PRELIMINARY RADIOLOGICAL CHARACTERISATION OF THE MAIN CIRCULATION CIRCUIT AT IGNALINA NPP FOR DECOMMISSIONING PURPOSES

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ABSTRACT

In this paper the data on a preliminary modeling of the main circulation circuit (MCC) equipment contamination at the reactor final shutdown and the analysis of the contamination variation with time are presented for RBMK-1500 reactor Unit 1 at Ignalina NPP. In order to perform this task the data about MCC elements characteristics, the coolant characteristics, the system operational conditions, etc. were collected. The modified computer code LLWAA-DECOM was used to determine contamination of the system elements. As a result nuclide vectors and surface dose rates of deposits for every MCC element were determined. Modeling results were compared to the dose rates measurement results.

The analysis shows that the deposits mostly consist of ^{55}Fe, ^{59}Fe, ^{58}Co, ^{60}Co, ^{54}Mn and ^{63}Ni. From the radiological standpoint during dismantling the important nuclides are nuclides, which have influence on the equipment dose rate. γ emission from contaminated MCC elements at reactor final shutdown is mostly determined by ^{59}Fe and ^{54}Mn, but after 5 years – by ^{60}Co. With time the percentage of the γ emission from ^{94}Nb and ^{137}Cs nuclides is increasing. In the period of 35 years after the final reactor shutdown the contamination of MCC elements is mainly defined by the long-lived nuclide ^{63}Ni, because short-lived nuclides (^{55}Fe, ^{59}Fe, ^{58}Co, ^{60}Co, ^{54}Mn) decay during this period of time.

Analysis of MCC elements total contact dose rate variation shows that dose rate decreases during the period of 5 years significantly (up to 80 %). This is a result of the short- lived nuclides ^{59}Fe, ^{58}Co and ^{54}Mn spontaneous decay. Such decrease of equipment dose rate during the period of 5 years after final reactor shutdown is very important and indicates that dismantling of MCC shouldn't be started immediately after final shutdown.

For validation of the calculations measurements of the equipment dose rates have been performed. A comparison showed that the predicted dose rates were in rather good agreement with measured values or slightly exceed the highest measured values.

INTRODUCTION

There is only one nuclear power plant in Lithuania – Ignalina NPP (INPP). It operates two similar units with power rating of 1500 MW(e) and present power level of about 1250 MW(e) each. They were commissioned (first grid connection) in 12/1983 and 08/1987 respectively and provide approximately 70-80 per cent of the electricity produced in Lithuania. The original design lifetime was projected out to 2010-2015. October 10, 2002 Seimas (Lithuanian Parliament) approved updated National Energy strategy where it is indicated that the first Unit will be shutdown before the year 2005 and second Unit in 2009 if funding for decommissioning is available from EU and other donors. November 26, 2002, Lithuanian government approved immediate dismantling strategy for Unit 1.

Nuclear power plant decommissioning is long lasting, expensive and complicated procedure. Preparation for this process lasts few years and it means a preparation for safe dismantling, for safe decommissioning waste management, etc. In order to plan ahead dismantling works, to implement the radioactive waste treatment and conditioning technologies, storage facilities and repositories, it is

necessary to know the preliminary amounts, radioactivity levels and activity inventory of future decommissioning waste 1.

During NPP operation the materials in the reactor core are irradiated by neutrons, and the other systems, such as a main circulation circuit (MCC), purification and cooling system, spent nuclear fuel storage pools, etc. are contaminated due to circulation of contaminated coolant. During the circulation the radionuclides from coolant deposit on the inner surface of the system equipment.

