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Status Of The Spent Fuel In The Reactor Buildings Of Fukushima Daiichi 1-4

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ABSTRACT

The ratios of the radio nuclides Cs-134g and Cs-137 deduced from measurements of liquid samples from the spent fuel pools in Fukushima Daiichi 1-4 are used to interpret the status of the spent fuel assemblies in the pools of the damaged reactor buildings. The different nature of the production of Cs-134g (neutron capture product of Cs-133) and Cs-137 (cumulative fission product from mass chain 137) and the different half-lives (2.06 years and 30.17 years respectively) require a complicated calculation of the mass and activity of the two nuclides. These masses are depending on the local burn up of the fuel, the burn up history and the radioactive decay. For the calculation of the radionuclide masses with ORIGEN-2 the user has to use different cross section tables depending on the maximum burn up of the fuel. These tables are based on data from activity measurements after irradiation of fissionable materials. Especially for the neutron capture product Cs-134g the calculation is more complicated, because the production of Cs-133 (stable cumulative fission product from mass chain 133) has to be taken into account. The neutron capture cross section for Cs-133 was fitted from a gamma scan of spent fuel rods in a hot cell. The method of the calculation of the nuclide activities and the interpretation of the gamma measurements of the spent fuel pool samples from Fukushima Daiichi 1-4 are described in detail.

1 INTRODUCTION

For the interpretation of the source of radioactive materials very often nuclide ratios are used. The most well-known ratio is that from I-131/Cs-137 (Fig. 1). Both are fission products and both can be measured easily using a high resolution (HPGe) gamma detector. In case of the presence of I-131 activity in the gamma measurements from Fukushima samples it can be deduced, that the activity was released from fuel from one of the reactor cores of the units 1-3. The short half-live of I-131 (8.02 days) allows this possibility of interpretation only for a few weeks. Afterwards the activity of I-131 will be below the detection limit of the gamma detectors. For the long-term analysis of the origin of the measured activity the ratio of the nuclides Cs-134g/Cs-137 is of high importance. An additional advantage of this ratio, unlike I-131/Cs-137, is the fact that both nuclides undergo the same chemical reactions. Independently of its release path, its chemical combination with other elements or its physical state of matter the ratio will not change. This ideal pair is now used to interpret the change of the origin in the samples of the Fukushima spent fuel pools.

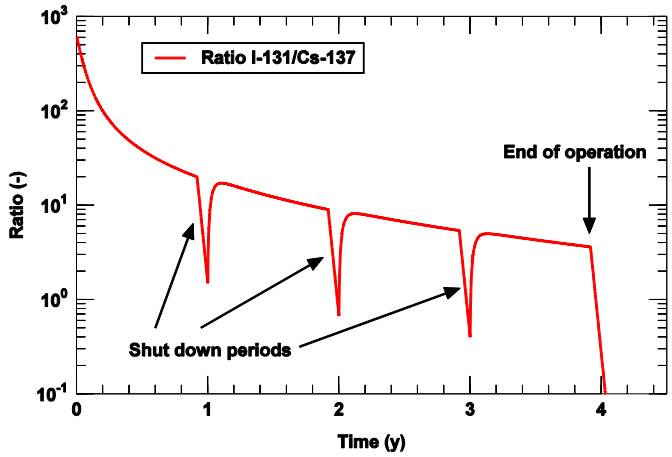

Figure 1: Nuclide ratio of I-131/Cs-137 during operation

2 NUCLIDE PRODUCTION OF CS-134G AND CS-137

The nuclear fission process from U-235 produces hundreds of fission products. Most of them are short living nuclides which are decaying into long living or even stable nuclides. Some of the produced radionuclides undergo a more complicated production way like the transuranium nuclides and neutron capture products such as Cs-134g. Fig. 2 shows the minimum potential energy curve (MPE) for the fission process of the neutron induced fission of U-235 together with the stable nuclides and the most important fission products and nuclear capture products with half-lives longer than 1 hour. The fission products produced along the MPE-Curve have high excitation energies and will cool down by neutron emission. With the assumption, that the heavy fission product releases two neutrons and the light fission partner releases one neutron the full line after neutron emission was constructed. All the independently produced nuclides are far from this curve and therefore they cannot be produced by the fission process. Fig. 3 shows part of the nuclide chart including the caesium isotopes.

