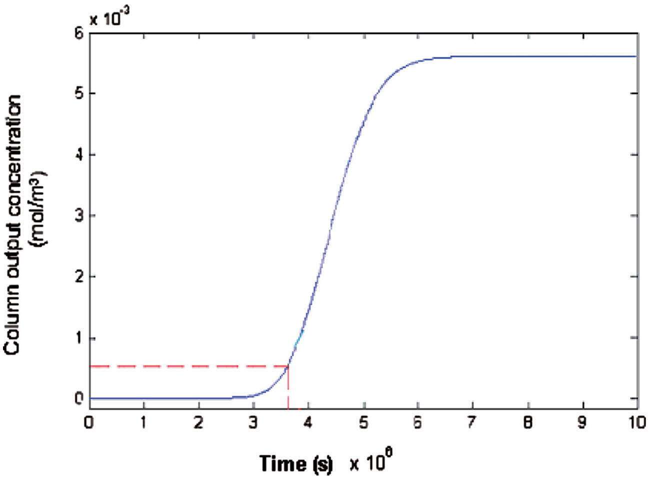

Fig. 5. Break through curve for Rb; $U_L = 0.0003$ m/s and $L = 3$ m.

Table 2
Calculated dimensions and other parameters for ion-exchange columns

Parameter	Options for the column size and liquid velocity	
	$U_L = 0.0003$ m/s et $L = 3$ m	$U_L = 0.004$ m/s et $L = 1.5$ m
Required number of columns	11,550	3415
Required resin volumes (m^3)	6400	240
Time before saturation (days)	42	1.3
Pressure drop (Pa)	9300	64,000
% of resin used	82	70

The 3415 columns in the second solution can be contained in a square of about 59 × 59 m^2, which does not seem to be unreasonable.

To recover Rb, an elution step is required after resin saturation.

Resin regeneration is generally realized by washing the resin with hydrochloric acid (to form chlorides) and by a second wash with sodium chloride (or sodium hydroxide). According to our calculations, the total elution time (second solution in Table 2) is about 3 h.

Table 3 shows the relative values brine volume and concentrations of Na and Rb in the initial brine flux and after the elution step.

Potassium would be, after rubidium, the most attracted element. A further purification would be necessary to separate these two materials. Some potassium-selective membranes make this separation possible. The composition of these membranes is 33 wt% PVC, 66 wt% plasticizer and 1 wt% ionophore. The ionophore is *cis* bis-15-crown-5 and the plasticizer is *o*-nitrophenyloctyl ether (NPOE).

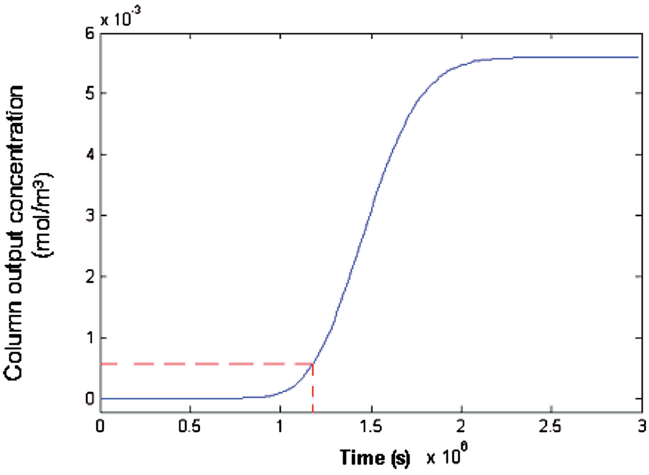

Fig. 6. Breakthrough curve for Rb; $U_L = 0.004$ m/s and $L = 1.5$ m.

With such low volumes (which could be further reduced by evaporation), it is possible to separate Rb by a liquid–liquid extraction technique as reported in [1], to improve the efficiency of recovery.

Note that these results (first ion-exchange, then elution) are obtained with just one step (one column for Rb fixation, one column for Rb elution).

3. Extraction of rubidium by complexation–nanofiltration (NF)

The main difficulty in extracting Rb from the rejected brine comes from its very low concentration in the initial seawater feed. Obviously, the efficiency of the resin would depend upon the concentration.

A solution for considerably increasing the ion-exchange resin efficiencies could be through the use of NF, assisted by complexation by specific molecules. A complex is a polyatomic edifice in which a central atom is linked to molecules (or ions) called ligands.

This approach has been developed and perfected at Cadarache Atomic Centre (CEA) [5] in the context of decontamination of highly saline effluents.

The basic principle of the process is as follows:

One selects a specific molecule for complexation in such a way that the ligands are not able to pass through the pores of a given NF membrane (Fig. 7).

