INTEGRALLY ESTIMATED NUCLIDE INVENTORY AND DECAY HEAT FROM POTENTIAL NUCLEAR WASTE REPRESENTED BY THE SPENT FUEL FROM THE FUKUSHIMA DAIICHI UNIT 1

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Assuming an accident that may damage spent fuel assemblies at an NPP, nuclear fuel in the spent fuel pool can be considered as radioactive waste. Data on its nuclide composition and heat generation is seen important for the safety assessment of its storage and potential radiological consequences in case of an accident. The SOCRAT/V3 integral code designed to simulate severe accidents at nuclear power plants with VVER power units is able to estimate the nuclide inventory of irradiated fuel based on a one-through calculation and a built-in model. This code was used for a computational analysis of the spent fuel from the Fukushima Daiichi Unit 1, namely, evaluated was its nuclide composition and its decay heat release. The results were compared with the reference data from JAEA (Japan). The calculations demonstrate that in case of severe accident simulations, the use of an integral approach causes no large errors in the estimated nuclide inventory. For most dose-forming radionuclides in the spent fuel, relative deviation of activity from the reference data does not exceed 15%, which appears to be lower than the typical uncertainty under source term predictions. Despite certain overestimation tendency that can be noticed, the decay heat levels calculated for the spent fuel pool appear to be quite similar to those presented in the JAEA report differing by no more than 16% in the first years after the accident, and by 5% ten years after the accident.

Keywords: *radionuclide, fission product, radioactive waste, spent fuel pool, Fukushima Daiichi.*

Introduction

In many respects, the accident that occurred at the Fukushima Daiichi nuclear power station (NPS) in 2011 is seen as a unique event in the history of peaceful atomic energy use. In particular, it was the first severe accident (SA) driven by a catastrophic external impact (tsunami). For the first time ever, it affected three power units at once with the mutual influence of the evolving processes and events observed at each unit. Also, it has been the first SA when a large-scale core damage was observed at three reactor units at once driving potential risk of fuel collapse in four spent fuel pools (SFP). In addition, the accident was followed by successive explosions of a hydrogen-air mixture that destroyed the upper structures of three reactor buildings.

Given its large scale, the accident at Fukushima Daiichi nevertheless yielded relatively small radiological consequences, which was mainly due to a favorable coincidence of circumstances (weather conditions), effective engineering designs and operational staff response. However, potential risk of a large radioactive release was still great, which could be mainly associated not with the degradation of the reactor cores themselves, but rather with the potential failure of fuel in the SFPs sited outside the containment of the reactors. Even though

the SFP draining could be prevented at all power units during the accident, large fragments of the reactor building, namely, its roof and other building structures collapsing into the SFP after hydrogen explosions could potentially break the integrity of the fuel rods and violate the cooled geometry of the spent fuel assemblies (SFA). Currently, there is no evidence indicating the damage caused to the fuel elements in the SFP. At power units No. 3 and No. 4, the fuel has been completely unloaded from the respective SFPs into a common storage pool. Similar operations have not been launched yet at power unit No. 1 and were scheduled for 2027—2028 [12].

After all spent fuel assemblies are removed from the SFPs to the common pool, the fuel will be examined more carefully to identify any signs of damage. However, either way, the question associated with SFA categorization into damaged or undamaged (i. e., formally suitable for reprocessing) fuel still remains open [3]. If damaged, the spent fuel assemblies may be actually categorized as nuclear radioactive waste (RW), since damaged fuel either cannot be reprocessed or its reprocessing appears to be prohibitively expensive. At present time, Japan does not look to have clear plans regarding further management of this fuel, since its national industrial capacities for spent nuclear fuel (SNF) reprocessing are expected to be limited in the near future (commissioning of the Rokkasho plant and the temporary SNF storage facility in Mutsu has been constantly postponed, the Tokai plant is currently undergoing decommissioning [4], the world community advocates for further reduction of the plutonium stockpile). Reprocessing abroad (in France, UK) in accordance with the program that used to be implemented previously, may turn to be challenging specifically for the spent fuel stored at the damaged power units of Fukushima Daiichi. On the other hand, its disposal in Japan without any prior reprocessing seems to be quite unlikely given the geological instability of the region and the public opinion. In this study, the SFA removed from the fuel pools of the Fukushima Daiichi NPS are considered as radioactive waste (RW).