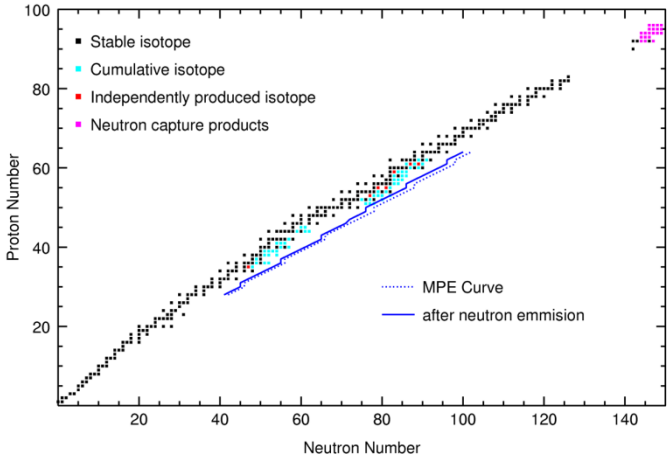

Figure 2: MPE Curve of U-235 (n,f) reaction

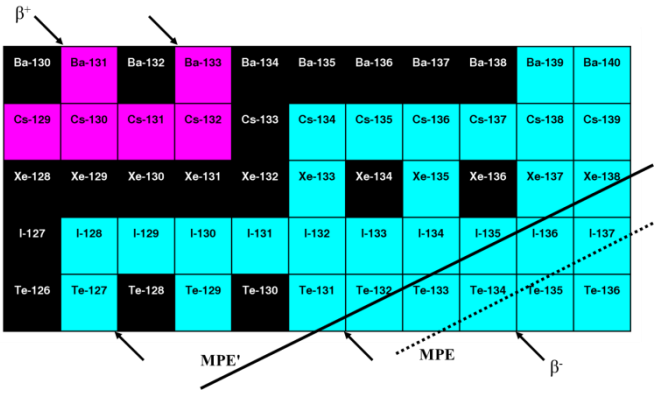

Figure 3: Part of nuclide chart with caesium nuclides

In case of the neutron capture process a neutron is captured by the mother nuclide and forms an excited daughter nuclide which cools down under gamma emission. The transuranium nuclides start from U-235 and U-238, where the production of Cs-134g started from Cs-133. The latter nuclide firstly has to be produced by the fission process as decay product from the mass chain 133 because it does not exist in the fresh fuel. While the production of Cs-133 Eq. (1) and Cs-137 Eq. (2) is very similar the production of Cs-134g Eq. (3) by replacing the number of Cs-133 moles with Eq. (1) is a square function of the time t, see Eq. (4).

$$N_{Cs\text{-}133}=N_{U\text{-}235} * \sigma_{n,f} * \Phi_n * 0.06609 * t \quad (1)$$

$$N_{Cs\text{-}137}=N_{U\text{-}235} * \sigma_{n,f} * \Phi_n * 0.06236 * t \quad (2)$$

$$N_{Cs\text{-}134g}=N_{Cs\text{-}133} * \sigma_{n,\gamma} * \Phi_n * t \quad (3)$$

$$N_{Cs\text{-}134g}=N_{U\text{-}235} * \sigma_{n,f} * \sigma_{n,\gamma} * \Phi_n^2 * 0.06609 * t^2 \quad (4)$$

N: Number of moles
$\sigma_{n,f}$: Fission cross section
$\sigma_{n,\gamma}$: Neutron capture cross section
Φ_n: Neutron flux

After addressing the decay of the radionuclides and investigating the neutron capture cross section for the Cs-133 (n,γ) reaction the ratio of the caesium nuclides could be calculated in dependency from the burn up. In the chart of nuclides [1] the cross section for thermal neutrons is given with 29 barn. By using this cross section the calculation of the Cs-134g/Cs-137 ratio never exceeded a value of 0.2 also up to very high burn up.

