



Application of reverse osmosis at NPP and verification of the process for primary coolant treatment in temelín nuclear power

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Abstract: Drained primary coolant from nuclear power plants containing boric acid is currently treated in the system of evaporators and by ion exchangers. Reverse osmosis as an alternative process to evaporator was investigated. Using reverse osmosis, the feed primary coolant is separated into two output streams: retentate and permeate. Retentate stream consists of concentrated boric acid solution together with other components, while permeate stream consists of purified water. In the first phase of the project the reverse osmosis modules from several manufactures were tested on a batch laboratory apparatus. Certain modifications to the pH of the feed solution were needed to enable the tested membranes to concentrate the H_3BO_3 in the retentate stream, separate from the pure water in the permeate stream. Furthermore, the separation capability for other compounds present in primary coolant such as K, Li or NH_3 were evaluated. In the final phase of the project the pilot-plant unit of reverse osmosis was tested in nuclear power plant Temelín. It was installed in the Special Purification System SVO-6 for the regeneration of boric acid. The aim of the tests performed in Temelín nuclear power plant was to verify possible use of reverse osmosis for the treatment of primary coolant.

Keywords: *Reverse osmosis, H_3BO_3 , primary coolant, nuclear power plants.*

I. INTRODUCTION

Demineralized light water is used as a coolant in primary circuit of nuclear power plants (NPP) with water-water energetic reactor (VVER type). Several chemicals are added into the coolant to adjust its chemistry: H_3BO_3 is added to control the core reactivity by absorption of neutrons, NH_3 is added as a source of H_2 to suppress negative effects of radiolytic decomposition of cooling water and KOH is added to adjust coolant pH. Furthermore, other compounds can be present in the coolant: Li is formed as a product of neutron absorption by boron and radioactive isotopes, such as ^{134}Cs and ^{137}Cs , are products of fission reaction.[1][2][3]

Drained primary coolant from nuclear power plants containing boric acid is currently

treated in the system of evaporators and by ion exchangers. Reverse osmosis (RO) was suggested as an alternative solution for the treatment of the drained primary coolant in nuclear power plants (NPP) instead of currently used system of evaporators. The advantages of usage of RO over traditional evaporator are its flexibility, mild temperatures of process fluids, quick start-up and shut-down time. Nowadays the evaporator remains still in operation thanks to the robustness of the process and long-term experience in this usage. However, as the membrane technologies have been implemented in various fields so they could be one day in the field of rad-waste processing and/or as a support technology in auxiliary systems of nuclear power plant (investigated in this work).

When using RO, a feed solution is separated on the semi-permeable membrane

into two output streams: retentate and permeate. The retentate contains majority of dissolved compounds while the permeate is almost clean solvent (water). Separation of individual species on RO unit is dependent on many factors [4][5], e.g. feed flow rate, recovery rate (ratio of the permeate flow rate to the feed flow rate) and pH of the feed solution, whose influence was tested within this work.

II. EXPERIMENTS

The whole experimental plan was divided into 3 phases:

1. Non-radioactive tests at lab-scale RO apparatus (RO module size 2540)
2. Non-radioactive tests at pilot-scale RO apparatus (RO module size 4040)
3. Radioactive tests at pilot-scale RO: Field test at NPP with primary coolant

1st Phase: lab-scale apparatus

The scheme of the laboratory apparatus used for testing of reverse osmosis membranes is depicted in Fig.1. It consists of a feed solution reservoir, pump (Hydra-Cell G03 series), heat exchanger and pressure vessel with a spiral wound module of reverse osmosis membrane. Various membranes were tested. The membranes of interest were polyamide composite reverse osmosis membranes commercially used for sea water desalination produced from three manufacturers, namely Ropur FR80-2540-S, Filmtec SW30-2540 and Hydranautics SWC-2540). One particular membrane was used only for few experiments (2–4 experiments), and for further experiments, a new identical membrane of the same type and