Since the SFA are planned to be stored in an on-site common pool for the next decade, their nuclide composition should be known. Inter alia these are initial data for further simulation of radioactive releases in the event of a hypothetical severe accident at the common pool. The nuclide composition of SFA also affects the decay heat release, which is important to predict the time before the SFA dry up and degrade, and radioactive releases into the environment start to occur. Nuclide composition of SNF is formed during the fuel campaign in the reactor core prior to its unloading into the SFP and depends on the fuel cycle characteristics. During its storage in SFP, it also depends on the time elapsed since the irradiation of a given fuel batch in the core was stopped (cooling time in the SFP).

Radioactive releases may be basically predicted using the so-called integral codes for SA analysis: they can be used to model processes and events at NPP units starting from the accident initiating event to the time when a radioactive release into the environment (so called "source term") occurs. Nuclide composition of nuclear fuel has been previously estimated numerically with integral codes to address real-life emergency situations, in particular, to provide prompt response to the accident at the Fukushima Daiichi NPP in 2011 and the events at the Zaporozhskaya NPP in 2022.

This paper focuses on the numerical estimate of nuclide composition of nuclear fuel in the SFP at Unit No. 1 of the Fukushima Daiichi NPS and its quantitative comparison with the reference data [5] calculated by the Japan Atomic Energy Institute (JAEA) with the ORIGEN2 code.

Assessment method and initial data

The key analytical method implemented in this study is modeling of radionuclide build-up in the fuel throughout a fuel campaign and their subsequent transformations at the cooling stage, as well as comparing the results obtained from the integral formulation with those of high precision calculations (cross-verification). The modeling object is the fuel in the SFP at Unit No. 1 of the Fukushima Daiichi NPS at the time of the accident in 2011, as well as 10 and 15 years after the accident.

Information on the history of SFP loading at Unit 1 of the Fukushima Daiichi NPS provided in the JAEA report [5] were taken as the initial data. This report also provides the results of high precision assessments of fuel burnup, radionuclide inventory and decay heat in SNF calculated using the ORIGEN2 code at the time of the accident initiation and at various times after the accident initiation. These data are considered as referent data for the simulation results obtained with integral code SOCRAT/V3 [6].

Previously, the SOCRAT/V3 code was used to assess the radioactivity accumulated in the cores of damaged power units when preparing the Report of the IAEA Director General on the accident at the Fukushima Daiichi NPP [7]. Later, after the nuclide kinetics model BONUS was refined in the SOCRAT/V3 code, the updated estimates were published in an article [8]. Data on the nuclide composition of the SFP fuel calculated with the SOCRAT/V3 are published for the first time.

In calculation of thermal processes in SFP, one should account for the presence of SFA of different burnup in fuel batches that are unloaded from the core under partial reloading operations. For example, fuel unloaded from VVER units usually includes fuel assemblies that have experienced different numbers of irradiation cycles (for example, 6 and 7 for VVER-440 fuel; 2 and 3 for VVER-1000 fuel), i.e., it consists of fuel assemblies with different burnups. Theoretically, in the calculation model, it is possible to divide each batch of unloaded fuel into 2 groups with the same burnup levels. However, since the number of such batches accommodated in a SFP typically amounts to 5—6, the number of effective groups becomes too big, which considerably complicates the calculations. As for the SFP of the Fukushima Daiichi NPP, there are no data on the burnup distribution in the unloaded fuel batches. The JAEA report provides average characteristics for each SFA batch. This may introduce some error into the estimated nuclide composition, which is nevertheless considered negligible, since the spread of burnup values in each fuel batch is usually small and does not exceed 10 %.

Most integral codes for numerical analysis of SA are not fitted with built-in models enabling the calculation of the initial fuel composition, variation of nuclide inventory during the accident due to radioactive transformations, and the corresponding change of the decay heat. Typically, the initial radionuclide concentrations in the fuel prior to the accident and the decay heat generation curve are calculated based on standalone neutronics calculations and are specified in the integral models of NPP units as initial and boundary conditions. The specific point of the Russian integral code SOCRAT/V3 is a corresponding built-in model BONUS [9]—[11] that allows the user evaluating independently and consistently the fuel burnup levels in fuel during a fuel campaign and the variation of nuclide composition of fuel following reactor shutdown. The decay heat generation is also calculated in the SOCRAT/V3.

