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What Caused the High Cl-38 Radioactivity in the Fukushima Daiichi Reactor #1? 福島第一原発の１号機（タービン建屋）から検出された高濃度放射性塩素３８の原因は何か？ •Japanese text available

Arjun Makhijani, Ferenc Dalnoki-Veress

What Caused the High Cl-38 Radioactivity in the Fukushima Daiichi Reactor #1?[1]

A bilingual Japanese-English text is available here.

F. Dalnoki-Veress with an introduction by Arjun Makhijani

Important article update April 23, 2011 A Japanese translation by Kyoko Selden of this update is available here.

In its press release of April 20, TEPCO has retracted the Cl-38 radioactivity concentration measurement (1.6 MBq/mL) for the seawater used to cool reactor #1that it had issued on March 25, saying that it was "below minimum detectable density". Based on this original measurement, we had determined that the value was too high to be explained without invoking the possibility of inadvertent, transient criticalities. We are pleased that TEPCO has retracted this result and has set out to improve its analysis protocol as described in the same press release. But we would appreciate further explanation of why previous results were simply retracted with inadequate categorization and explanation of the errors, as in the TEPCO press release. (The Cl-38 reading was changed on April 20th from 1.6MBq to a value "below detection limit" with the following explanation: "Identification and determination of radioactivity density were conducted based on main peaks.") For example, the main gamma lines of Cl-38 are at 1.64 MeV and 2.16 MeV. What lines did these interfere with that required a downscaling of 6 orders of magnitude? If the count rate could not be attributed to Cl-38 what isotope had a count rate equivalent to 1.6 MBq/mL?

While appreciating the steps that TEPCO has taken since the April 4th NISA reprimand, we recommend further rigor in isotopic measurement protocol and timely reporting of results. Otherwise, public trust in the important measurements that TEPCO is making will further erode. We therefore recommend that TEPCO take the following steps:

1) Release full spectra data (not just a number)

2) Release the time/date sample was taken

3) Release the time/date sample was measured including counting time and dead time

4) Repeat measurements at different times of the day

5) Please measure other isotopes of interest (such as Te-129, which was retracted by TEPCO on April 20th as well), even if they are below the detection limit

6) If retractions are necessary due to an honest mistake, please provide full explanation of the mistake

7) If third-party, independent analyses are done, please state the name of the analyst/lab that has cross-checked TEPCO's interpretation of the results

TEPCO/NISA and the Japanese government have a monumental task ahead of them and important decisions will be based on measurement results. Therefore, it is important that rigorous protocol be followed both in analysis and in communicating the results. F. Dalnoki-Veress

This is a first for The Asia-Pacific Journal: publication of a technical scientific paper addressing critical issues pertaining to the leakage of radioactive water at the Fukushima reactors. Our goal is to make this information available to the Japanese and international scientific communities, to Japanese government authorities, and TEPCO as they address the formidable issues of cleanup and safety. But we also believe that the information is of importance to informed citizens and the press in the face of further dangers that have gone unmentioned not only in government statements, but also in the press. Arjun Makhijani's introduction provides a lucid explanation of the problem and the issues, followed by F. Dalnoki-Veress's paper. Asia-Pacific Journal

Introduction by Arjun Makhijani

The presence of highly radioactive water in three turbine buildings at the Fukushima Daiichi nuclear plant is widely understood to be from the damaged fuel rods in the reactors. This has rightly raised concerns because it indicates several problems including extensive fuel damage and leaks in the piping system. Less attention has been paid to the presence of a very short-lived radionuclide, chlorine-38, in the water in the turbine building of Unit 1. The following paper evaluates whether its presence provides evidence of a serious problem – one or more unintended chain reactions (technically: unintended criticalities) – in the reactor. Such chain reactions create bursts of fission products and energy, both of which could cause further damage and aggravate working conditions that are already very difficult.

Chlorine-38, which has a half-life of only 37 minutes, is created when stable chlorine-37, which is about one-fourth of the chlorine in salt, absorbs a neutron. Since seawater has been used to cool, there is now a large amount of salt – thousands of kilograms – in all three reactors. Now, if a reactor is truly shut down, there is only one significant source of neutrons, namely, the spontaneous fission of some heavy metals which are created when the reactor is working and remain present in the reactor fuel. The most important ones are two isotopes of plutonium and two of curium. But if accidental chain reactions are occurring, it means that the efforts to completely shut down the reactor by mixing boron with the seawater have not completely succeeded. Periodic criticalities, or even a single accidental one, would mean that highly radioactive fission and activation products are being (or have been) created at least in Unit 1 since it was shut down. It would also mean that one or more intense bursts of neutrons, which cause heavy radiation damage to people, have occurred and possibly could occur again, unless the mechanism is understood and measures taken to prevent it. Measures would also need to be taken to protect workers and to measure potential neutron and gamma radiation exposure.