The experimentally measured ratio for different burn up could be deduced from a gamma scan of fuel pins from a BWR with different mean burn up. The scan result of a high burn up fuel pin is shown in Fig. 4. The mean burn up of the fuel pins is known and the local burnup can be easily calculated by using the distribution of the Cs-137 activity as value which is proportional to the local burn up. From this information the burn up dependent nuclide ratios could be drawn (Fig. 5). Two curves can be recognized, which can be explained by the different cross section due to different neutron energy distributions. In the lower part of the BWR the void coefficient is lower and therefore the moderation of the neutrons is better which results in a lower mean neutron energy. This lower neutron energy results in a lower neutron capture cross section. The same effect is used in the fast breeder reactor, where the higher neutron mean energy is used to enhance the neutron cross section for the production of plutonium. A fit using the production rate functions to this data results in a neutron capture cross section for the lower curve of 180 barn and for the upper curve of 200 barn. A test

calculation for the verification of the data was executed and compared with data points of the VERDON 2 experiment (Fig. 6). The difference of about 7% compared with the data can be explained by the detector efficiency difference for the two gamma lines of the caesium nuclides, which should be between 5% and 10%.

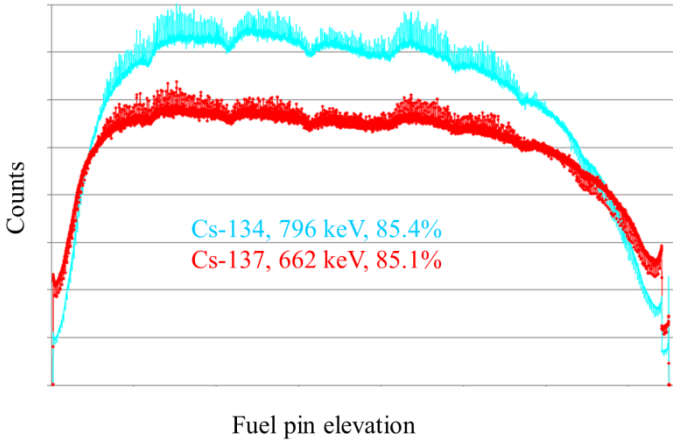

Figure 4: Gamma scan of high burnup fuel pin

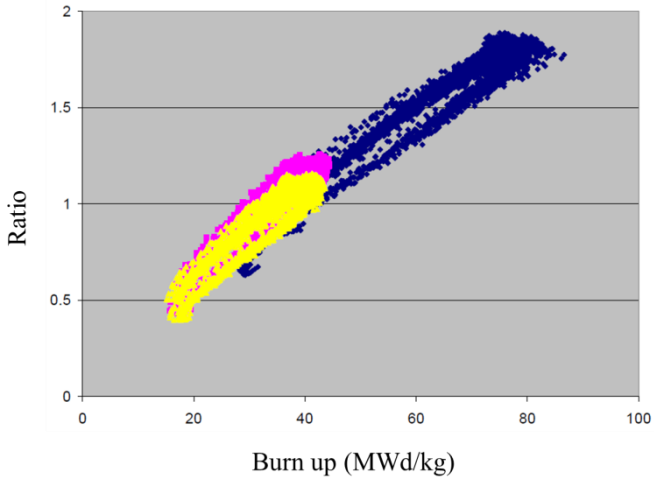

Figure 5: Cs-134g/Cs-137 ratio from fuel pins

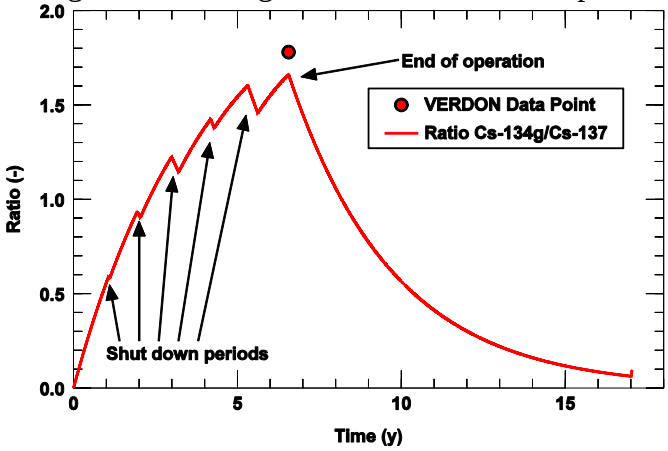

Figure 6: Cs-134g/Cs-137 ratio calculation

3 INTERPRETATION OF FUKUSHIMA SPENT FUEL POOL DATA

The activity data measured in the Fukushima spent fuel pool samples will be interpreted by using the development of the ratio of the two caesium nuclides Cs-134g and Cs-137. Important data for the interpretation is the time schedule of the accident in Table 1. Each of