Under integral SA calculations, the BONUS model is requested not only at the beginning of the calculation process, but regularly throughout the entire calculation session to track changes in the residual heat release and the concentrations of actinides and fission products (FP) with an account taken of their radioactive decay. An important advantage of the model is its high performance. A typical share of processor time that is spent for the operation of the BONUS model in the integral SA calculations amounts to about 0.2 %. Such a high speed of operation is attained by few simplifying model assumptions described in [10], [11].

High performance of the BONUS model is offset by lower accuracy of prediction compared to stand-alone codes of the ORIGEN2 class, which may be unacceptable when addressing some specific problems (for example, in the fuel re-criticality issues or in the SNF transportation safety assessment). However, this accuracy is acceptable for SA modeling, since it is balanced by the modeling error typical for other processes affecting the release and further transport of radioactive substances and materials from the fuel into the environment.

Assessing the nuclide composition of fuel in SFP

Considering the radiation consequences of SA at NPP units, a representative isotopic composition of a source term would depend on the following parameters:

- accumulated activity;
- dose characteristics of radionuclides (taking into account the internal and external human exposure);
- half-life of radionuclides.

To compare the results obtained in SOCRAT/V3 with those of the JAEA study, the following nuclides were previously considered in the article [8] as important in terms of the radiation exposure:

- actinides ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{239}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm;
- fission products ^{140}Ba, ^{141}Ce, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{138}Cs, ^{131}I, ^{132}I, ^{133}I, ^{134}I, ^{135}I, ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{140}La, ^{99}Mo, ^{88}Rb, ^{103}Ru, ^{106}Ru, ^{89}Sr, ^{90}Sr, ^{91}Sr, ^{131m}Te, ^{132}Te, ^{133}Xe, ^{135}Xe.

This paper also adds to this list the fission products taken into account in deterministic calculations of the source term when simulating the SA at VVER NPPs with the SOCRAT/V3 code (1), as well as fission products that are considered in software tools for rapid assessment of source terms [12]—[15] (2):

(1) ^{133m}Xe, ^{135m}Xe, ^{147}Nd, ^{149}Nd, ^{95}Zr, ^{144}Ce, ^{127}Sb, ^{95}Nb;

(2) ^{85}Kr, ^{105}Rh, ^{90}Y, ^{91}Y, ^{99m}Tc, ^{127m}Te, ^{137m}Ba, ^{129}Te, ^{143}Pr.

Additionally, the ^{239}U actinide was considered as well.

It should be noted that the above list contains radionuclides listed in article [16] as relevant in terms radioactive release regulation and control (with the exception of ^{3}H, ^{14}C, ^{22}Na, ^{41}Ar, ^{54}Mn, ^{60}Co, ^{65}Zn, ^{110m}Ag, ^{222}Rn nuclides, which are specific to SA).

In general, this list of nuclides is typical both for the fuel in the core and the fuel in the SFP (if an accident involving recently unloaded SFA batch is considered). The accident at Unit 1 of the Fukushima Daiichi NPS started almost 1 year after the last SFA batch was unloaded. Almost 13 years have

passed since the accident to date, and some of the nuclides from the above list have decayed completely and are no longer of interest.

The nuclide composition of fuel in the SFP was calculated in SOCRAT/V3 by grouping the SFA within each batch unloaded from the core into the SFP in different years. Respective information on SFP loading is provided in the JAEA report [5]. This method of SFA grouping corresponds to the approach used to simulate the SA in VVER SFP. However, in this case, to assure more consistent conditions of comparison between the SOCRAT/V3 results and the JAEA data, the SFA cooled for over than 3 years have not been collected in one special group, as is commonly done in simulations of SA at VVER in order to save the computational resources. In VVER simulations, such grouping is reasonable, since the contribution of SFA cooled for a long time to the radiation consequences and heat release is deemed insignificant compared to SFA loaded into the pool more recently.

Thus, the following data was used on each SFA batch:

- number of SFA in a batch;
- irradiation time in the core;
- cooling time in SFP;
- specific SFA power;
- specific mass of uranium in SFA (assuming the same value for all batches).

In the JAEA report, the average initial enrichment of fresh fuel that is loaded into the core and irradiated during the fuel cycle, was not specified for each SFA batch, therefore it was assumed that all batches had the same initial enrichment.