This paper examines whether spontaneous fission alone could be responsible for the chlorine-38 found in the water of the turbine building of Unit 1. If that could be the only explanation, there would be less to be concerned about. However, the analysis indicates that it is quite unlikely that spontaneous fission is the sole or even the main explanation for the measured concentration of chlorine-38. Presuming the reported measurements are correct, this leaves only one other explanation – one or more unintended chain reactions. This paper is presented in the spirit of encouraging discussion of whether further safety measures might be needed, and whether supplementary measures to bring the reactors under control should be considered. It is also presented as a preliminary analysis for scientific discussion of a terrible and technically challenging nuclear crisis at the Fukushima Daiichi plant.

Arjun Makhijani March 30, 2011

I have been consumed over the last few weeks by the events unfolding in Japan. I keep 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 appeared on the Kyodo news website (relevant text italicized below for emphasis) and I did the following analysis. FD-V March 30, 2011

"Neutron beam observed 13 times at crippled Fukushima nuke plant

TOKYO, March 23, Kyodo

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.*

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.

The utility firm said it will measure uranium and plutonium, which could emit a neutron beam, as well.

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.

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."

==Kyodo News

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 why these measurements were taken so late in the crisis (or why the information was released so late) is unclear at present.

Radioactive Nuclide	Concentration (Bq/cm^3)
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 radioactivity from a sample taken in the turbine building of reactor #1[2]

The measured levels of Cesium and Iodine, Cs-137 and 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, the fact that it was there at all, and in such high concentration, 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 λ_{38} = 0.00031021 s^{-1}. So that: $dN_{38}/dt = -\lambda_{38}N_{38}$ where, 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 present in the seawater in the first place? The mass of chlorine in seawater is 19345 mg/kg = 19.345g 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

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 it adjusts the fraction Cl-38 by mass to be 25%) of 19.345 g Cl/kg = 4.89g 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 λ_{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, ϕ is the flux in n/cm^2.s, and $\sigma_{(\gamma,n)}$ = 383.7mb 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 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 $\sigma_{(\gamma,n)}N_{37}/\lambda_{38}$ = 0.098445192.

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

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

We assume that the production of Cl-38 started with the deliberate introduction of seawater on March 23rd (according to the TEPCO press briefing[4]) into reactor #1. Therefore, since the measurement appears to have been done on March 25th it means that 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 T0.4 days.

A lower limit in the flux is set when T is long (i.e. > 0.5 d) so that the denominator approaches unity. We call this flux (ϕ = 5.241e10 n/cm^2.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 =ρ_{iso}	Br_{SF} = SF Isotope Branching Ratio (%)	Half- Life =$T_{1/2}$ in years	Decay Constant of isotope = λ_{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

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 can be calculated for each isotope by summing the contribution of spontaneous fission by each isotope.

$(dN\_n)/dt=\Sigma_{i=1:iso}[\lambda_i M_i \rho_i (Br_{i,SF})/100)\nu_i]$; where ν 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 consider several hypothetical 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.).