these events is connected with radioactive release into the environment from possibly different sources. The ratio of the caesium nuclides and especially the presence of the short living I-131 can clearly show, if the activity measured is released from a reactor core operating until start of the accident or from a spent fuel pool.

Table 1: Time table of Accident events

Important dates	Event	Reason
March 11, 2011, 15:46	Start of Accident	Tsunami
March 12, 2011, 15:36	Explosion in unit 1	Deflagration
March 15, 2011, ~06:00	Explosion in unit 2	Deflagration in suppression chamber
March 14, 2011, 11:01	Explosion in unit 3	Deflagration
March 15, 2011, 06:00	Explosion in unit 4	Deflagration

3.1 Unit 1

The explosion of the unit 1 reactor building happened only one day after the initiation of the accident. Until this time there was no radioactive release from the other units. The hydrogen deflagration destroyed the complete upper part of the reactor building and produced the first major release of radioactivity into the environment. Some activity measurements from samples of the spent fuel pool of unit 1 are shown in Fig. 7. The measured activity is shown on the right hand axis and the ratio of the two caesium nuclides is shown on the left hand axis. The full line shows the calculated development of the nuclide ratio fixed on the first point of measurement. Because there are no uncertainties given from the measured activities, no significant change of the nuclide ratio due to release from spent fuel can be deduced from the data. Almost all of the activity originates from the fallout in the first phase of the accident and not from later radioactive release from possibly destroyed spent fuel assemblies. Because the first measurement was done after about 3 month after initiation of the accident, all data include possible fallout from the explosions in the other units. The measured Cs-137 activity stays still at about $2 \cdot 10^4$ Bq/ml.

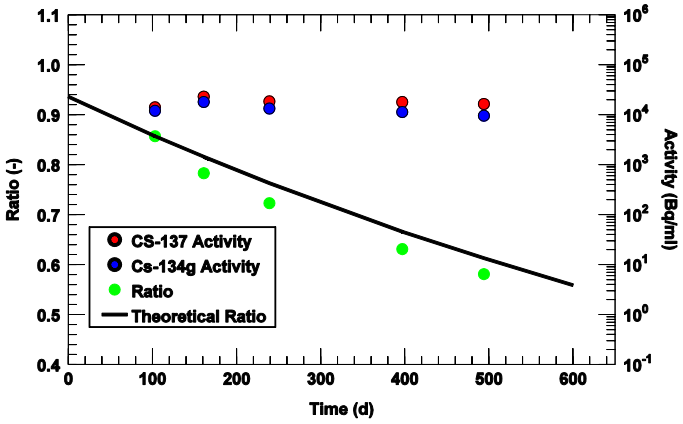

Figure 7: Nuclide ratio of unit 1 activity measurements

3.2 Unit 2

The explosion of unit 2 is not mentioned in most of the Fukushima reports. There was a sound in the area of the suppression chamber at about 6:00 am on 15th of March in 2011. The upper part of the reactor building stayed intact and fallout from the other units could not