Published data evidence that there were 292 SFA in the SFP at the time of accident initiation with the most recent batch of 64 SFA which has been loaded into the SFP almost 1 year before the accident. It is interesting to notice that the SFP stored 74 SFA that were unloaded from the core back in the 1970s.

Tables 1 and 2 present the SFP radioactivity calculated in the SOCRAT/V3 code. As noted above, by the time when the accident at Unit 1 started some considerable part of the dose-forming FP had already decayed either almost or completely. Figures 1 and 2 show relative deviation of the SOCRAT/V3 results from the data presented in the JAEA report. As follows from the figures, the differences in the activity for most of the remaining radiologically significant nuclides does not exceed 15 %, but for some nuclides (^{136}Cs, ^{106}Ru, ^{144}Ce, ^{144}Pr, ^{147}Pm, ^{148m}Pm, ^{241}Am, ^{242}Cm) it amounts to 20—30 %.

It should be noted that for ^{239}Np, JAEA data demonstrate a constantly high activity of 3.4E+13 Bq regardless of the cooling time. The correctness of

Table 1. Activity (Bq) of dose-forming fission products in the SFP of Unit 1 according to integral calculations with SOCRAT/V3 code, for different times since the accident initiation

Nuclide	Time (years)		
	0	10	15
^{137m}Ba	$1.60\cdot10^{17}$	$1.28\cdot10^{17}$	$1.14\cdot10^{17}$
^{140}Ba	$2.06\cdot10^{9}$	0	0
^{141}Ce	$2.11\cdot10^{14}$	0	0
^{144}Ce	$1.75\cdot10^{17}$	$2.45\cdot10^{13}$	$2.89\cdot10^{17}$
^{134}Cs	$1.07\cdot10^{17}$	$3.77\cdot10^{15}$	$7.04\cdot10^{14}$
^{135}Cs	$1.11\cdot10^{12}$	$1.11\cdot10^{12}$	$1.11\cdot10^{12}$
^{136}Cs	$1.21\cdot10^{8}$	0	0
^{137}Cs	$1.70\cdot10^{17}$	$1.36\cdot10^{17}$	$1.21\cdot10^{17}$
^{85}Kr	$1.27\cdot10^{16}$	$6.73\cdot10^{15}$	$4.88\cdot10^{15}$
^{140}La	$2.38\cdot10^{9}$	0	0
^{95}Nb	$1.79\cdot10^{16}$	0	0
^{103}Ru	$8.02\cdot10^{14}$	0	0
^{106}Ru	$1.45\cdot10^{17}$	$1.60\cdot10^{14}$	$5.33\cdot10^{12}$
^{89}Sr	$1.53\cdot10^{15}$	0	0
^{90}Sr	$1.16\cdot10^{17}$	$9.15\cdot10^{16}$	$8.12\cdot10^{16}$
^{127m}Te	$1.70\cdot10^{14}$	0	0
^{90}Y	$1.16\cdot10^{17}$	$9.16\cdot10^{16}$	$8.12\cdot10^{16}$
^{91}Y	$3.89\cdot10^{15}$	0	0
^{95}Zr	$8.29\cdot10^{15}$	0	0
^{147}Nd	$1.09\cdot10^{17}$	0	0
^{154}Eu	$8.13\cdot10^{15}$	$3.66\cdot10^{15}$	$2.45\cdot10^{15}$
^{147}Pm	$1.09\cdot10^{17}$	$7.86\cdot10^{15}$	$2.10\cdot10^{15}$
^{143}Pr	$6.13\cdot10^{9}$	0	0
^{144}Pr	$1.75\cdot10^{17}$	$2.45\cdot10^{13}$	$2.89\cdot10^{11}$

Table 2. Activity (Bq) of actinides in the SFP of Unit 1 according to integral calculations with the SOCRAT/V3 code, for different times since the accident initiation