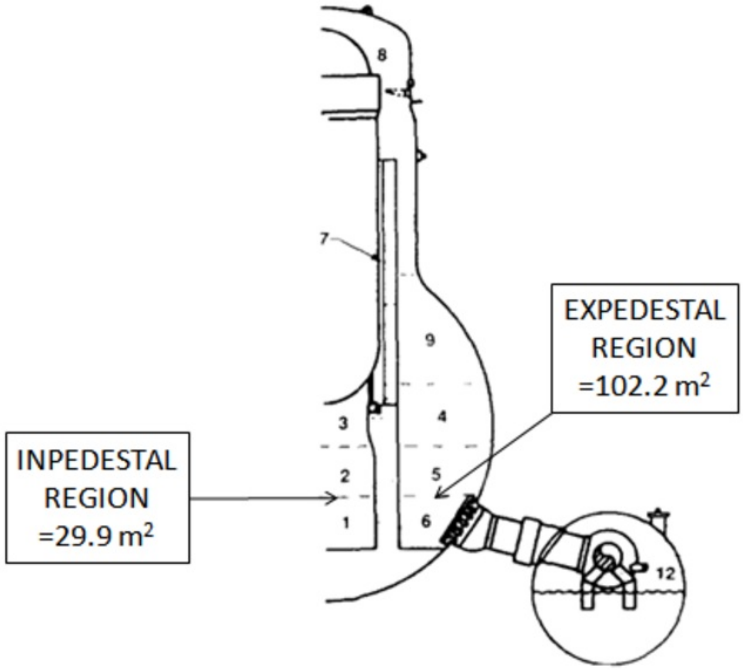

Figure 1: Figure showing the pressure vessel and Mark I containment and the inpedestal and expedestal regions which are the regions where it is assumed that the melted fuel would assemble (Figure adapted from C. R. Hyman, Nucl. Eng. and Des., 121, 1990, Fig 2).

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).

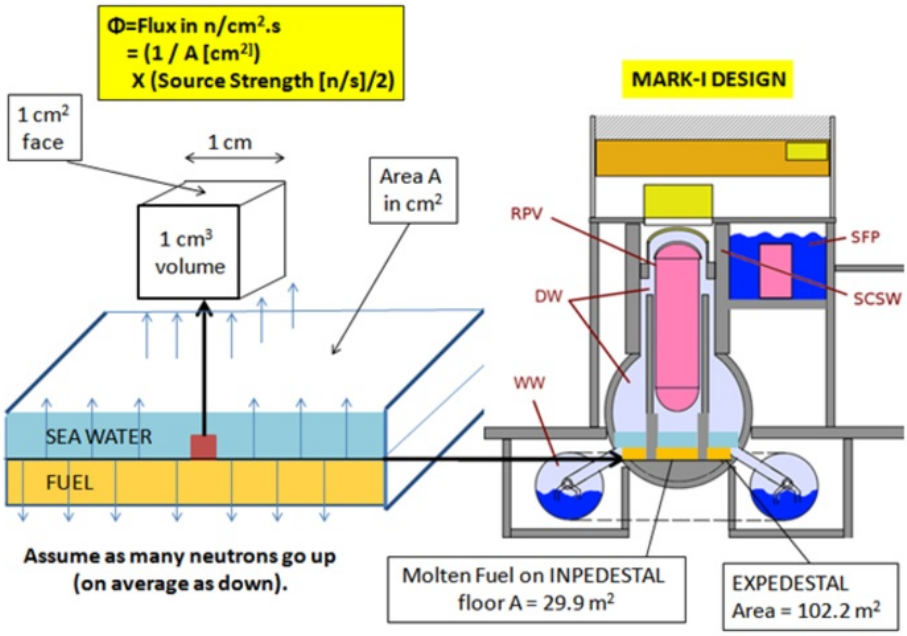

Figure 2: Figure showing how the neutron flux is calculated. We assume a simple slab geometry where the seawater covers the fuel and ½ of the neutrons source travels up and half travels down. The flux intersecting the neutrons is the ratio of the area of 1 cm^3 to the area of the slab which is assumed 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).