contaminate the spent fuel pool of unit 2. The high activity at the first measurements of 10^5 Bq/ml (Fig. 8) can only originate from fission products in the vicinity of this explosion in the suppression chamber of unit 2, being carried way into the reactor building and sediment into the spent fuel pool. About 240 days after the initiation of the accident most of the activity was removed by an ion exchange process running some weeks and a follow on desalination process. These cleaning processes removed more than 3 orders of magnitude from the activity at the beginning. About 1 year after the accident the ratio started to reduce faster than the theoretical curve. This fact has to be interpreted as due to an additional release of caesium from (mechanically) damaged spent fuel. A calculation of the total content of Cs-137 in one 9x9 fuel assembly with a mean burn up of 55 MWd/kg dissolved completely in the water of the full spent fuel pool would result in an activity of 10^6 Bq/ml. This means that less than 1% of caesium released from one single fuel pin is sufficient to explain the change in the nuclide ratio.

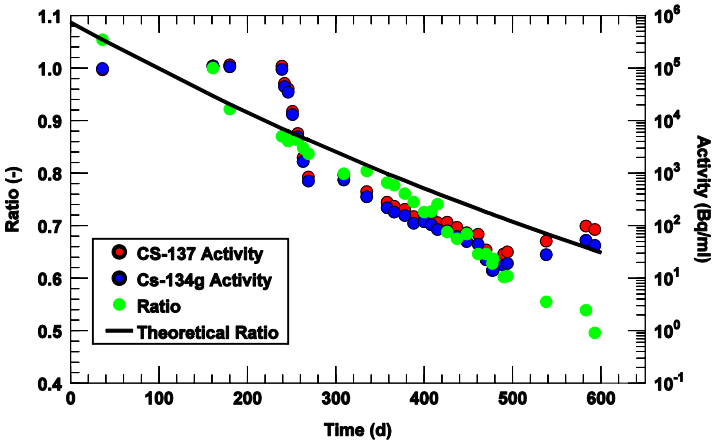

Figure 8: Nuclide ratio of unit 2 activity measurements

3.3 Unit 3

The second explosion during the Fukushima Daiichi accident destroyed completely the upper reactor building of unit 3. In several photos available in the internet it could be seen that a lot of steel carriers and concrete blocks must have been fallen into the spent fuel pool. It would be almost impossible not to assume greater mechanical damage of the spent fuel in this pool. Surprisingly at Fig. 9 it can be seen clearly that there is absolutely no change from the measured ratios compared with the theoretical curve even not during the ion exchange phase 300 days after the accident. There are two possible explanations, firstly there is no damage in spent fuel pool of unit 3 or secondly the activity of about $4 \cdot 10^3$ Bq/ml covers a small possible damage which should not be greater than that in unit 2.