Nuclide	Time (years)		
	0	10	15
^{241}Am	$1.32\cdot10^{15}$	$3.18\cdot10^{15}$	$3.80\cdot10^{15}$
^{242}Cm	$5.94\cdot10^{15}$	$1.07\cdot10^{9}$	0
^{244}Cm	$3.92\cdot10^{15}$	$2.69\cdot10^{15}$	$2.22\cdot10^{15}$
^{237}Np	$4.50\cdot10^{11}$	0	0
^{238}Pu	$5.94\cdot10^{15}$	$5.56\cdot10^{15}$	$5.34\cdot10^{15}$
^{239}Pu	$5.09\cdot10^{14}$	$5.12\cdot10^{14}$	$5.12\cdot10^{14}$
^{240}Pu	$8.71\cdot10^{14}$	$8.76\cdot10^{14}$	$8.76\cdot10^{14}$
^{241}Pu	$1.47\cdot10^{17}$	$9.14\cdot10^{16}$	$7.17\cdot10^{16}$
^{242}Pu	$3.56\cdot10^{12}$	$3.58\cdot10^{12}$	$3.58\cdot10^{12}$
^{235}U	$4.73\cdot10^{10}$	$4.77\cdot10^{10}$	$4.77\cdot10^{10}$
^{236}U	$4.72\cdot10^{11}$	$4.75\cdot10^{11}$	$4.75\cdot10^{11}$
^{238}U	$5.75\cdot10^{11}$	$5.80\cdot10^{11}$	$5.80\cdot10^{11}$

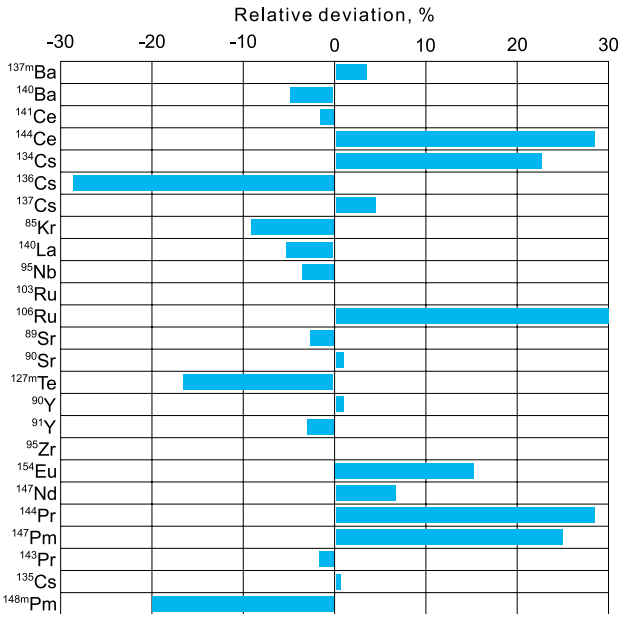

Figure 1. Relative deviation of calculated FP accumulation in the SFP of Unit 1 from the JAEA data at the time when the accident started

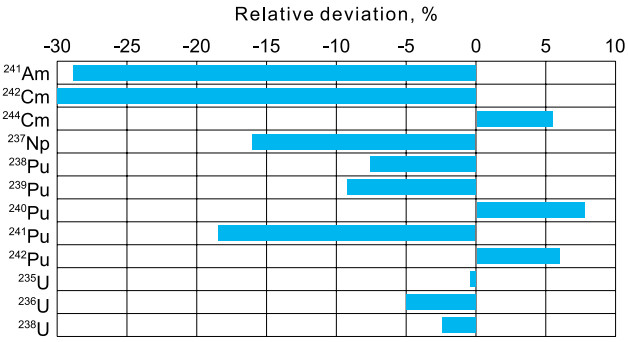

Figure 2. Relative deviation of calculated actinides accumulation in the SFP of Unit 1 from the JAEA data at the time when the accident started

this result is questionable, given the short half-life of ^{239}Np relative to the cooling time (2.4 days and 351 days, respectively), the rapid complete decay of the parent nuclei ^{239}U (half-life of 23 minutes) and, conversely, very slow α-decay of the parent nuclei ^{243}Am.

The deviations in the activities of ^{144}Pr and ^{144}Ce coincide because the activity of ^{144}Pr is governed by the radioactive decay of ^{144}Ce.

Calculations assuming a cooling time of 10 years since the beginning of the accident (that is, the last unloaded fuel batch has been cooled for 11 years) showed similar deviations from the JAEA data (Figures 3, 4). For most nuclides considered important in terms of radiation exposure and having high activities, the deviation did not exceed 15 %; for ^{144}Ce, ^{106}Ru, ^{106}Rh, ^{144}Pr, ^{147}Pm, ^{241}Am, ^{242}Cm it amounted to 20—30 %. Only for ^{127m}Te the deviation increased from – 16% to – 55 %.