The buildup of activity in the circulating coolant, on the coolant piping inner surfaces and on the core surfaces does pose problems to the reactor operation and maintenance, and during decommissioning. On one hand these problems are closely linked with the coolant purification, use of filters and corrosion inhibitors and on the other hand these are related to any rise in coolant temperatures, change in flow rates and perturbations in neutron flux. Various studies have shown that water becomes very corrosive at the high temperatures and pressures. The decomposition of water by radiation increases its corrosive nature. The corrosion products may originate as soluble and insoluble oxides, or in other particulate forms. The rate of corrosion in the reactor primary system keeps on increasing as the time of reactor operation at full power increases

In pressurized water reactors (PWRs) the corrosion product activity is primarily due to short-lived ^{56}Mn and ^{24}Na, and nearly all the longer-lived activity in the coolant is due to iron, molybdenum and cobalt with most significant radionuclides as ^{59}Fe, ^{99}Mo and ^{60}Co 2. The neutron activation of structural ^{27}Al and activation of ^{23}Na from salt impurities in water can produce ^{24}Na. The use of high-purity water, demineralization of water and the presence of filters keep the amount of dissolved salts to less than 0.05 ppm 3. Operating parameters of the reactor also strongly affect the types of radionuclides formed, the levels of saturation activity reached and the rates at which the saturation is reached. These include the composition of the materials in contact with the coolant, amount and the types of the impurities present in the coolant, reactor power, residence time of coolant in core, temperatures and pressure, coolant flow rates, corrosion rates, filter efficiency and deposition rates of radioactive elements in coolant. During the past ten years, a series of experimental and numerical studies were done on effects of flow rate (fast and slow transients) on dose rates due to corrosion activity in coolant of PWRs. Power perturbations remained in focus also. Brief review of these activities and new data on the joint effect of the accelerating corrosion rates (corrosion rate does increase slowly with plant aging; it also increases with temperature and pressure) and different types of flow rate perturbations are presented in 2

Recently, accumulation of radioactive corrosion products on steel surfaces of VVER type nuclear reactors has been studded experimentally. These were laboratory scale studies for ^{110m}Ag and ^{60}Co buildup on steel surfaces (austenitic stainless steel type 08X18H10T and magnetite-covered carbon steel often to be used in Soviet-panned VVERs) 4,5. It is indicated in these studies that the most important corrosion product radionuclides in the primary coolant of PWRs are ^{60}Co, ^{58}Co, ^{54}Cr, ^{59}Mn as well as ^{110m}Ag in some Soviet-made VVER-type reactors.

Experimental investigations demonstrate that the main radionuclides in the primary coolant of RBMK-1000 reactors are ^{59}Fe, ^{54}Mn, ^{56}Mn, ^{51}Cr, ^{58}Co, ^{60}Co, ^{95}Zr and others 7,8,9. The quantities of the nuclides in the reactor water depend on the reactor regimes and removal by filters. Detailed measurements of the impact of the different nuclides to the effective dose rate depending on the operation time were performed in 8. With time the activity of the nuclides in the reactor water is increasing and depending on the decay constant reaches the stabilization 8,9. The activity of the rather short-lived nuclide ^{56}Mn in the primary coolant is changing in similar way as power rate of the reactor is changing, ^{51}Cr, ^{59}Fe – are reaching the stabilization after half of the year, ^{58}Co, ^{54}Mn, – after one and a half year. Activity of ^{60}Co is increasing permanently and the stabilization can appear only close to the operation lifetime of the

NPP. On the basis of the performed experimental investigations and regularities revealed few calculation models have been developed 6,9,10.

In this paper a preliminary assessment of MCC equipment contamination for RBMK-1500 Unit 1 at Ignalina NPP is presented. First of all information necessary for such assessment was collected: characteristics of the MCC elements, characteristics of the coolant, operation conditions, etc. The modified computer code LLWAA-DECOM (Belgium) was used for determination of the contamination of the elements. As a result nuclide vectors and dose rates were determined for every element. Modeling results were compared to the measurements of the dose rates.

MAIN CIRCULATION CIRCUIT (MCC)

The main purpose of MCC is to supply water to the fuel channels, where it is converted to the steam-water mixture due to nuclear reactions heat generation, and to direct this mixture to the turbines. The main MCC elements are presented in Table 1.