Dozol et al. [6] particularly developed the crowned ether molecule calixarene (calix-4-arene-bis-crown 6-tetrahydroxyle), which has the property of complexing caesium at high pH values (pH > 11).

Table 3
Initial and post elution volumes and concentrations

	Brine	Solution obtained	Gain
Volume over 24 h	84,000 m^3	1850 m^3	The volume has been reduced by a factor of 45
Na concentration	1.8 mol/L	0.06 mol/L	Na concentration reduced by a factor of 30
Rb concentration	0.0056 mol/m^3	0.375 mol/m^3	Rb concentration increased by a factor of 67

Since Cs and Rb are chemically very similar, we consider that the conditions for extraction of Cs are also valid for the extraction of Rb, although experiments need to be made to verify this assumption.

In a brine solution, the calixarene molecules form a complex with Na^+ or with Cs^+/Rb^+:

$$\text{Cal} + \text{Cs}^+ \rightarrow (\text{Cal. Cs})^+ K_1 = 23{,}000 \text{ L/mol}$$
$$\text{Cal} + \text{Na}^+ \rightarrow (\text{Cal. Na})^+ K_2 = 3.5 \text{ L/mol}.$$

Here Cal. Cs indicates that the complex is formed between calixarene molecule and Cs ion.

Calixarene is found to have an affinity for Cs ions that is 6570 times higher than that for the Na ions. We expect that this figure would also be the same for Rb ions, but this assumption needs to be verified experimentally.

Let us considering the reaction Cal. + Rb^+ → (Cal. $Rb)^+$.

Let $[\text{Cal.Rb}]/[\text{Rb}]_{\text{total}}$ be the rate of complexation. Then this rate can be calculated by solving the following system of equations:

$$K_1 = \frac{[\text{Cal.Rb}]}{[\text{Cal.}]_{\text{total}} - [\text{Cal.Rb}]) \cdot ([\text{Rb}]_{\text{total}} - [\text{Cal.Rb}])}$$

$$K_2 = \frac{[\text{Cal.Na}]}{[\text{Cal.}]_{\text{total}} - [\text{Cal.Na}]) \cdot ([\text{Na}]_{\text{total}} - [\text{Cal.Na}])}$$

The rate of complexation of Rb can be correlated to the rate of retention of the NF membrane by:

Rate of retention $R = 1 - (C_{\text{permeate}}/C_{\text{feed}})$

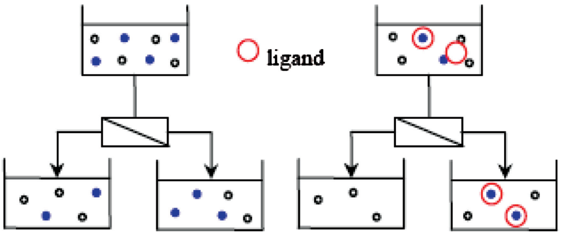

Fig. 7. Principle of complexation–NF.

We consider a brine solution with the following characteristics:

$$[\text{Rb}] = 5.6\text{E}^{-6} \text{ mol/L or } 0.48 \text{ mg/L}$$
$$[\text{Na}) = 1.8 \text{ mol/L}.$$

The results of calculations are presented in Table 4.

3.1. Use of diafiltration

Diafiltration is a modification of the ultra-filtration process in which water is added to the feed as filtration proceeds in order to wash out feed components that will pass through the membrane (Fig. 8).

This process can thus be applied to our case to further reduce the Na concentration without affecting the Rb concentration.

Ideally, the main purpose of a diafiltration stage (which may be preceded by a NF stage to further reduce the rates of flow) is to obtain the lowest possible brine concentration. However, the rate of retention of Rb is a function of the Na concentration.

The rate of retention of Rb would thus vary during the diafiltration operation. The state of the system can be determined by numerical integration using the following mass balance equations.

$$V \cdot \frac{dC_{\text{Rb}}}{dt} = -P \cdot (1 - \text{Re}) \cdot C_{\text{Rb}}$$

$$V \cdot \frac{dC_{\text{Na}}}{dt} = -P \cdot 0.95 \cdot C_{\text{Na}}$$

$$\text{Re} = f(C_{\text{Na}}, C_{\text{Rb}})$$

where V is the reservoir volume, P is the permeate flow rate and Re is the retention rate of Rb.

Table 4
Calculated amounts of calixarene for different reaction rates

Desired retention rate	0.6	0.7	0.8	0.9
Required calixarene concentration (mol/L)	0.0005	0.0007	0.0013	0.0029

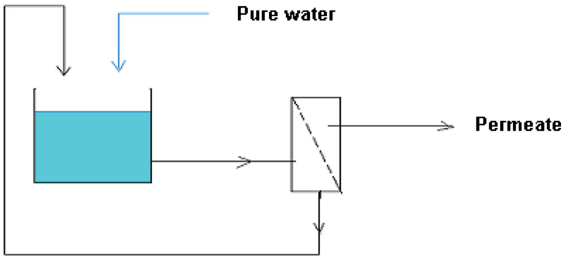

Fig. 8. Principle of diafiltration.