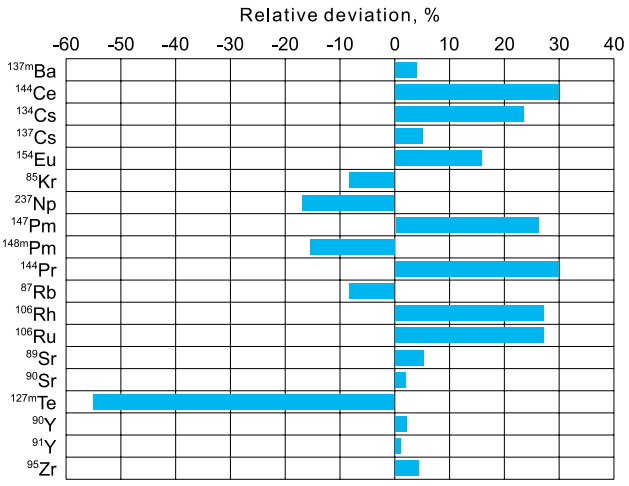

Figure 3. Relative deviation of calculated FP accumulation in the SFP of Unit 1 from the JAEA data 10 years after the accident

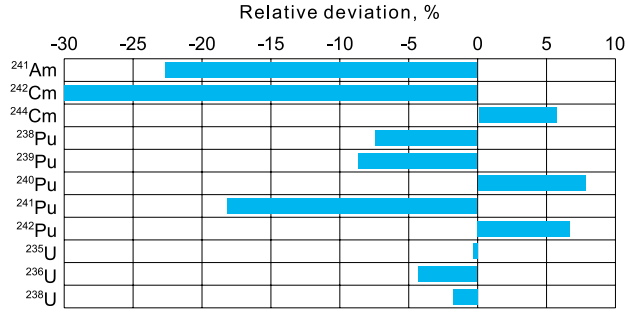

Figure 4. Relative deviation of calculated actinides accumulation in the SFP of Unit 1 from the JAEA data 10 years after the accident

Estimated SFA decay heat generation in the core and in the SFP

The nuclide composition calculated in SOCRAT/V3 was used to calculate the decay heat generation in the SFP of Unit 1(Table 3). The calculated value of power in the SFP is in good agreement with the value of 180 kW predicted by TEPCO [17]. It can be noted that the decay heat generation in the SFP was relatively low at the time of the accident initiation, which provided a significant margin of time before the water started boiling and, accordingly, for taking measures to supply water to the pool on March 31, 2011, i. e. almost three weeks after the accident started. At present time, the heat generation in the SFP of Unit 1 has decreased by over an order of magnitude.

Table 3. The calculated dependence of decay heat generation in the SFP on the time elapsed since the beginning of the accident

Time, years	0	1	2	5	10	15
Power, kW	178	117	89	60	49	12

The decay heat values calculated for different times ranging from 0 to 10 years elapsed since the onset of the accident were compared with those calculated by JAEA with the ORIGEN2 code. Figure 5 presents relative deviations of the calculated decay heat from the JAEA referent data. The SOCRAT/V3 models tend to overestimate the SFA power calculated by JAEA by 12—16 % in the first 2 years, and by almost half of this value afterwards. Figure 5 shows relative deviations of the SOCRAT/V3 results from the JAEA data at the same times, but for the core fuel. It should be noted that, strictly speaking, direct comparison of the cross-verification results obtained for the SFP and for the core is incorrect, since at the time of the accident the fuel burnup and nuclide composition in the core and in the SFP were different. However, a qualitative comparison shows that for different cooling times longer than 1 year the SOCRAT calculation overestimates the JAEA data on the decay heat in a same way both in the core and SFP.