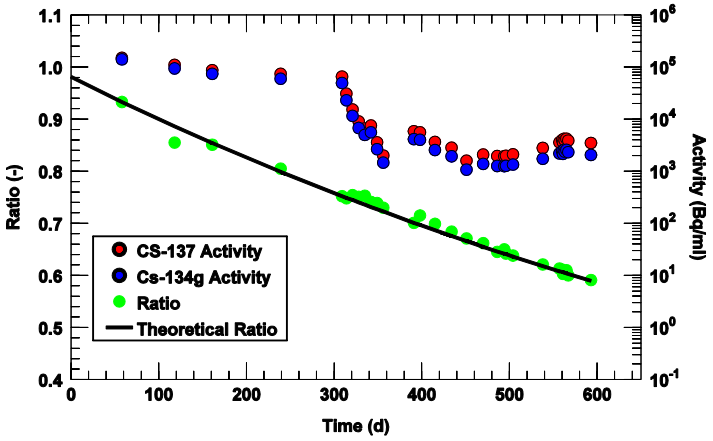

Figure 9: Nuclide ratio of unit 3 activity measurements

3.4 Unit 4

The most surprising event during the Fukushima accident was the explosion of the reactor building of unit 4. This explosion went in parallel with a strong increase of the radioactivity monitored at the Fukushima area. This caused a lot of confusion also in the groups of severe accident experts. Measured activities of the caesium nuclides and also I-131 showed that spent fuel was not the source of the release but reactor fuel that was recently undergoing fission. The event of the explosion in unit 2 suppression chamber at almost the same time was responsible for the increase of radiation at the Fukushima area but a common pipe connecting units 3 and 4 with the common offgas unit was responsible for a slow hydrogen flow from unit 3 into the reactor building of unit 4 which led to the observed deflagration after a given hydrogen concentration was reached. Measurements of the activity in the unit 4 spent fuel pool about one month after the accident showed clearly that the inventory of the spent fuel pool did not undergo any degradation (Fig. 10). Firstly the activity was very low with 10^2 Bq/ml compared with the measurements of the other pools and secondly the starting value of the caesium nuclide ratio was identical with that from unit 3 spent fuel pool. This confirms that hydrogen (and aerosols) from unit 3 where transferred into the unit 4 reactor building. Another point which supports this argument is the measured soil contamination from the playground about 500 m from the reactor buildings measured on 31st of March 2011. The value for Cs-137 was $4.9 \cdot 10^5$ Bq/kg. Under the assumption that the sample was taken as standard sample (1 cm thick) and the density is in the order of 2 g/cm^3, this corresponds with an activity of 10^7 Bq/m^2. If all the activity in the spent fuel pool of unit 4 would originate from fallout, the fallout must have been in the order of 10^9 Bq/m^2 for Cs-137.

The ion exchange and desalination processes further reduced the activity of the Cs-137 to less than 1 Bq/ml. The strong scattering of the nuclide ratio implies that the relative uncertainty of the gamma measurement is much higher compared with the measurements of the other units. It cannot be excluded, that very slight damage of spent fuel may have happened in the first 3 months, but this damage should be much less compared with unit 2.

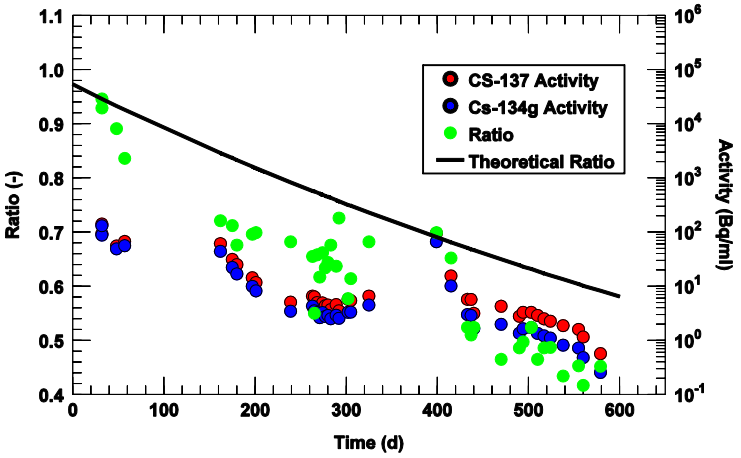

Figure 10: Nuclide ratio of unit 4 activity measurements

4 CONCLUSION

The spent fuel pools in Fukushima which have a much higher risk for long-term land contamination because of their high content of long living radio nuclides compared with the reactor cores were not damaged by the accident. The spent fuel pools of units 1 and 3 do not show any significant change in the measured nuclide ratio compared with the theoretical curve. But still it cannot be excluded that a very slight damage occurred but the evidence is masked by the relative high remaining activity (10^3 – 10^4 Bq/ml). In the spent fuel pool of unit 2 with an activity of 100 Bq/ml Cs-137 a clear trend is visible, suggesting a small release of caesium from the spent fuel that may be due to mechanical damage. In the most important and most endangered spent fuel pool of unit 4 only very low activities were measured and any possible damage would be lower as in the pool of unit 2. The last measured C-137 activity was in the order of 600 Bq/l which can be compared with the natural activity of seawater which is of the order of 20 Bq/l from K-40.

ACKNOWLEDGMENTS

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[1] G. Pfennig, H. Klewe-Nebenius, W. Seelmann-Eggebert, Karlsruher Nuklidkarte, Druckhaus Haberbeck, D-32791 Lage, Germany, December 1998