# WHAT WAS THE CAUSE OF THE HIGH Cl-38 RADIOACTIVITY IN THE FUKUSHIMA DAIICHI REACTOR #1[1]

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I have been totally consumed the last few weeks by one thing, day and night, and those are the events unfolding in Japan. I keep on alternating between complete disbelief and acceptance of the gravity of the situation, but mostly disbelief. And I am not the only one. Most of the nuclear physicists and engineers with whom I have spoken since the incident cannot - will not - believe that it is possible that some of the fuel that is melting could somehow produce little pockets that could go critical. I believed them for the longest time until the following came on the Kyodo news website (relevant text italicized below for emphasis) and I did the following analysis.

> “Neutron beam observed 13 times at crippled Fukushima nuke plant
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> TOKYO, March 23, Kyodo
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> **Tokyo Electric Power Co. said Wednesday *it has observed a neutron beam, a kind of radioactive ray, 13 times on the premises of the Fukushima Daiichi nuclear plant after it was crippled by the massive March 11 quake-tsunami disaster.***
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> TEPCO, the operator of the nuclear plant, said the neutron beam measured about 1.5 kilometers southwest of the plant's No. 1 and 2 reactors ***over three days from March 13 and*** is equivalent to 0.01 to 0.02 microsieverts per hour and that this is not a dangerous level.
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> The utility firm said it will measure uranium and plutonium, which could emit a neutron beam, as well.
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> In the 1999 criticality accident at a nuclear fuel processing plant run by JCO Co. in Tokaimura, Ibaraki Prefecture, uranium broke apart continually in nuclear fission, causing a massive amount of neutron beams.
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> In the latest case at the Fukushima Daiichi nuclear plant, such a criticality accident has yet to happen.
>
> But the measured neutron beam may be evidence that uranium and plutonium leaked from the plant's nuclear reactors and spent nuclear fuels have discharged a small amount of neutron beams through nuclear fission.”
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> ==Kyodo News, http://english.kyodonews.jp/news/2011/03/80539.html

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[1] Thanks go to Drs Patricia Lewis (CNS, MIIS) and Arjun Makhijani (IEER) for carefully reviewing this memo and for thoughtful and stimulating discussions.

Also, on March 25th, TEPCO made public a measurement of the contributions of different isotopes to the extremely high measured radioactivity of the seawater used to cool reactor #1. The reasons as to why these measurements were taken so late on in the crisis (or why the information was released so late on) is unclear at this stage.

| Radioactive Nuclide | Concentration (Bq/cm3) |
|---|---|
| Cl-38 | 1.6e6 |
| As-74 | 3.9e2 |
| Y-91 | 5.2e4 |
| I-131 | 2.1e5 |
| Cs-134 | 1.6e5 |
| Cs-136 | 1.7e4 |
| Cs-137 | 1.8e6 |
| La-140 | 3.4e2 |

**Table 1: The contribution of different isotopes to the radio-activity from a sample taken in the turbine building of reactor #1.**[2]

The measured levels of Cs-137, I-131 were expectedly very high. The very high concentration of one isotope however – Cl-38 – was the figure that drew my attention. Why worry? Cl-38 has a 37-min half-life beta decay; in a couple of days it will be gone. However, it was the fact that it was there at all, and in such high concentration, that puzzled me. Could it be that the incident flux of neutrons converted the 24% Cl-37 present naturally in salt to Cl-38 through radiative neutron capture (a simple reaction: add a neutron give up a gamma, and you have Cl-38)? What flux could have produced the observed radioactivity? In what follows, I attempt to calculate the neutron flux that would have been able to produce the observed radioactivity. There is a bit of math, but you can skip to the conclusions. All calculations assume that the TEPCO measurements reported in Table 1 are correct.