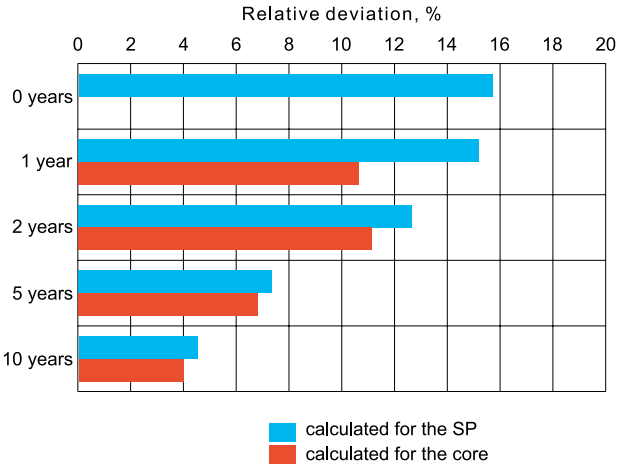

Figure 5. Relative deviation of the decay heat calculated by SOCRAT/V3 from the decay heat in JAEA data, for the SFP and the core

JAEA data shows that the decay heat generation in SFA in the first cooling years is mainly contributed by the following radionuclides (Figure 6): ^{106}Ru (19 %), ^{144}Pr (18 %), ^{134}Cs (16 %), ^{90}Y (11 %), ^{137m}Ba (10 %), ^{242}Cm (7 %). As shown above in the estimate of the nuclide composition of SFP fuel, the activities of ^{106}Ru, ^{144}Pr and ^{134}Cs calculated using the SOCRAT/V3 exceed those presented in the JAEA data by 20—30 %, whereas for ^{90}Y and ^{137m}Ba, the corresponding relative deviation of the calculated activities is very small. The activity of ^{242}Cm according to SOCRAT/V3 is 30 % less than the one specified by JAEA, nevertheless its contribution to decay heat is several times lower than the that of nuclides with overestimated activity. Thus, the cross-verification results for the decay heat can

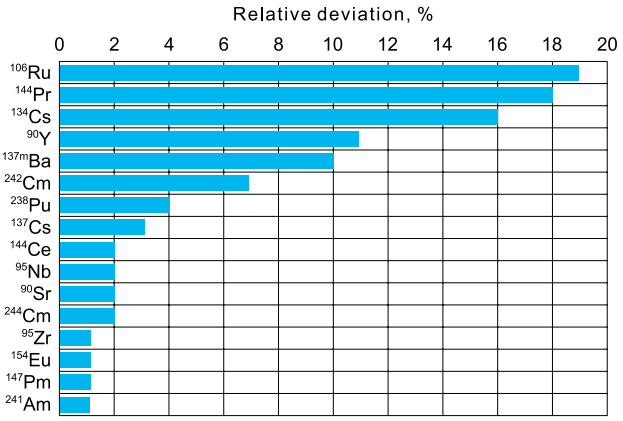

Figure 6. Relative contribution of nuclides to SFA decay heat in the SFP of Unit 1 at the beginning of the accident (according to JAEA data)

be deemed consistent with the calculated nuclide composition.

A noticeable overestimation of ^{106}Ru and ^{134}Cs build-up relative to the JAEA data has been identified for the core as well [8]. However, validation calculations performed with SOCRAT/V3 using the SFCOMPO2.0 data as referent reveal an error of [−8 %; +10 %] for ^{134}Cs, i. e., no overestimation tendency is observed, and the maximum value is almost half as large as the deviation from the JAEA data for the SFP at the beginning of the accident.

As noted in [8], a quite considerable overestimation of ^{134}Cs and ^{106}Ru build-up may be produced by differences in calculations of the main fissile actinides (^{235}U and ^{239}Pu), since in accordance with the yield curve the direct yield of these fission products tends towards the maximum probability. On the other hand, the deviations may be also driven by some specific features of the models and different evaluated nuclear data libraries used to set the cross-sections, as well as insufficient information on specific nuclides and various corresponding assumptions. A more detailed information on the underlying data and the assumptions made by the JAEA in their study is required to explain the reason for the observed discrepancy in the results.

Similarly, the decreased overestimation of decay heat power after 10 years of cooling in SOCRAT/V3 calculation is also explained by differences in the activities of radionuclides which provide the biggest contribution to the decay heat. In this case, these are ^{90}Y (29 %), ^{137m}Ba (25 %), ^{238}Pu (12 %), ^{137}Cs (8 %), ^{241}Am (8 %), ^{90}Sr (6 %), ^{244}Cm (5 %) (Figure 7). With the exception of ^{241}Am, the activities of all these radionuclides in the SOCRAT/V3 calculations and in the JAEA data are quite similar with a slight overestimation tendency in the former case. Therefore, in general, the difference in the predicted decay heat power appears to be quite small.