The Euler method and the SCILAB software were used to resolve the above system.

Results are presented in Figs. 7 and 8 for a total reservoir volume of 7560 m^3 and permeate flow rate of 1575 m^3/h.

It may be observed that at the end of 24 h, the Na concentration is reduced to 20 mol/m^3 from the initial 2500 mol/m^3.

Rb concentration also decreases but very slowly because of the high retention rate. At the end of the operation, there remains about 98% of Rb due to the less quantity of Na.

The above figures are obtained for a permeate flow rate of 1575 m^3/h and the required diafiltration surface of 7875 m^2 (Figs. 9 and 10).

At the end of diafiltration stage, the solution may be subjected to a reverse osmosis (RO) stage in order to further concentrate the Rb and obtain the pure water for the diafiltration stage.

In a final stage, the calixarenes can be precipitated by lowering the pH of the solution.

The combined NF–complexation protocol thus obtained is schematically represented in Fig. 11.

It should be underlined that the proposed protocol is still in the early conceptual stage. To begin with, all calculations have been made from the data available on Cs and not on Rb. Experiments need to be performed to verify this assumption and the precipitation of Rb through acid treatment.

Yet another drawback of the protocol is the rather high quantities of calixarenes required. Investigation on recycling the calixarenes should therefore be carried out to reduce the amounts and thus the costs involved.

4. Conclusions

Current demands for rubidium are relatively limited and are adequately met by available resources but certain experts believe that these demands may increase the view of novel applications in the future. It is the reason that investigations were undertaken to use the rejected brine by desalination systems to extract rubidium along with certain other metals present in the brine.

The above investigations have shown that whatever the method used, the extraction of Rb from the brine rejected by desalination plants would require large sized installations. The commercial interest of the rubidium thus extracted would thus strongly depend on

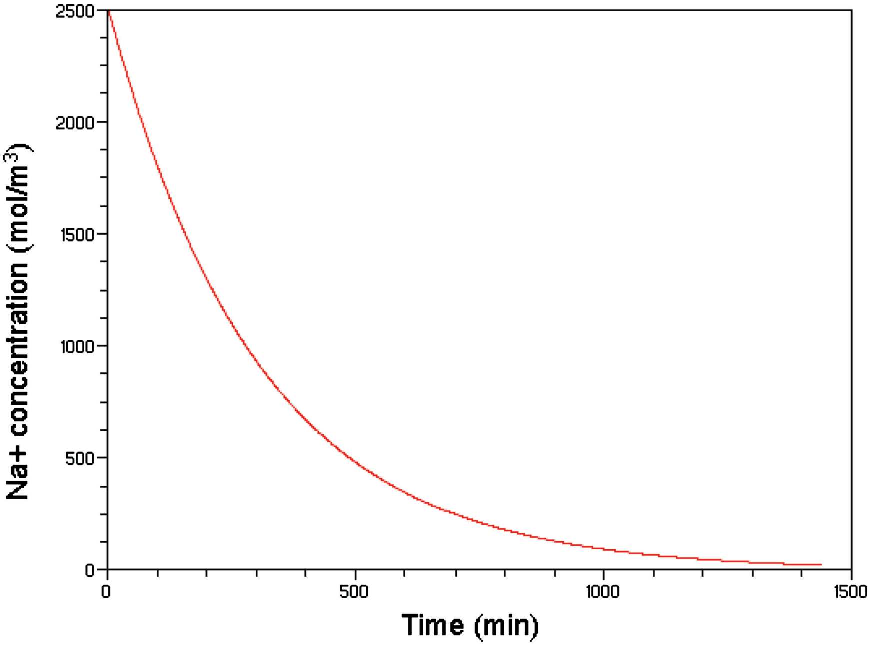

Fig. 9. Variation of Na^+ as a function of time in the diafiltration stage.