Table 1 Main MCC elements

No	System element	Number per unit	Mark

1.	Separator drum:*
	     Liquid phase
	     Steam phase
		4	MCC 01L, MCC 01S

2.	Bypass between separator drums	4	MCC 02
3.	Downcomers (not insulated)	48	MCC 03
4.	Downcomers (insulated)	48	MCC 04
5.	Suction header	2	MCC 05
6.	MCP suction pipes	8	MCC 06
7.	MCP tanks	8	MCC 07
8.	MCP pressure pipes	8	MCC 08
9.	Bypass between headers	12	MCC 09
10.	Pressure header	2	MCC 10
11.	Group distribution headers	40	MCC 11
12.	Bottom water pipes	1661	MCC 12
13.	Fuel channels, below the core	1661	MCC 13
14.	Fuel channels, within the core	1661	MCC 14
15.	Fuel channels, above the core	1661	MCC 15
16.	Steam- water pipes	1661	MCC 16

*About 77% of separator drum inner surface is in contact with coolant, and the rest 23% is in contact with steam.

MCC consists of two loops, whose components are arranged symmetrically with respect to the vertical axis of the reactor. Each loop has two separator drums (1), which separate steam from steam-water mixture exiting from the fuel channels in core. Separator drums are interconnected both in the lower, liquid filled, and upper, steam filled, levels by pipes (2). Water separated from steam is directed to the suction header (5) by 24 bypass pipes (3, 4), then to the four main circulation pumps (7). During normal reactor operation three pumps are operating in each loop, the forth pump is in reserve. These pumps are of a vertical, centrifugal, single-stage configuration. Nominal efficiency of the pump is 8500 m3/h, and pressure is 1,96 MPa.

From main circulation pumps water flows through pressure header pipes (8) to the pressure header (10). Suction and pressure headers are connected between itself by six bypass pipes (9). Each of these pipes

has a gate and check valve. Mentioned bypass pipes ensure the natural circulation of the coolant in case the main circulation pumps are shut down.

From pressure header (10) water is supplied to twelve group distribution headers (11) by twelve pipes. Each group of distribution headers is connected to 40 – 43 bottom water pipes (13) leading to fuel channels. Flow rate in each pipe and, therefore in each fuel channel (14), is set by control valves. Steam separated from steam-water mixture generated in the reactor core is directed to the turbines by steam pipelines.

CONTAMINATION ASSESSMENT METHODOLOGY

Contamination is of two general types: loose contamination capable of being removed by simple mechanical means or fixed contamination requiring more aggressive removal methods. Contamination deposited on internal and external surfaces of the plant is due to the transport and/or leach out of activated corrosion and erosion products or fission products and actinides. Contamination generally accumulates on the facility and equipment surfaces and does not (except in concrete) penetrate very deeply.

For the assessment of MCC equipment radioactive contamination the modified computer code "LLWAA – Decom" (Belgian) was used 11. Mentioned code allows to determine the activity (Bq/m^2) of the deposits located on the system equipment inner surface taking into account the coolant specific activity (Bq/m^3) and the constructional data of system elements (constructional materials, geometrical measurements, etc.).

Deposits activity assessment is based on the following equation for each system element:

$$\frac{dW_i}{dt} = K_d * Cv_i * (1 - frspr_i) - W_i * (K_r + \lambda_i)$$
(Eq. 1)

Here W_i - activity of i type nuclide deposited on equipment surface, Bq/m^2;

K_d - deposition rate, m/s;

Cv_i - i type nuclide (specific) activity in the MCC fluid, Bq/m^3;

$frspr_i$ - fraction of i type nuclide specific activity in soluble form;

K_r - release rate coefficient , s^{-1}; λ- i type nuclide decay rate, s^{-1}; t - time, s.