First we calculate the number of Cl-38 nuclei that are present that would explain the observed radioactivity. The half-life of Cl-38 = 37.24 min which corresponds to a decay constant of $\lambda_{38}$=0.00031021 $s^{-1}$. So that: $\frac{dN_{38}}{dt} = -\lambda_{38} N_{38}$ where, $\frac{dN_{38}}{dt} = 1.6e6\ \ s^{-1}$ and $N_{38}$ =5.16e9 Cl-38 nuclei. This means that the activity measured is consistent with the production of 5.16e9 Cl-38 nuclei. The next question is how much Cl-37 was there present in the seawater in the first place? The mass of chlorine in seawater is 19345 mg/kg = 19.345 g Cl /kg [3]. Also, the fraction of Cl-37 in natural Cl is =24.23 % (see Table 2 below).

| Isotope | Molar Mass | % |
|---|---|---|
| Cl-35 | 34.9688527 | 75.77 |
| Cl-37 | 36.9659026 | 24.23 |

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[2] See: http://www.nisa.meti.go.jp/english/files/en20110325-6.pdf

[3] See: http://www.seafriends.org.nz/oceano/seawater.htm

**Table 2: The isotopic abundance and molar mass of chlorine**

The mass of Cl-37 can then be found to be 25% (we must account for the difference in molar mass of the two isotopes it is a very small difference but adjusts the fraction Cl-38 by mass to be 25%) of 19.345 g Cl /kg = 4.89 g Cl-37/kg. Using Avogadro's number we can calculate the total number of Cl-37 nuclei/ g of seawater to be $N_{37}$ =7.96e19.

We now know that $N_{37}$ =7.96e19 Cl-37 nuclei/g of seawater and we observed that 5.16e9 of these have been converted to Cl-38. The question then becomes what flux could have produced this many Cl-38 nuclei?

We now assume Cl-38 was produced as the seawater was being circulated through the fuel. What is the flux of neutrons we need to produce the observed $N_{38}$?

Since Cl-38 is radioactive with a decay constant given by $\lambda_{38}$ the rate of change of the number of Cl-38 nuclei is given by:

$$\frac{dN_{38}}{dt} = \phi\, \sigma_{(\gamma,n)} N_{37} - \lambda_{38} N_{38}$$

This is the familiar equation of series decay where one isotope is being produced and at the same time is decaying. This equation can be easily solved (see for example I. Kaplan, Nuclear Physics, 1958, p 463.):

$$N_{38}(t) = \phi \left[\frac{\sigma_{(\gamma,n)} N_{37}}{\lambda_{38}}\right] \left(1 - e^{-\lambda_{38} t}\right)$$

Where, $\phi$ is the flux in n/cm$^2$.s, and $\sigma_{(\gamma,n)}$=383.7 mb is the radiative capture cross-section which would result in the production of Cl-38 at the Maxwellian distribution average temperature. Note that the thermal neutron cross-section is not very different at 432 mb so the similar results would be obtained if we assumed that all the neutrons are thermalized.

Now, we know that after activation we produced $N_{38}(t)$ =5.16e9 Cl-38/cm3, so we let t=T, the time when activation stopped so that $N_{38}(T) = 5.16e9$ nuclei/cm$^3$. We also know the value of the factor $\left[\frac{\sigma_{(\gamma,n)} N_{37}}{\lambda_{38}}\right]$=0.098445192.

So that the flux can be expressed very simply as a function of irradiation time T:

$$\phi = \frac{5.2415e10}{\left(1 - e^{-\lambda_{38} T}\right)}$$

We assume that the production of Cl-38 started with the deliberate introduction of seawater on March 23rd (according to TEPCO press briefing[4]) into reactor #1. Therefore, since the measurement appears to have been done on March 25th it means we have a maximum activation time of 2 days. In fact, we really have two regions of flux that are significant. The first region is where the denominator is < 1 (corresponding to activation time T<4 h) where the flux changes appreciably with a change in activation time, and the second region is where the denominator ~1 which happens when T>0.4 days.

A lower limit in the flux is set when T is long (ie. > 0.5 d) so that the denominator approaches unity. We call this flux ($\phi$=5.241e10 n/cm2.s) and it is the lower limit of the flux that could have produced the Cl-38 nuclei radioactivity observed.

## What might have caused the concentration of Cl-38?

The first possible explanation to consider is that the seawater was circulated among the core intercepting neutrons from natural spontaneous fission of the used nuclear fuel. The second possible explanation to consider is localized criticalities.