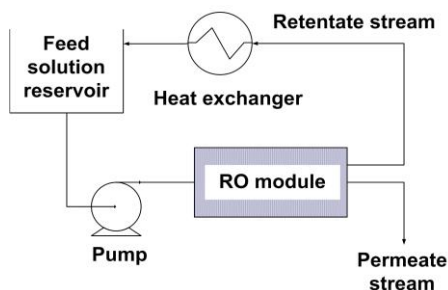


Fig.1. Flow diagram of lab-scale RO apparatus

manufacturer was used in order to minimize influence of membrane fouling and aging. All experiments were conducted in semi-batch mode when the retentate stream was recycled into the feed tank and permeate was drained.

2nd Phase: pilot plant

In the second phase, more sophisticated apparatus was developed. The apparatus was equipped with control unit for data acquisition (conductivity, flow rate, pH, pressure, temperature) and process control (flow rate, pH).

3rd Phase: testing at NPP

During the last (third phase) of the project the pilot-plant unit of RO was installed in Temelín NPP in auxiliary purification system (SVO-6) for impure boric acid solution treatment. Three tests were performed with real primary coolant in order to verify possible use of membrane system for the treatment of primary coolant under real conditions. The tests were focused on the separation efficiency species commonly occurring in the primary coolant, i.e. H_3BO_3 , NH_3 , K, Li and radioactivity (represented by total radioactivity and radioactivity and concentration of ^{134}Cs and ^{137}Cs isotopes). The aim was to obtain a concentrated H_3BO_3 solution together with other components of the primary coolant and radioactivity in the retentate stream and purified water in the permeate stream. Altogether, we performed three tests with real primary coolant of various composition (attention was focused mainly on various concentration of boric acid). Primary coolant pH was adjusted by KOH addition directly to the flow of the feed primary coolant. For each pH value, the flow of the feed primary coolant was set to 2000 and 3000 l/h and recovery (ratio of permeate flow rate to feed flow rate) to 10 and 15 %. Each experimental setup was stabilized for 20 minutes, after which samples for analysis were taken. Chemical analysis consisted of evaluation of conductivity and of H_3BO_3 , NH_3 , K and Li concentrations and radioanalysis consisted of evaluation of the total radioactivity and of the radioactivity and concentration of ^{134}Cs a ^{137}Cs isotopes.

