Design of treatment procedure for reprocessing of large volume of highly radioactive liquid waste after severe accident on VVER reactor

V. Brynych*[1], P. Franta[1], V. Havlová[1], L. Szatmáry[1], P. Sázavský[1], M. Gajdoš[2]

[1]ÚJV Řež, a. s.; Waste Management & Fuel Cycle Chemistry Division; Hlavní 130; 250 68 Husinec-Řež; Czech Republic

[2]Slovenské elektrárne, a.s.; Mlynské nivy 47; 821 09 Bratislava 2; Slovak Republic

*vojtech.brynych@ujv.cz

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Introduction

The apparatus designed for contaminated water treatment in the case of the reactor core damage on the VVER reactor type is being developed as an integral part of the implementation of measures after the Fukushima Daichi accident in VVER units operated Slovenské elektrárne, a.s. The volume of treated water is expected to be 10 000 m^3, presuming that the treatment of the contaminated water would start 6 months after the successful stabilization of the accident progression based on ERO response according to plant-specific SAMGs. The current scope of coolant volume limitation is related to adopted SAMG strategies for long-term removal of residual heat. However, the volume of treated water can be increased by scaling of the apparatus. The main presumed contaminants in the coolant solution are radioisotopes of caesium and strontium.

Decontamination apparatus

The basis of treatment apparatus was designed in the aftermath of Chernobyl accident in the UJV. The apparatus consist of several vessels with various sorbent materials, being related to the type of the radioisotope to be eliminated.

The decontamination apparatus have to fulfill following requirements:

- Decreasing the concentration of radionuclides to required limits or to levels necessary for reusing water for cooling of reactor.
- Vitrification of spent sorbent to the desired form for temporal or final storing.
- Decrease of specific radioactivity of gaseous emissions from decontamination apparatus to levels necessary for connection with air conditioning system of power plant.
- Decontaminated around 10 m^3 liquid waste every 24 hours.
- Equip the apparatus according regulations of work with nuclear waste - manipulators, shielding etc.
- The apparatus have to consists from several mobile modules so it can be fast assembled on required place. It have to be easy scalable if necessary.

Modules

The decontamination apparatus consists from three basic modules - sorption, vitrification modules and module for capturing of gaseous contaminants. Another necessary parts of apparatus are transporting and manipulation machinery, cooling, decontamination module and shielding. Sorption module consists from vessels with sorbent. Decontaminated water continue to control tank and if necessary the water can continue to alpha vessel. This vessel is utilized for decontamination of mainly alpha radionuclides by precipitation with ferrous hydroxide or hydrated titanium dioxide.

The water is filtrated and stored for standard treatment of liquid waste or it can be utilized for cooling of nuclear reactor.

The spent sorption vessels are vitrified in vitrification unit. The gaseous contaminants from vitrification units are captured and send back to first sorption vessel. The decontaminated gases are send to air condition system of power plant.