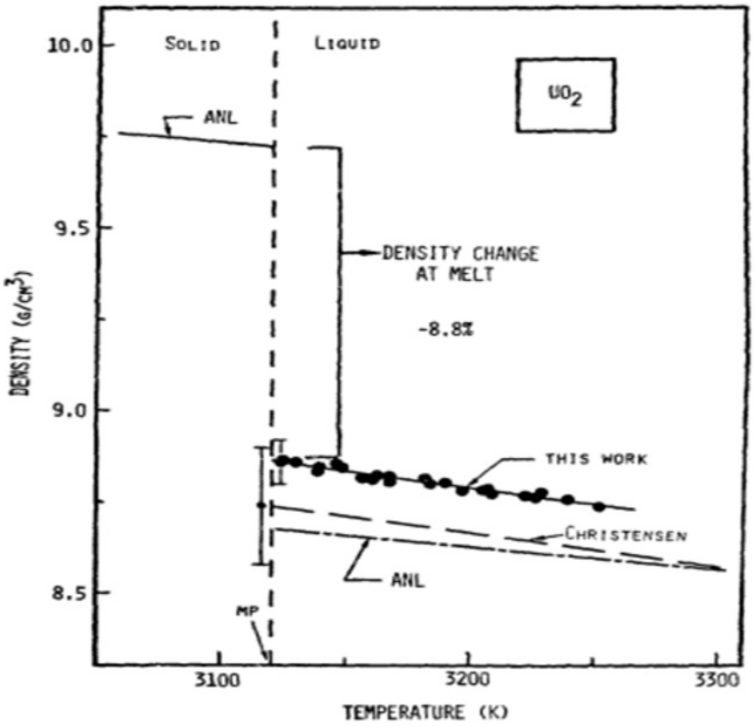

Figure 3: Figure showing how the UO2 fuel density changes as a function of temperature (Figure taken from W.D. Drotning, Thermal Expansion of Molten Uranium Dioxide, CONF-81069601).

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 cm3 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 $7x10^2$ which again is certainly not enough to explain the observed Cl-38 radioactivity of 1.6 MBq.So this scenariois 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 in whicha small 1 cm3 sample is embedded in 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 seawater intercepting neutrons fromnatural spontaneous fission of the used nuclear fuel. There has to be another reason.

Assuming that the TEPCO measurements are correct,this analysis seems 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, without knowing 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 out localized criticality and TEPCO should assure that the workers take the necessary precautions.

For a discussion of the article at *Nature* see Jeff Brumfiel, Japan faces more than a decade of nuclear clean-up.

For further discussion of related issues see Fukushima Physicists Forum.

Ferenc Dalnoki-Veress Response to Comments (Update)

Thanks everyone for all your excellent comments. I wrote this paper because I wanted to rule out criticality in reactor #1 and with the scenarios that I invoked came to the uncomfortable conclusion that I could not. A colleague of mine (Patricia Lewis) had been wondering about the temperatures of the molten core and whether we could any longer think of the integrity of the fuel. We had investigated already the melting point of reactor grade steel, the effect of heating on volumes and the possible viscosities of the molten fuel and the reported 13 "beams" of neutrons also added into our concerns; so we were aware that transient critical masses could not be ruled out. In addition, we were concerned about the basing the entire analysis on one reported measurement of Cl-38 but that amount of Cl-38 activity would have been a red flag to any physicist and I did not have a reason at the time to dispute it. A healthy type of skepticism in all of the measurement numbers coming from Fukushima is absolutely necessary, but, I share *Peter Raffaele's* statement "What can go wrong will. And if you are not paranoid you are naïve". What is the probability that the reactor will not go critical again, if only for an instant? If your answer to that question is well it's not zero, then you are where I am. In emergency preparedness we have to stretch our mind a bit further than we might want to. We have to imagine the impossible even if it goes against every instinct until it is ruled out absolutely.

Can We Believe the Cl-38 Number?

TEPCO has made many measurement errors, from mixing up I-131 and I-134 to adjusting the numbers for Tc-99m etc. So it is absolutely reasonable to question whether TEPCO correctly measured the Cl-38. *Red_Blue* is correct that the start time of seawater injection was March 12th, not March 23rd as I surmised. However, this does not change the analysis much because of the equilibrium that is established due to production and decay. I whole-heartedly agree with *Red_Blue* and many others that conclusions could be drawn when TEPCO re-samples and measures the water. Although, that would only be true if there have been further high neutron flux incidents. If these had been transient criticalities over a given period of time, the short half-life of Cl-38 would mean that resampling would not necessarily help us.

I also agree that a valid criticism of my analysis is the assumption that the seawater is pumped through the system in a continuous flow with a salt concentration identical to that of seawater. However, we can relax this assumption and take the maximum salt concentration (40 g/mL) which is a factor of 20 more salt/mL than my assumption. However, this is not large enough to account for the large Cl-38 concentration of 1.6 MBq/cm3. We are talking six orders of magnitude difference. That is quite a significant difference.