Deposition rate and release rate coefficients are the functions of fluid characteristics (velocity, temperature, Reynolds number), the system equipment characteristics (geometry, inner walls roughness, friction factors), the characteristics or radioactive particles (its specific weight, diameter).

As it was mentioned above equipment contamination is concentrated in the surface layer. Contamination occurs due the contact with contaminated coolant. Only the fuel channels (MCC elements) located in the reactor core are contaminated mostly due the activation process in the core.

The dose rates from different nuclides in reactor water and nuclides deposited on the inner wall of the elements have been determined also. Calculations show that dose rate from MCC fluid is much smaller than dose rate from deposits. Coolant will be removed before the dismantling process. But there is an opportunity to compare predicted and measured dose rates even for the systems filled with the coolant.

Activation products appear in MCC water by two different ways 12. First one is that during operation of a nuclear reactor, most metallic surfaces oxidize and form a layer of corrosion film. This layer, represented by oxides of structural elements, is exposed to high pressures and temperatures. It erodes and, together with coolant is transported to the area of high neutron flux within the reactor core. Here, the oxides may be deposited and then activated by neutrons to form activation products and, following erosion, circulate throughout the reactor system and be deposited on inner surfaces. The other way is erosion of neutron-activated material. In order to assess the activity of activation products (determined by ORIGEN code) in MCC the parameters such as equipment material characteristics, surface area, corrosion rates, fluid purification rate, should be determined. ^{14}C is the activation product also. Activation of oxygen is dominant reaction in MCC water and as a result MCC water is contaminated by ^{14}C nuclide. Total MCC contamination (activity) is a result of all mentioned processes.

RESULTS

Nuclide inventory

The analysis shows that the deposits mostly consist of ^{55}Fe, ^{59}Fe, ^{60}Co, ^{54}Mn, ^{63}Ni, and ^{58}Co. The nuclides mentioned above are a consequence of the MCC equipment material activation and corrosion processes. As it was already indicated, deposition of the radioactive particles onto equipment inner surface depends on coolant characteristics (pH, temperature), which have influence on corrosion rate, and system operational conditions (characteristics) (MCC purification rate, length of operation cycle). The concentration of radionuclides in the deposits depends on its concentration and solubility in the MCC water (coolant).

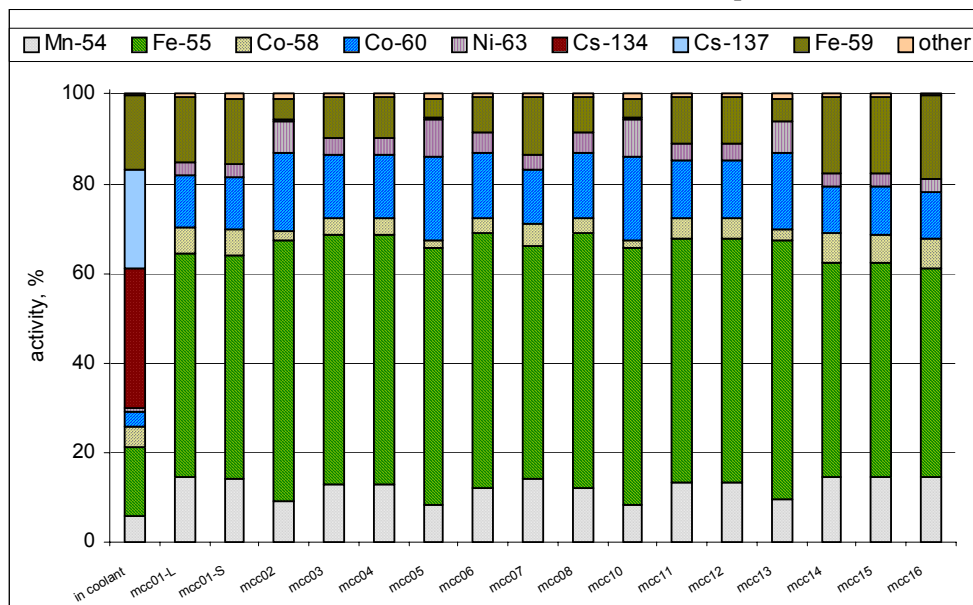Nuclide inventories in the RBMK-1500 reactor coolant and in the deposits are shown in Figure 1.