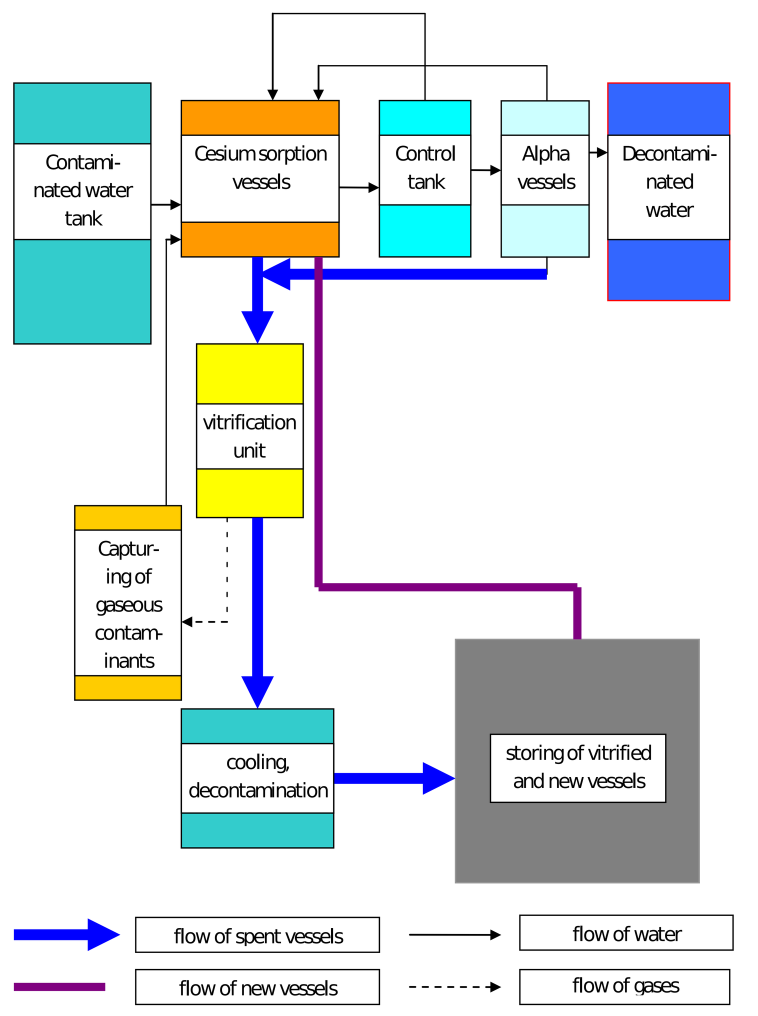

Figure 1: Decontamination and vitrification process[1]

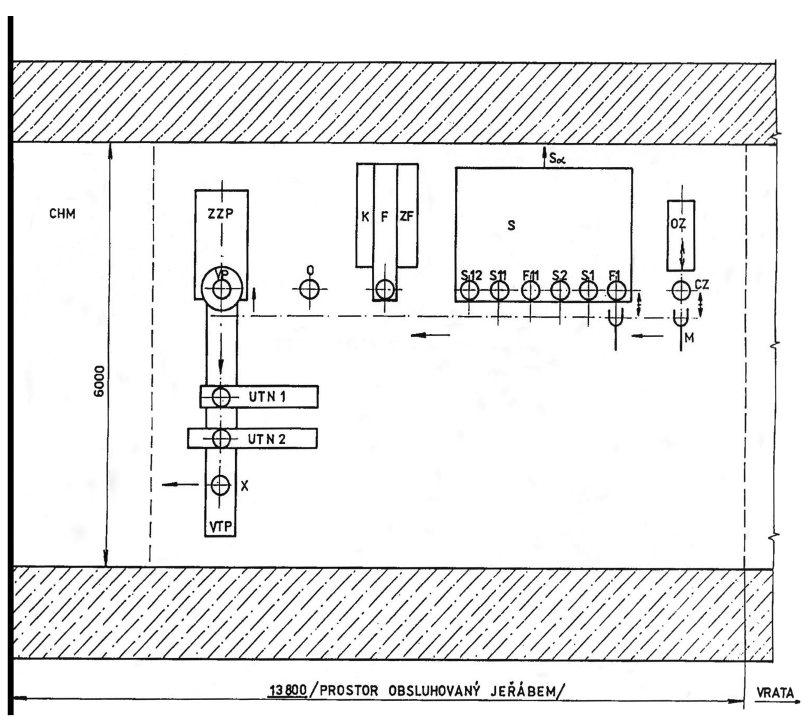

Figure 2: Layout of Decontamination apparatus[1]

CZ - entrance position of line
M - jaws of not marked manipulator
S - decontamination vessels
F1, F11 - places for vessels working as mechanical filters
S1, S2, S11, S12 - places for sorption vessels
K - system for condensation of emissions from vitrification unit
F - filters for capturing of emissions from vitrification unit
ZF - stack of filters
O - protruding place
VP - vitrification or drying unit
ZZP - manipulation system of vitrification unit
VTP - conveyor for transporting vessels from vitrification unit to closing machinery
UTN1 - machinery for closing vessels from vitrification unit
UTN2 - machinery for closing vessels after drying
CHM - cooling module
X - place for takeoff to cooling place
OZ - machinery for closing vessels
Note: alpha vessel is not marked on this layout

Source term

Source term describes the types, the quantities and chemical forms of radionuclides originated during accident. It depends on type of accident and the time after accident have to be taken into account. In this study it is expected that around 4 weight% of nuclear fuel will be released in the case of VVER reactor. The expected radioactivtiy of beta and gamma nuclides:

10^8-10^{10} Bq/l, total 10^{15}-10^{17} Bq/10 000 m^3

The expected radioactivtiy of alpha nuclides:

10^6-10^7 Bq/l, total 10^{13}-10^{14} Bq/10 000 m^3

The activity will decrease by one order after 6 month after accident.

nuclide	max. total activity [Bq]	%
^{89}Sr	$3.9\cdot10^{14}$	3.9
^{90}Sr	$3.2\cdot10^{14}$	3.2
^{91}Y	$2\cdot10^{13}$	0.2
^{95}Zr	$3\cdot10^{13}$	0.3
^{134}Cs	$5.04\cdot10^{15}$	50.4
^{137}Cs	$3.98\cdot10^{15}$	39.8
^{103}Ru	$3\cdot10^{13}$	0.3
^{106}Ru	$7\cdot10^{13}$	0.7
^{144}Ce	$9\cdot10^{13}$	0.9
^{147}Pm	$1\cdot10^{13}$	0.1
total:	$9.98\cdot10^{15}$	99.8

Table 1: Summary of the major radionuclides in contaminated water 6 months after accident[1]

Sorption vessels

The vessels are utilized for both sorption and vitrification, therefore the sorbent is mixed with glass substrate before sorption process. Thus, it can be vitrified after sorption without further manipulation with the sorbent, containing high concentration of caesium or other separated radionuclide.

There were designed two volumes of the sorption vessel - 25 and 50 liter. The vessel has integrated tube in the way which allow flow of the water going from from the top to the bottom where it continue through sorbent back to the top and to the next vessel. This design prevents the leaking of radioactive water outside of vessel even if tube rupture.

The various types inorganic sorbent were tested in the past, in order to determine sorption properties. The sorption experiments were performed in the solution containing H_3BO_3 and NaOH or KOH to model presumed waste solution originated during severe accident. The zeolites sorbents demonstrated the best sorption properties for caesium. As best type for sorption of caesium was determined mordenite. The klinoptilolit has best sorption properties for strontium. The capabilities of these zeolites can be improved by the modification with hexacyanoferrate for caesium or by the modification with titanium for strontium.

Leaching of concrete in Boric acid

In order to determine the contaminated solution composition, including radionuclides and additional components, experimental research focused on concrete leaching was performed. It is expected that the part of the concrete may be exposed to the contaminated water in the reactor or adjacent buildings due to possible cracks in steel liner. The content of concrete components leached out in the boric acid solution can influence the efficiency of sorbents in the process of radionuclide content reduction. To perform the initial quantification of this phenomenon, the small cube of concrete was placed inside the plastic bottle with a solution of H_3BO_3 and NaOH. The plastic bottle was stored inside the thermostatic chamber keeping the temperature equal to 90 °C. After one month the concrete cube was removed and the selected elements in the solution were measured by Atomic Absorption Spectroscopy (AAS). The concentration of calcium, potassium, sodium and magnesium were determined and the concentration of H_3BO_3 was measured by titration. The results were compared with the composition of the original solution. The concentration of calcium and potassium in solution was increased. The concentration of magnesium stays practically the same as in the original solution. On the other hand, the concentration of sodium and H_3BO_3 in solution was decreased.

	1 month				3 month		
	A0	A1	A2	A5	B0	B3	B6
m(g)	-	203,5	206,9	231,2	-	188,3	184,3
S/L (g/ml)	-	0,68	0,69	0,77	-	0,63	0,61
H_3BO_3 (g/l)	15,7	11,9	11,9	11,8	15,7	8,4	8,5
Ca (mg/l)	1,2	164,5	185,8	179,3	2,1	210,1	190,8
Mg (mg/l)	0,017	0,072	0,057	0,079	0,021	0,055	0,077
K (mg/l)	1,1	136	124	136,7	1,6	170,3	188,3
Na (mg/l)	1110,7	1010,3	1052,3	1003,7	1453,3	1238	1180,7

Table 2: The change of cations and H_3BO_3 concentrations after 1 and 3 months of concrete leaching in solution NaOH 1.9 g/l and H_3BO_3 15 g/l, temperature 90 °C. Samples A0 and B0 are blank experiments without presence of concrete[1]