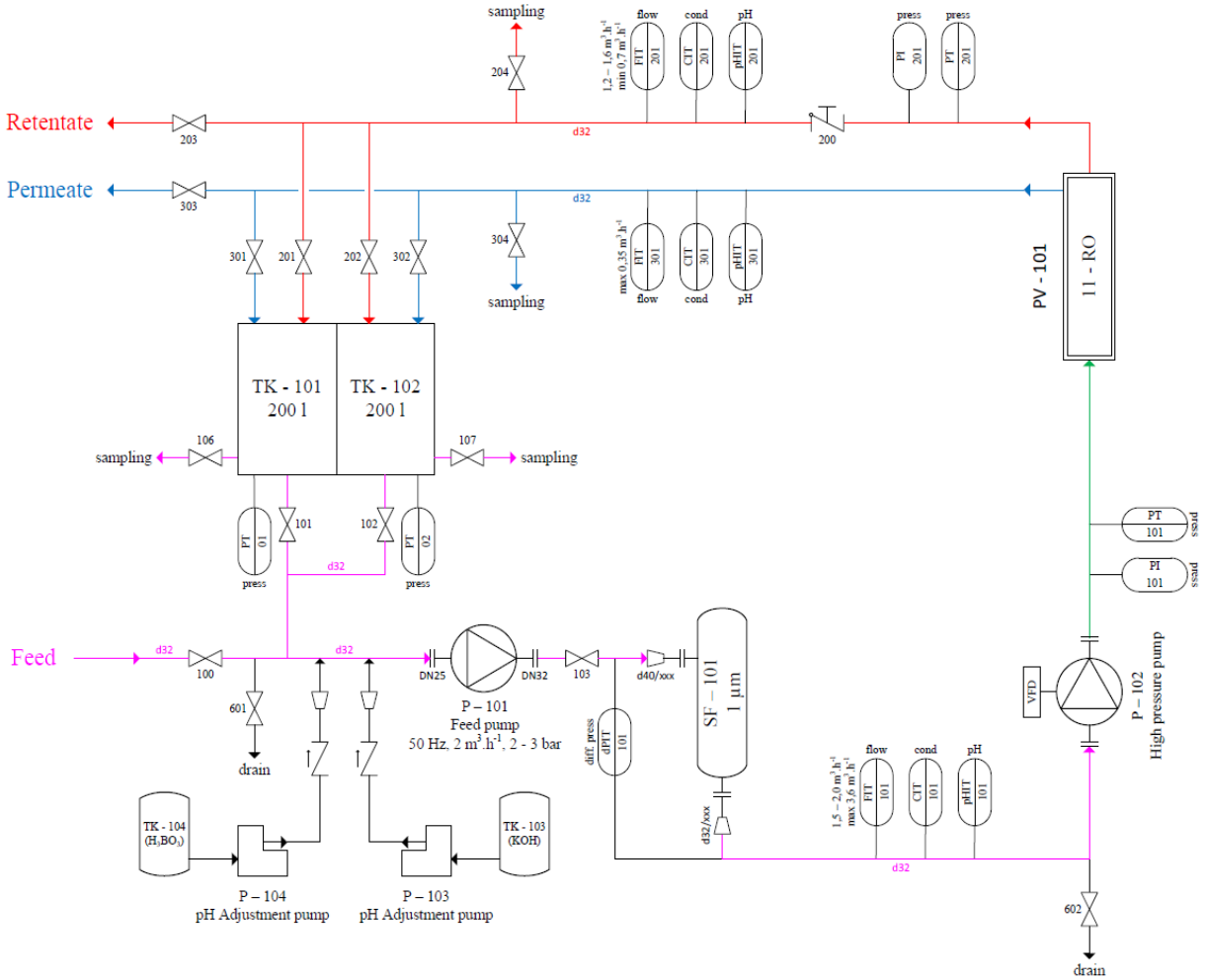


Table I. Composition of treated feed solution

	H ₃ BO ₃ , g/L	pH	Li, mg/L	K, mg/L	Radioactivity
1st phase	1.0-93.2*	4.6-12.2	0-43.8	3-40 510	none
2nd phase	0.6-13.7	5.6-11.2	0-7.9	5-8 780	none
3rd phase	2.2-9.4	6.8-11.1	0.1-0.2	6-4 338	1.5-11.7·10 ⁷ Bq/m ³

*exceeding boric acid solubility thanks to the high pH value

III. RESULTS

Rejection of boric acid

Rejection factor increases with increase of pH. The explanation of this phenomenon is

higher ratio of boric acid molecules form ions and build larger solvation layer. The amine groups in membrane's active layer are getting charged which helps to repulse ions of the same charge.