Recall that nuclear fuel changes its isotopic composition upon irradiation in a reactor. This is the reason why we are concerned about plutonium production in nuclear reactors from a nonproliferation point of view. We investigated this by calculating the number of spontaneous fissions from a typical BWR with 4% enriched fuel after 45 MWdth/kg burnup (see IAEA-TECDOC-1535, pg. 74). The inventory we get for 1 metric ton fuel for the primary neutron producing isotopes are shown in Table 2.

| Isotope | Isotope Inventory $M_{iso}$=# g/MTHM | Number of Isotope Nuclei/g =$\rho_{iso}$ | $Br_{SF}$ = SF Isotope Branching Ratio (%) | Half- Life =$T_{1/2}$ in years | Decay Constant of isotope = $\lambda_{iso}$ in $s^{-1}$ | Number of neutrons produced/sec |
|---|---|---|---|---|---|---|
| Pu-238 | 2.66E+02 | 2.53E+21 | 1.85E-07 | 8.77E+01 | 2.51E-10 | 9.35E+05 |
| Pu-240 | 2.57E+03 | 2.51E+21 | 5.75E-06 | 6.56E+03 | 3.35E-12 | 3.72E+06 |
| Pu-242 | 6.79E+02 | 2.49E+21 | 5.54E-04 | 3.73E+05 | 5.89E-14 | 1.65E+06 |
| Cm-242 | 2.02E+01 | 2.49E+21 | 6.37E-06 | 1.63E+02 | 1.35E-10 | 1.29E+06 |
| Cm-244 | 5.26E+01 | 2.47E+21 | 1.37E-04 | 1.81E+01 | 1.21E-09 | 6.49E+08 |

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[4] See: http://www.tepco.co.jp/en/press/corp-com/release/11032609-e.html. “At approximately 2:30 am on March 23rd, seawater was started to be injected to the nuclear reactor through the feed water system.”

**Table 2: The isotopic inventory, nuclei/g, branching ratio for spontaneous fission, half-life, and decay constant for different neutron producing isotopes present in spent nuclear fuel. The largest flux comes from even Pu isotopes and Cm. Note: MTHM= metric ton heavy metal and refers to the active component of the fuel SF= spontaneous fission. Isotopic inventory obtained from IAEA-TECDOC-1535, pg 74.**

The neutron production rate from spontaneous fission rate can be calculated for each isotope by summing the contribution of spontaneous fission by each isotope.

$\frac{dN_n}{dt} = \sum_{i=1}^{iso} \lambda_i M_i \rho_i (\frac{Br_{i,SF}}{100})\ \nu_i$; where $\nu$ is the average number of neutrons. We will assume that all neutrons will be thermalized and about 3 neutrons are produced per fission. The total neutron production rate found is 6.56e8 neutrons/sec for 1 metric ton. However, the full mass of fuel in the core is 69 metric tons. Therefore, the source strength of the core due to spontaneous fission is 4.53e10 neutrons/sec.

At this rate we can use the formula for simultaneous production and decay to calculate the number of Cl-38 produced as a function of time.

$$N_{38}(t) = \phi \left[\frac{\sigma_{(\gamma,n)} N_{37}}{\lambda_{38}}\right] \left(1 - e^{-\lambda_{38} t}\right)$$

However, knowing the source strength does not tell us the flux. To determine the flux we have to know the configuration of the fuel with respect to the seawater. This is difficult to determine given the little information that is known about the status of reactor #1. To get an estimate we will assume several scenarios:

1) Scenario 1: The fuel has melted, and has assembled in the bottom of the inpedestal and expedestal regions of the reactor vessel (the “bulb”) as shown in Figure 1. The seawater is assumed to come into contact and cover the melting fuel as shown in Figure 2. This scenario was predicted in C. R.Hyman’s report (“Contain calculation of debris conditions adjacent to the BWR Mark I drywell shell during the later phases of a severe accident”, Nucl. Engin. and Design., 121, 1990, p 379-393.).