I agree with *Eve* that TEPCO should have also monitored the Na-24 1.368 MeV and 2.754 MeV gammas. I don't presume to know why they didn't or why they didn't redo the Cl-38

measurements considering that these should have triggered an alarm. I agree with *JamesL* that it would have been nice to be able to time-correlate the reported observation of the "neutron bursts" with other measurements and I would urge TEPCO to publish them. I would also recommend TEPCO to publish the time when all samples are taken as well as when they are measured so the time difference can be accounted for. However, I disagree with *Red_Blue* and *Old Jim Hardy* that there are many fission product gammas that the Cl-38 could have been confused with. The spectra for the two isotopes are very distinct: Cl-38 has two gammas a 1.64 MeV and 2.17 MeV whereas I-134 has prominent emissions at 0.847 MeV and 0.884 MeV and no significant gammas at higher energies (see INL Spectra Catalog at http://www.inl.gov/gammaray/catalogs/ge/catalog_ge.shtml) . Beta spectroscopy is a little bit more complicated but Cl-38 has a 56% probability high 4.92 Q-value whereas I-134 has a complicated scheme with the highest beta endpoint at 2.2 MeV and most betas < 1.2 MeV. However, I come back to where I started: if you are absolutely certain that the reactor won't go critical then by all means dismiss the Cl-38 number. However, if you think the probability is non-zero then it is prudent to consider all possibilities since the consequences could be serious.

With respect to my esteemed colleague, I have to disagree with Dr. Jim Rushton of Oak Ridge National Laboratory's assertion "Even if they [inadvertent criticalities] did occur briefly, they would not add much radioactivity or heat beyond what workers are already dealing with from the radioactive material that accumulated when the reactors were running at full power (see New Scientist at http://www.newscientist.com/article/dn20322-are-nuclear-reactions-restarting-at-fukushima.html) . " Neutrons affect human tissue so very differently than gammas. We know from other transient (and non-explosive) criticality accidents that people have died very quickly from large neutron bursts – even recently in Japan, there have been fatal consequences of criticality accidents.

Consequences of Possible "Inadvertent Criticalities"

So now let's assume that "inadvertent criticalities" do occur in reactor #1. What could the consequences be and how might they manifest themselves? Many of you brain-stormed on this topic and I will reserve comments and summarize what you have all said across different blogs and fora. *Damfuzzy* reminds us of the fact that new BWR fuel assemblies are located on the refueling floor which at least in one Fukushima reactor is exposed to the atmosphere and may have been disrupted from explosions. He suggests the possibility that the "spontaneous fires" that have been reported may be due to criticality excursions. *Roger T Crouch* postulates that rod-collapse could lead to loose material where vibrations, water flow and structural collapse of the assembly grid and control rod systems could result in a self-sustaining chain reaction. Many of you have worried about the possibility of starting a chain reaction that becomes super-critical rather than turning itself off due to negative feedback effects. The main explanation for not going super critical is that – if transient criticalities have taken place – they are probably due to small globules of fissile material that is expanding and moving in a viscous soup of molten metals and oxides, thus continually changing their mean free path.

What Must be Done Right Now?

The purpose of the Cl-38 calculation was to exclude the possibility that an "inadvertent criticality" can occur at the Fukushima reactor #1 which I was unable to do. Therefore, it is prudent that TEPCO takes seriously the possibility of criticality excursions and monitors the neutron flux with independent neutron detectors close to the core. A sudden increase in the neutron flux would be immediately measurable above the background due to the spontaneous fission of the different actinides in the fuel. TEPCO must continue to mix Boric acid with the fresh cooling water to ensure that no criticality excursions can occur especially in reactor #1. All efforts must be made to protect the workers when the probability for "inadvertent criticalities" are non-zero. I suggest that TEPCO takes the following actions:

- Install a neutron detector to monitor the core of Fukushima Daiichi reactor #1
- Keep mixing neutron absorbers with the cooling water for cooling reactor cores and spent nuclear fuel ponds
- Give complete gamma spectra rather than just the summaries of the results
- Include not only sampling times but also measurement times for all measurements and repeat measurements to increase confidence in the results

Let's keep the conversation going. We owe it to all those heroic Fukushima Daiichi workers.

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Notes

[1] Thanks go to Dr. Patricia Lewis (CNS, MIIS) and Arjun Makhijani (IEER) for carefully reviewing this memo, and for thoughtful and stimulating discussions. Dr. Lewis may be contacted at patricia.lewis@miis.edu.

[2] Nuclear and Industrial Safety Agency, Ministry of Economy, Trade and Industry, News Release, March 26, 2011.

[3] Dr. J. Floor Anthoni, The Chemical Composition of Seawater (2000, 2006).

[4] Press Release (Mar 26, 2011) TEPCO News, Plant Status of Fukushima Daiichi Nuclear Power Station (as of 8:00 PM Mar 26th): "At approximately 2:30 am on March 23rd, seawater was started to be injected to the nuclear reactor through the feed water system."