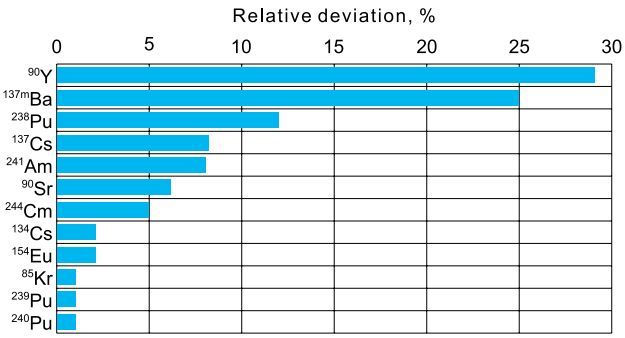

Figure 7. Relative contribution of radionuclides to the decay heat power in the SFP of Unit 1 ten years after the accident (according to JAEA data)

It can be noted that the calculated deviations of decay heat on the day of the accident and 10 years after (or 1 year and 11 years of fuel cooling in the SFP after scram) are qualitatively consistent with the validation results provided for the BONUS model based on RB-093-20 data [18]. It was demonstrated in [11] that decay heat in VVER fuel assemblies with fuel enrichment of 4.3 % and a burnup of 36 MWt·day/kgU was overestimated by 5 % if they have been cooled for 1 year, by 8 % in case of their cooling for 3 years and by less than 1 % when cooled for 10 years. Such quantitative difference in the degree of reference data overestimation can be explained by slightly different parameters of the SFA (enrichment of 3.7 % and 4.3 %, burnup of 40 MWt·day/kgU and 36 MWt·day/kgU), and by inconsistency in the reference data (local inconsistency of JAEA results were noticed above), or by incomplete description of assumptions adopted by JAEA. For example, the same enrichment was assumed for all fuel batches unloaded in different years. This assumption may be wrong, and different enrichment values were used in the JAEA study in fact.

Conclusion

A computational analysis of nuclide composition and decay heat generation has been performed in this work for the fuel in the SFP at Unit 1 of the Fukushima Daiichi NPP. Based on conservative assumptions on the impossibility of its reprocessing in the coming 10—20 years, this fuel has been referred to the RW category. The study is aimed at evaluation of the radioactivity of the fuel in case of a hypothetical severe accident either in the SFP of the Unit or in the common fuel pool at the NPP site, as well as at comparison of results with reference data from the JAEA (Japan). The Russian integral code SOCRAT/V3 was used in this study, which has both the model for radionuclides build-up and the model for consistent calculation of decay heat in fuel driven by radioactive transformations.

Results of integral calculations performed with SOCRAT/V3 code were compared with the reference data obtained by JAEA using the ORIGEN2 code, which provided data on relative deviations for the activity of dose-forming radionuclides and the decay heat power in the SFP.

As it comes to the nuclide composition of fuel that was located in the SFP at the time of the accident initiation and included several batches stored for 1—30 years, it is shown that the deviation in the activity of long-lived dose-forming radionuclides from the JAEA reference data does not exceed 30 % and is falling within the range of ± 10 % for most of the nuclides. The resulting deviations could be considered acceptable for issues of integral simulation of SA at SFP and reactor core.

Comparison of calculated decay heat generation in SFP with the JAEA data also reveals an overestimation tendency of 5—16 % (for fuel that has been cooled from 1 to 10 years). Thus, when simulating the processes in the SFP during a hypothetical accident at Unit 1 of the Fukushima Daiichi NPS using the SOCRAT/V3 code, one should expect a slight overestimation of the SFP heating and drying rate until the burst of fuel rod claddings, when the heat generation in the fuel rods starts decreasing due to release of semi-volatile FP from the fuel.

Comparison of the calculated data with the JAEA findings shows that when using the SOCRAT/V3 code in the future, particular attention should be paid to the accuracy of build-up prediction for the following radionuclides that correspond to the largest deviations obtained: ^{85}Kr, ^{106}Ru, ^{134}Cs, ^{136}Cs and ^{144}Ce. The lack of experimental data impedes the validation of the fuel burnup model. Thus, additional experimental studies are required to assess the predictive accuracy of nuclide kinetics model in SOCRAT/V3. This problem can be addressed through integral experiments where the decay heat power of the irradiated fuel samples was measured at different times after the end of their irradiation.

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