Fig. 1   Nuclide inventory of the reactor coolant and the deposits of each system element. The "other" means other nuclides, which influence on the total contamination does not exceed 1 %.

As can be seen, ^{134}Cs and ^{137}Cs are dominating nuclides in the coolant (31.04 % and 22.17 % respectively). Whereas the dominant nuclides in deposits are ^{55}Fe (46.61 % - 57.18 %), ^{60}Co (10.39 % –

18.69 %), ^{54}Mn (8.33 % – 14.46 %), ^{59}Fe (4.11 % – 18.56 %), although it's share in the coolant is 15,52 %, 3.32 %, 5.6 %, 16.63 % respectively.

Such nuclide distribution in the deposits depends on nuclide concentration and their solubility in the coolant. Only insoluble fraction of the nuclides deposits on the surface. In case of ^{134}Cs and ^{137}Cs the nuclide volumetric concentrations in the coolant are similar or even larger than ^{54}Mn concentration, but in the deposits there is larger part of ^{54}Mn nuclide than ^{134}Cs or ^{137}Cs nuclides. This is a result of higher solubility of Cs isotopes. Nuclide transition to the deposits layer is a complex process and depends on the chemistry of deposits formation.

Dose rates

Analysis of the data on MCC contaminated equipment dose rate at the time of reactor shutdown shows that the separator drums surface in contact with water, MCC pumps (7), group distribution header (11), fuel channels located in the rectors core (14), steam – water pipes (16) are the most contaminated elements. MCC equipment contamination changes due to spontaneous nuclide decays and Fig. 2 presents the dynamics in time of the nuclide inventory for the equipment.

For the period, analyzed the long-lived nuclide ^{63}Ni starts dominate because of the decay of the short-lived nuclides (^{55}Fe, ^{59}Fe, ^{58}Co, ^{60}Co, ^{54}Mn). The contribution of the long-lived nuclides to the total contamination increase as its activity practically remains unchanged. An exception is ^{241}Am, which activity increases as a consequence of ^{241}Pu (T$_{1/2}$ = 14.35 y) decay.
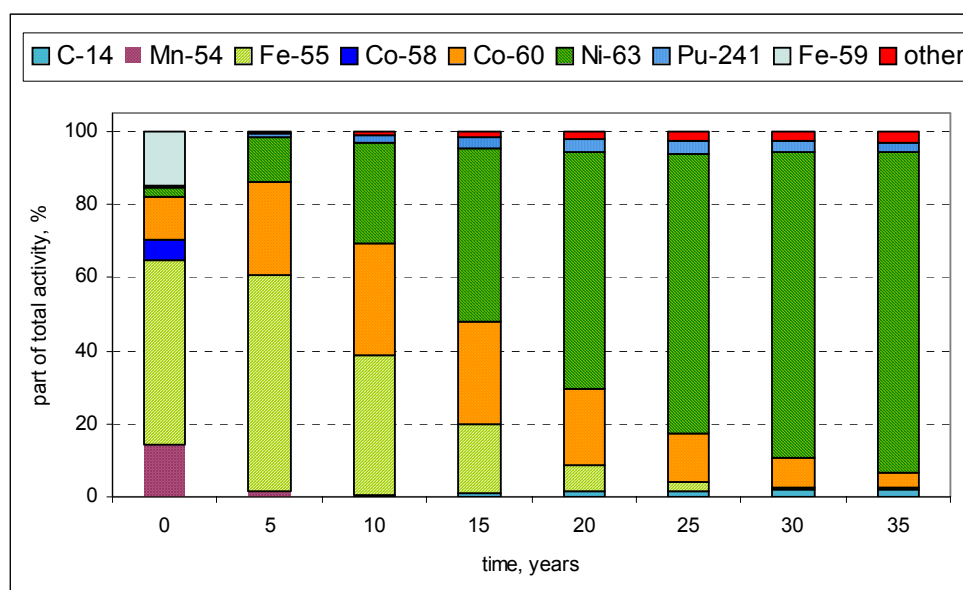