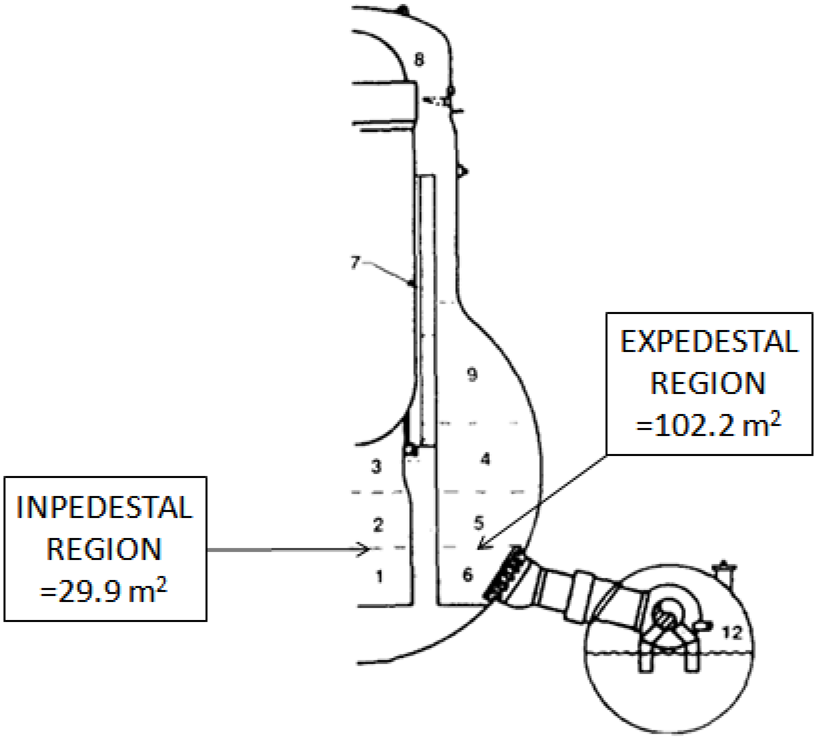

**Figure 1: Figure showing the pressure vessel and Mark I containment and the inpedestal and expedestal regions which are the regions where it is assumed that the melted fuel would assemble (Figure adapted from C. R. Hyman, Nucl. Eng. and Des., 121, 1990, Fig 2).**

The flux is calculated by assuming a simple slab geometry as is shown in Figure 2 where the neutron source is assumed to rest underneath the layer of water and half of the neutrons are expected to go on average up and half down. The flux is defined by the number of neutrons that intersect a 1 cm$^2$ area which is half the source strength divided by the area of the slab. We assume that the slab area is the sum of the inpedestal and expedestal areas (according to C. R. Hyman op cit).