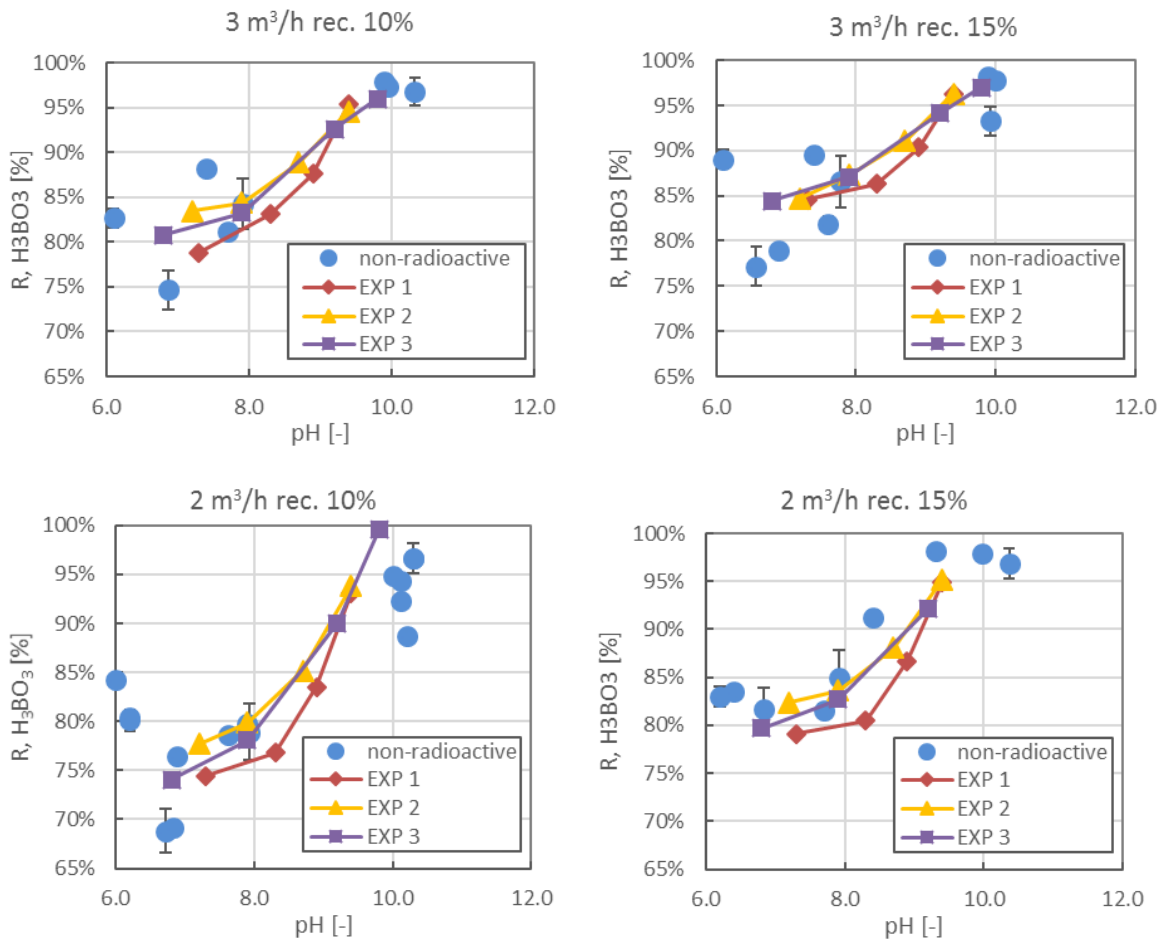
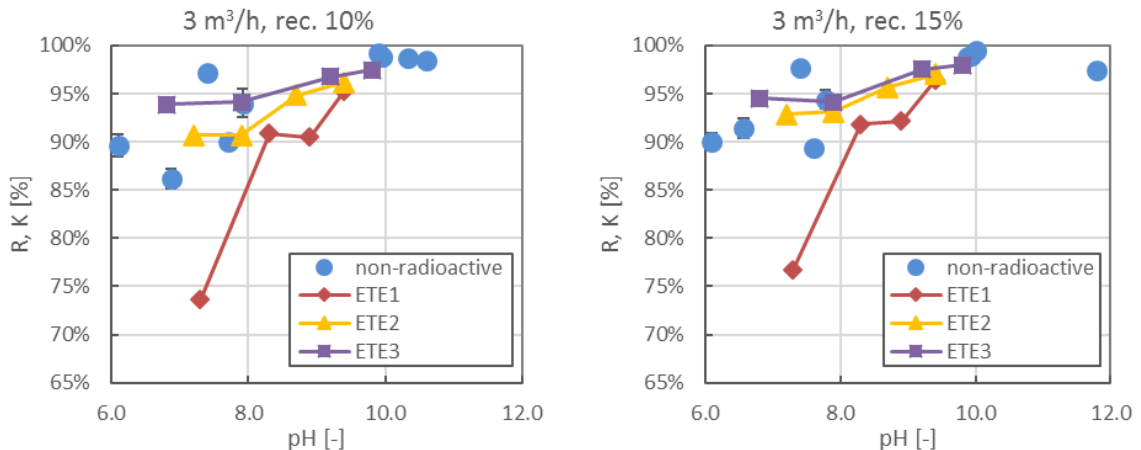