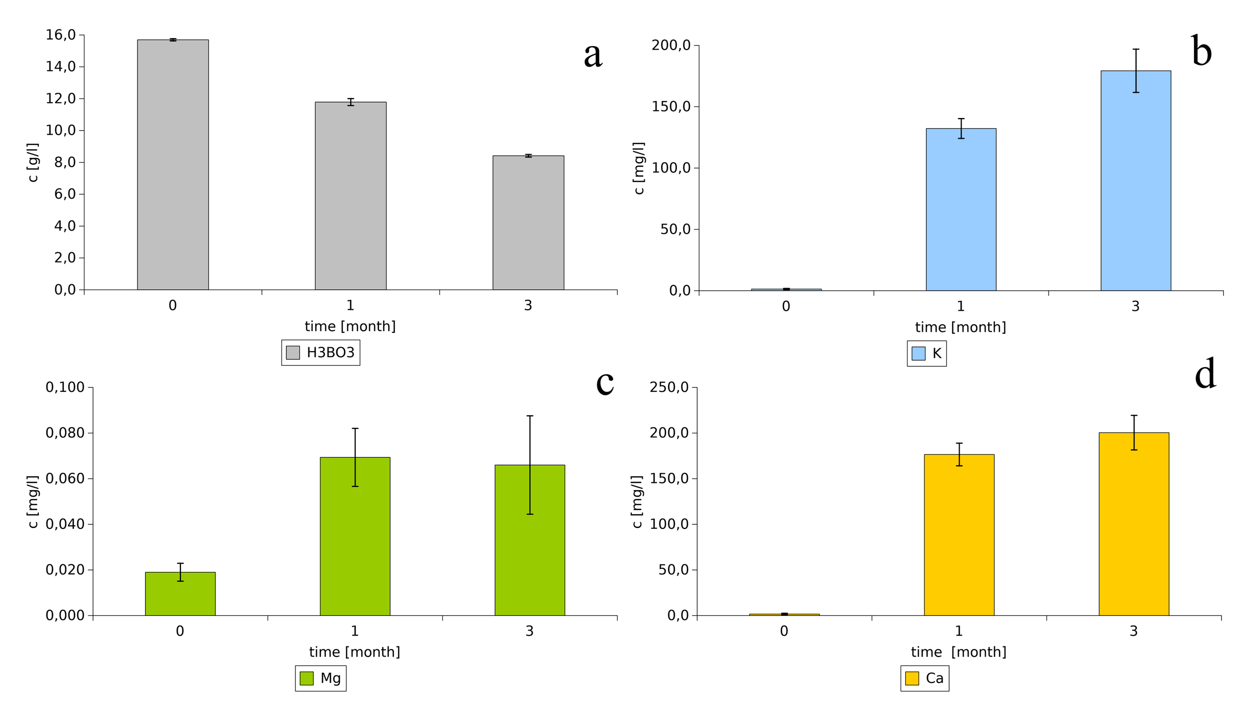

Figure 3: The change of cations and H_3BO_3 concentrations after 1 and 3 months of concrete leaching in solution NaOH 1.9 g/l and H_3BO_3 15 g/l, temperature 90 °C; a)Boric acid; b) Potassium; c) Magnesium; d) Calcium[1]

Conclusions

After Fukushima Daichi accident the demand for preparation of post-accident measures has arose. One of the main challenges is related to the highly radioactive waste waters reprocessing that are generated during the severe accident mitigation. Therefore UJV Řež in the cooperation with Slovenské elektrárne, a.s. (an ENEL group company) has started to review the concept and design of radioactive contaminated water treatment apparatus in the case of the VVER 440 NPP severe accident, that would be including new experiences and requirements.

UJV Řež has long lasting experience with the treatment of the various types of the radioactive wastes, including liquid ones. The development of design of the apparatus, used for liquid radioactive waste reduction, had started in the aftermath of Chernobyl accident. The results from the past were nowadays reconsidered and extended with the new information, including Fukushima Daichi experience and short term experimental research. Based on the knowledge and experience gained, a treatment procedure together with an apparatus was designed to meet the specific conditions of VVER reactors.

References

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