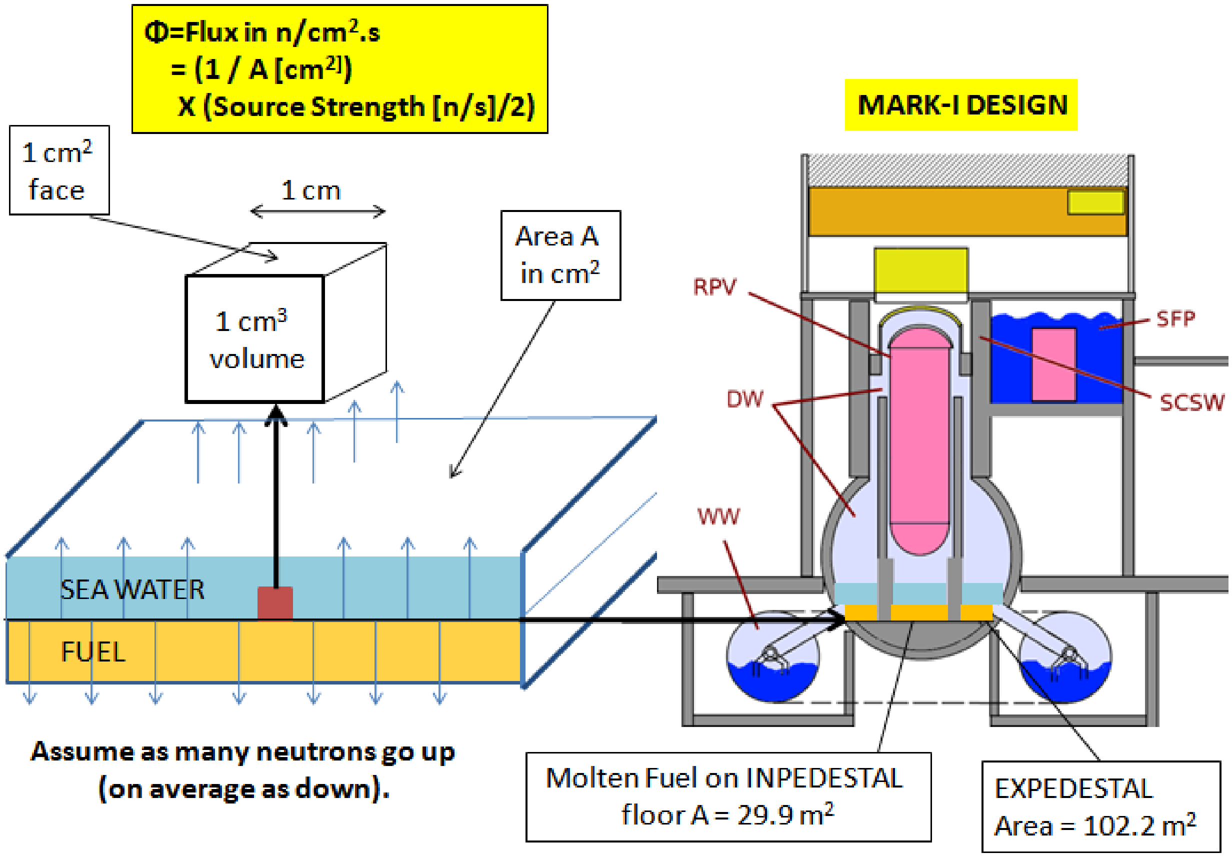

**Figure 2: Figure showing how the neutron flux is calculated. We assume a simple slab geometry where the seawater covers the fuel and ½ of the neutrons source travels up and half travels down. The flux intersecting the neutrons is the ratio of the area of 1 cm$^3$ to the area of the slab which is assumed to be the sum of the inpedestal and expedestal areas (illustration of Mark-I adapted from Wikipedia).**

We use the familiar equation from before and find that:

$$N_{38}(t) = \phi\left[\frac{\sigma_{(\gamma,n)}N_{37}}{\lambda_{38}}\right]\left(1 - e^{-\lambda_{38}t}\right)$$

$$N_{38}(T) = 1.71e4\left(1 - e^{-\lambda_{38}T}\right)$$

Now, the maximum number of Cl-38 nuclei are produced when T is long and is maximum at 1.71e4Cl-38 nuclei. As time increases as many Cl-38 nuclei are produced as decay and an equilibrium is established. So assuming that the seawater covers the fuel in the floor of the "bulb" it is clear that in this proposed scenario not enough neutrons are produced to account for a 1.6 MBq Cl-38 radioactivity.

2) Scenario 2: The second scenario is if the fuel partially melts but the core leaves crevices through which the seawater can flow. In this case the 1 $cm^3$ water is assumed to be surrounded by a homogeneous neutron emitting fuel.

The flux is calculated by calculating the ratio of the 1 $cm^3$ as compared to the complete volume of the fuel. We know that the total mass of the fuel is 69 metric tons and the density of the fuel changes considerably at high temperatures (see Figure 3).