Fig. 2. Rejection factor of boric acid for different feed flows and recovery rates

Rejection of potassium

Comparison of the results obtained at nuclear plant with those obtained from experiments with non-radioactive model

solution shows that both anions and cations exhibit the same separation properties regardless radioactivity.



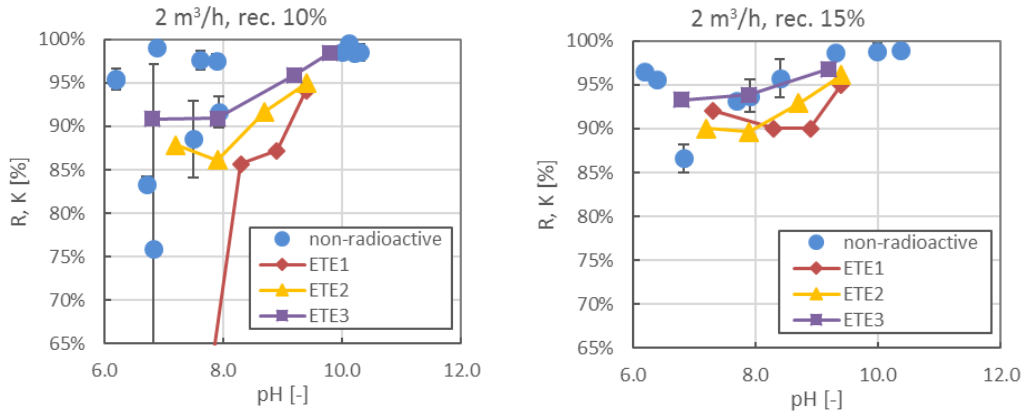


Fig.3. Rejection factor of potassium for different feed flows and recovery rates

Rejection of lithium

Rejection factor of cations increases from EXP 1 to EXP 3. Concentration of boric acid increases from EXP 1 to EXP 3 (Fig. 2). As stated in [6][7] K and Li rejection is pH independent, because steric dominated retention is the main mechanism attributing to the

generally high retention of both ions (>90 %). However due to nonideality of the membrane part of the boric acid passes (convective flux) as an anion so cations are being held by condition of electro neutrality on both sides of the membrane.