Fig. 2 Activity variation during the period of 35 years. "Other" – nuclide, which activity does not exceed 0,12 % of total activity

As it was already mentioned ^{63}Ni is weak β- emitter and its fission product is ^{63}Cu in ground state. Thus ^{63}Ni decay is not followed by γ emission. So it does not contribute to the contact dose rate. From the radiological standpoint during dismantling the important nuclides are nuclides which have influence on the equipment dose rate, especially ^{60}Co. Equipment dose rate decrease during analyzed period, and the importance of the nuclide influencing dose rate changes also (Fig. 3).
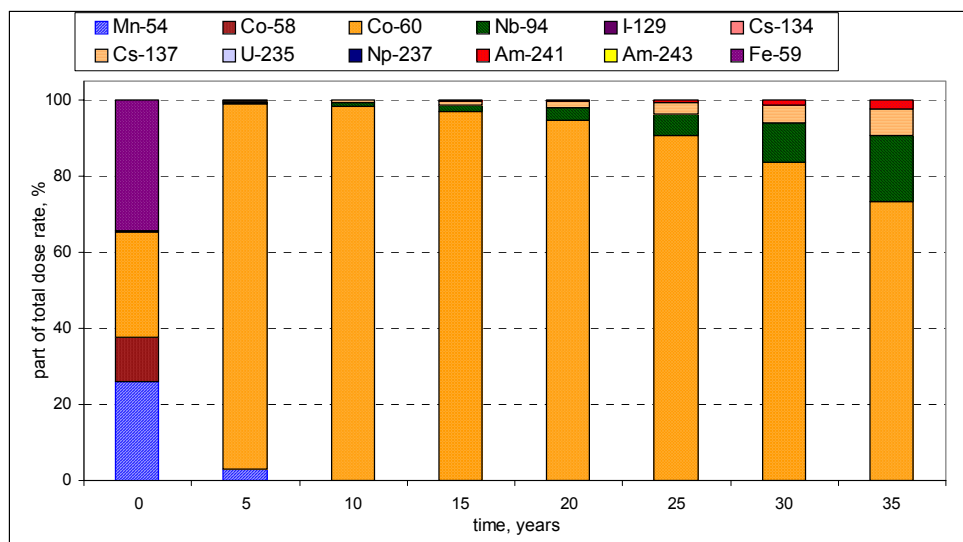
Fig. 3 The influence of each nuclide to the total dose rate

Analysis of MCC elements total contact dose rate variation shows that dose rate decreases during the period of 5 years significantly (up to 80 %) (Fig. 4).
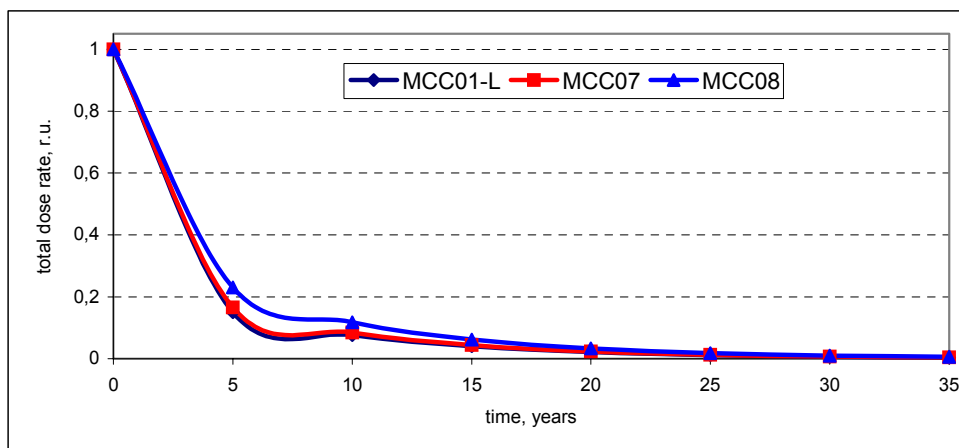