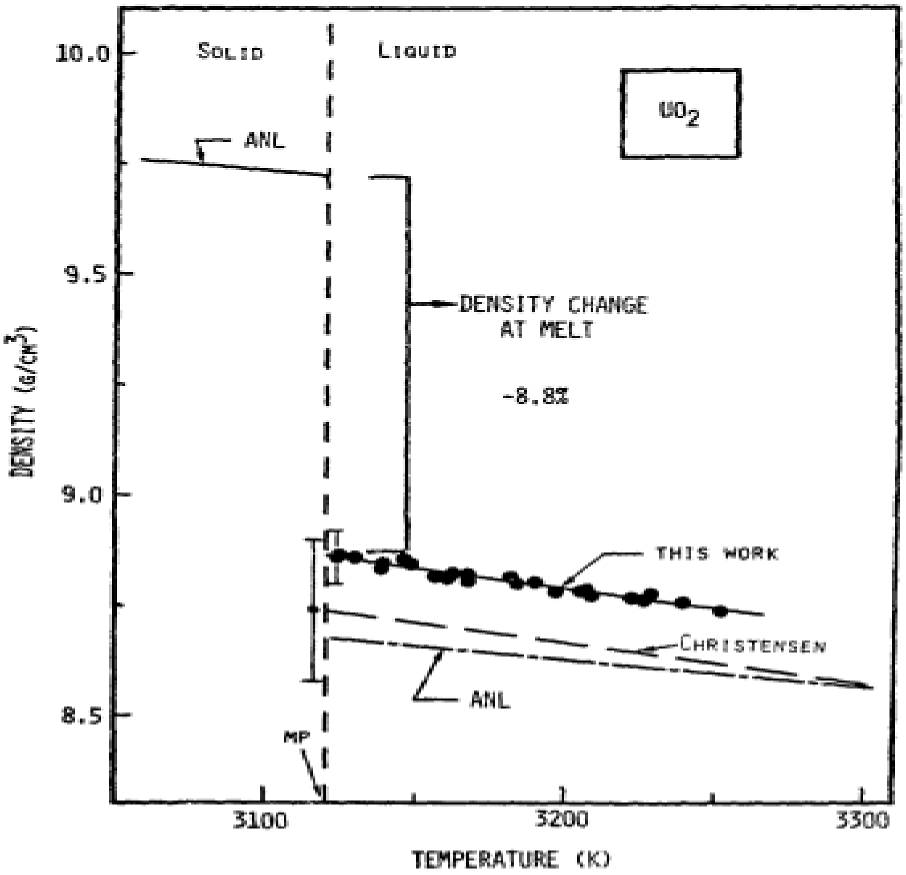

**Figure 3: Figure showing how the $UO_2$ fuel density changes as a function of temperature (Figure taken from W.D. Drotning, Thermal Expansion of Molten Uranium Dioxide,CONF-81069601).**

We assume that the density is approximately 8.86 $g/cm^3$ at temperatures exceeding 3120 K so that the volume occupied by the fuel is 6.77e6 $cm^3$. Therefore the fraction of the flux that is intercepted by the 1 $cm^3$ volume is 1.48e-7. We assume that the flux through the 1 $cm^3$ volume is also proportional to this fraction. Therefore, the flux is assumed to be=4.53e10*1.48e-7=6703 $n/cm^2.s.$ and the number of Cl-38 nuclei can be calculated as before:

$$N_{38}(t) = \phi \left[\frac{\sigma_{(\gamma,n)} N_{37}}{\lambda_{38}}\right] \left(1 - e^{-\lambda_{38} t}\right)$$

$$N_{38}(T) = 658.8 \left(1 - e^{-\lambda_{38} T}\right)$$

In this scenario we find that the number of Cl-38 nuclei reaches a maximum at $<7\text{x}10^2$ which again is certainly not enough to explain the observed Cl-38 radioactivity of 1.6 MBq. So this scenario is just as implausible as scenario 1 above, making it obvious that spontaneous fission cannot account for the reported concentration of Cl-38.

To summarize: We can compare the calculated number of Cl-38 nuclei determined from the measured Cl-38 radioactivity, to the upper limit of the number of Cl-38 nuclei assuming the two scenarios and express this as a percentage. We find that the scenario where the molten fuel pours into the inpedestal and expedestal areas suggests a Cl-38 number that is 3.3e-4% of what is needed to explain the observed Cl-38 radioactivity. Also, the second scenario where a small 1 $\text{cm}^3$ sample is embedded into a uniform neutron flux suggests a Cl-38 number which is even smaller at 1.3e-5%. Barring significant information that we do not possess, neither spontaneous fission and seawater option explains the observed radioactivity.

## Conclusions

So we are left with the uncomfortable realization that the cause of the Cl-38 concentrations is not due to seawater intercepting neutrons from natural spontaneous fission of the used nuclear fuel. There has to be another reason.

Assuming that the TEPCO measurements are correct, the results of this analysis seem to indicate that we cannot discount the possibility that there was another strong neutron source during the time that the workers were sending seawater into the core of reactor #1. However, since we don’t know the details of the configuration of the core and how the seawater came in contact with the fuel it is difficult to be certain. Given these uncertainties it is nonetheless important for TEPCO to be aware of the possibility of transient criticalities when work is being done; otherwise workers would be in considerably greater danger than they already are when trying to working to contain the situation. A transient criticality could explain the observed 13 “neutron beams” reported by Kyodo news agency (see above). This analysis is not a definitive proof but it does mean that we cannot rule localized criticality out and the workers should take the necessary precautions.