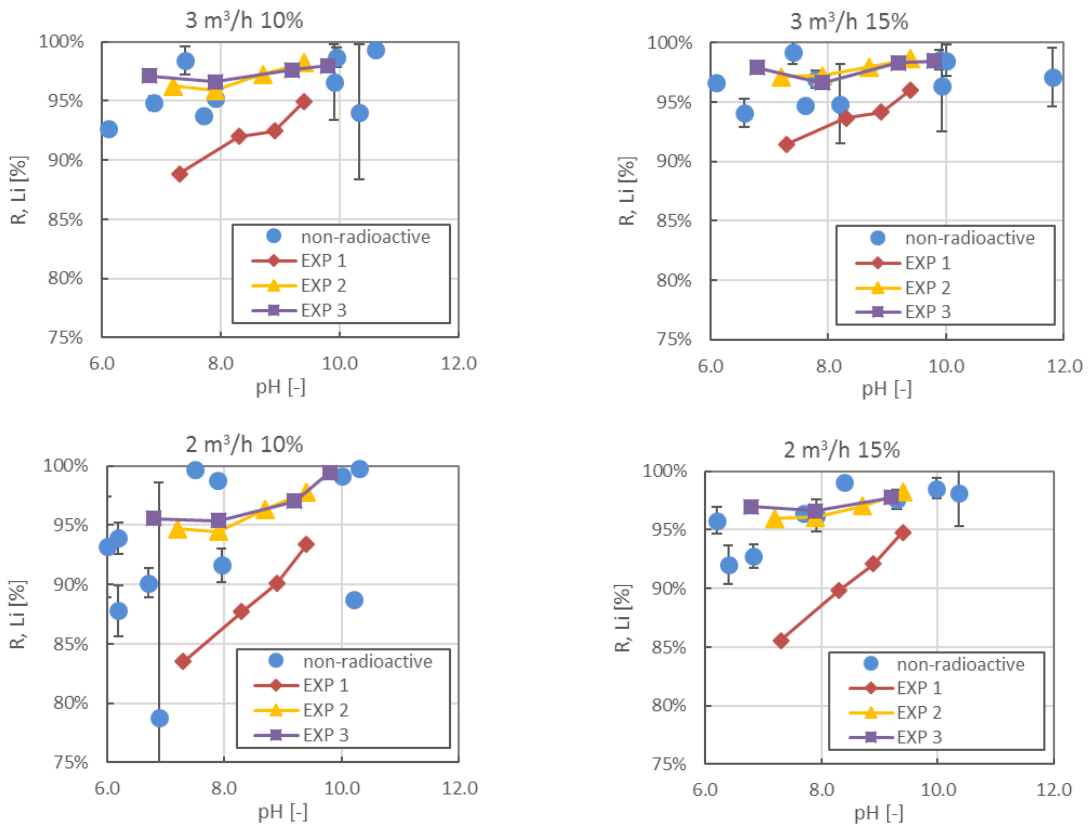


Fig.4. Rejection factor of lithium for different feed flows and recovery rates

Rejection caesium and decontamination factor

Rejection of radioactive isotopes, such as ^{134}Cs and ^{137}Cs , is generally rather high (80–100 %), but is dependent on pH: it is strongly enhanced by electrostatic interactions at low pH, while by Donnan’s effect at high pH [8].

Decontamination factor of total radioactivity is a ratio of the radioactivity in the feed and permeate stream, respectively [9]. Therefore the majority of radioactive isotopes in the solution are in form of cations the decontamination factor exhibits the same behavior as separation of cations.