Fig. 4   Variation of the total dose rate for several MCC elements during the period
          of 35 years

This is a result of the short-lived nuclides ^{59}Fe, ^{58}Co and ^{54}Mn spontaneous decay. Such decrease of equipment dose rate during the period of 5 years after final reactor shutdown is very important and indicates that dismantling of MCC shouldn't be started immediately after final shutdown. According to the strategy for spent nuclear fuel management at INPP part of unburned nuclear fuel assemblies will be transferred from Unit 1 to the Unit 2 and spent nuclear fuel will stay in the pools for a 5 years time before transferring to the interim storage facility. This will really allow the short-lived nuclides of MCC to decay.

For validation of the calculations measurements of the equipment dose rates have been performed. Comparison showed that the predicted dose rates were in rather good agreement with measured values or slightly exceed the highest measured values (by a factor limited to 3). This results from the following conservative assumptions:

- ^{60}Co activity in MCC fluid taken for the calculation was exceeding the most frequently measured activities by factor 2 to 3.
- Fluid velocities in the equipment calculated on the basis of the total MCC flow rate of 48000 m^3/h, corresponding to an electrical output of 1,25 GW, while the actual output does not exceed 70 % of the mentioned value on the average basis.

Future activities

Detailed knowledge of the activity content (inventory) of radioactive waste is necessary for assessment of the potential risk associated with the handling, storage and especially the long-lived disposal of the waste.

A list of nuclides, which are important for the radioactive waste disposal, is defined in 13:

- Most important nuclides that must be determined are ^{14}C, ^{59}Ni, ^{63}Ni, ^{60}Co, ^{94}Nb, ^{90}Sr, ^{129}I, ^{137}Cs, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am.
- Important nuclides that should be determined are ^{36}Cl, ^{99}Tc, ^{234}U, ^{237}Np, ^{238}Pu and ^{244}Cm.

Most of these nuclides are hard-to-measure nuclides. Thus their activity could be assessed using scaling factors. In the future on the basis of the obtained information more attention must be paid to the analysis of all nuclides important to disposal of the waste. Also, based on obtained results, radioactive waste streams for landfill and near surface repositories must be defined, and acceptance criteria for disposal of separate system elements must be analyzed.

CONCLUSIONS

After the radiological evaluation of MCC equipment contamination, as well as the analysis of variation of contamination in time the following conclusions can be drawn:

1. The nuclide composition of deposits on the surface of MCC equipment substantially differs from the nuclide composition of circulating coolant.
2. γ emission from contaminated MCC elements at reactor final shutdown is mostly determined by ^{59}Fe and ^{54}Mn, but after 5 years – by ^{60}Co. With time the percentage of the γ emission from ^{94}Nb and ^{137}Cs nuclides is increasing.
3. In the period of 35 years after the final reactor shutdown the contamination of MCC elements is mainly defined by the long-lived nuclide ^{63}Ni, because short-lived nuclides decay during this period of time (^{55}Fe, ^{59}Fe, ^{58}Co, ^{60}Co, ^{54}Mn).
4. The total radiation dose rate for each MCC element after 5 years since reactor final shutdown substantially decreases; therefore it is necessary to take this into account when planning the dismantling of the systems

ACKNOWLEDGEMENTS

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