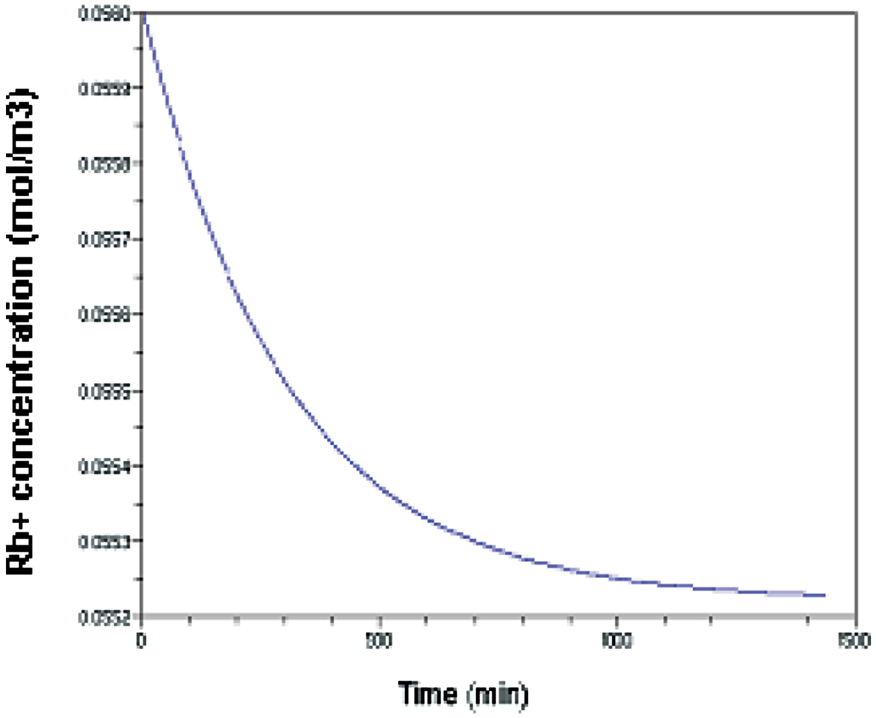

Fig. 10. Variation of Rb^+ as a function of time in the diafiltration stage.

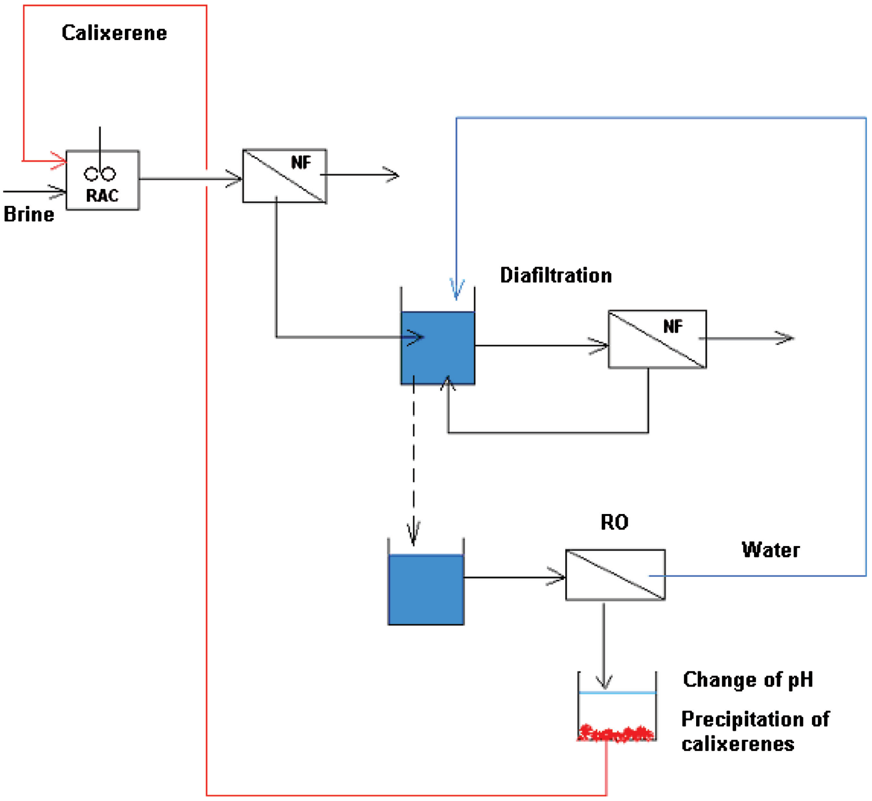

Fig. 11. Global protocol for Rb extraction by complexing and diafiltration–NF.

the costs that these installations and corresponding extraction protocols would entail.

After initial determination of various reaction constants, the optimal solution probably would comprise a mixture of several approaches e.g. initial membrane filtration for further concentrating the solution in the desired ion, followed by a passage on ion-exchange resins followed by complexation–diafiltration, etc. It must be underlined that such a combination of methods will have to incorporate the strategy of the reuse of the reagents used and their safe disposal, if needed in some cases.

We believe that for further investigations, it is necessary to:

- Determine the real adsorption isotherm for Rb extraction by the IE-911 resin in the presence of a highly saline medium.
- Determine the constants for the complexation of Rb by calixerene molecules.
- Determine the real costs of the protocol evolved.

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