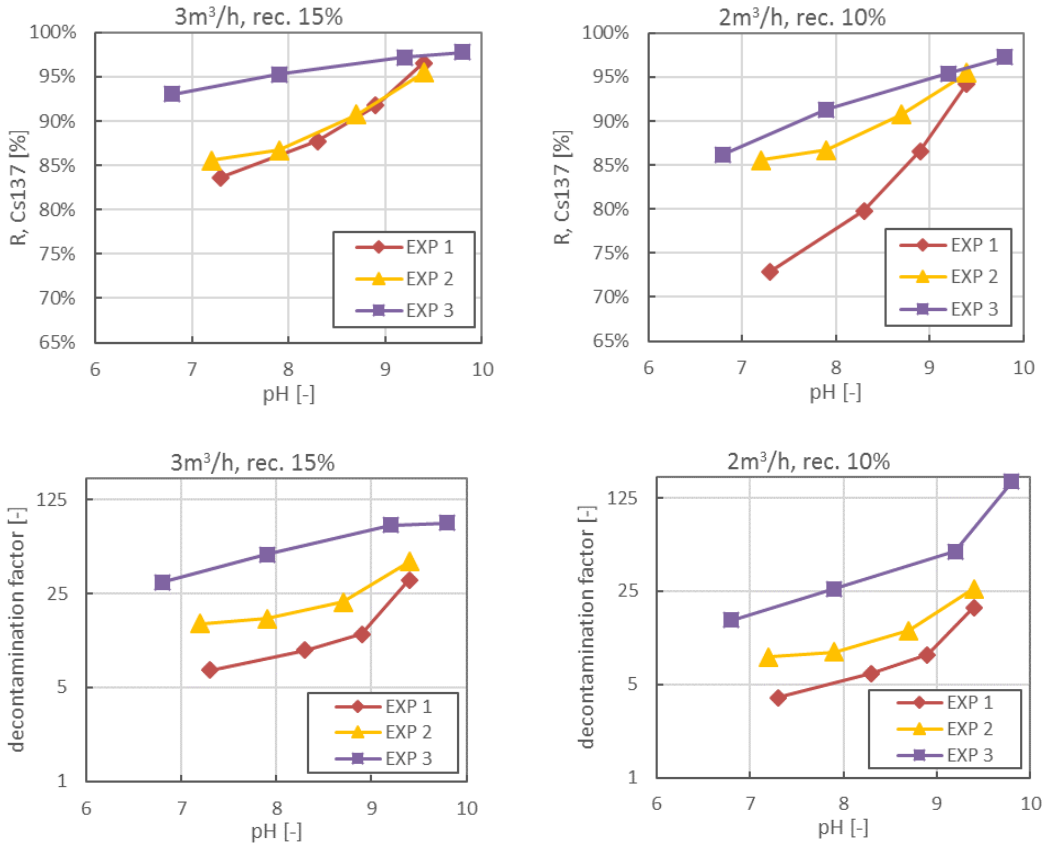


Fig. 5. Rejection factor of ^{137}Cs and decontamination factor for different feed flows and recovery rates

IV. CONCLUSIONS

	No. experiments	No. samples/ data points	Apparatus
1 st phase	38	900	Lab-scale
2 nd phase	33	650	Pilot-scale
3 rd phase	3	60	NPP tests

Table II. The project overview

Considerable number of experiments with model solution have been carried out on lab-scale and later pilot-scale apparatus. The separation ability for various compounds presented in primary coolant has been proven. The results confirmed that rejection of H_3BO_3 and NH_3 was strongly dependent on pH of the feed solution, while rejection of K and Li was high across the whole tested pH range. The experiments were performed with feed solutions containing various concentrations of H_3BO_3 in a range commonly occurring in primary coolant.

The pH of the feed solutions ranged from 4.6 to 12.2. Our results confirmed that the pH of the feed solution plays the most important role in membrane separation efficiency of H_3BO_3 .

In the final phase the pilot-plant unit of reverse osmosis (RO) was implemented into Special Purification System (SVO-6) in nuclear power plant (NPP) Temelín and its performance was studied during three experimental tests with real primary coolant. Three experimental tests with primary coolant of various composition and total radioactivity were performed across a wide range of feed primary coolant pH and with various total flows through the membrane modulus and recovery rates (ratio of permeate flow rate to feed flow rate). During the tests, samples of output streams were taken and analyzed for chemical parameters (pH, conductivity and H_3BO_3 , NH_3 , K and Li concentrations) and radioactivity parameters (total radioactivity and radioactivity and concentration of ^{134}Cs and ^{137}Cs isotopes). The tests were focused on the separation efficiency of basic components occurring in the primary coolant (H_3BO_3 , NH_3 , K and Li) and on the separation efficiency of radioactivity (total radioactivity and radioactivity and concentration of ^{134}Cs a ^{137}Cs isotopes) on the membrane modulus. Rejection of radioactivity exhibited the dependence on pH of the feed solution in the same way as H_3BO_3 , i.e. rejection of radioactivity increased as pH of the feed solution increased. The radioactive isotopes were trapped in the membrane modulus, mainly near its input, as well as in the mechanical filtered located ahead of the membrane modulus. However, the measured surface activity on these components was acceptably low.

Performing the tests with a pilot-plant RO unit directly in NPP Temelín with real primary coolant, we achieved a verified technology.

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