XENON DETECTION AT FUKUSHIMA NPP

UNIT 2

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An attempt to confirm the origin of Xe^{135} detected in the Fukushima Daiichi II Nuclear Power Plant in November 2011

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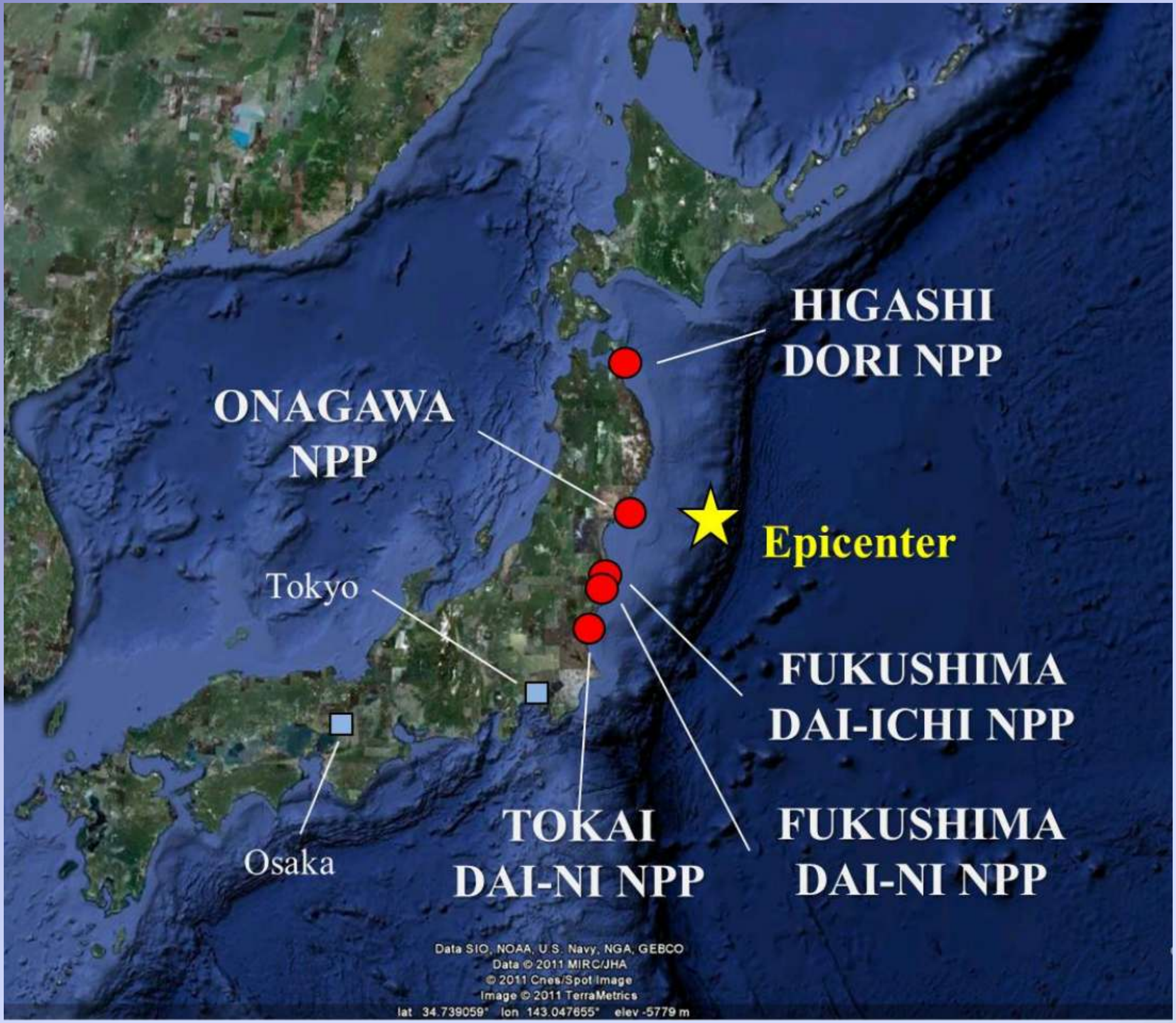
ABSTRACT

The recent detection of ^{135}Xe activity opens doubts about the evidence of nuclear recriticality in Fukushima Daiichi Nuclear Power Plant – Unit 2. The nuclear plant operator, after some investigation, reported that the measured concentration would be consequence from spontaneous fission process from damaged fuel. This work aims to perform an independent calculation to confirm the origin of ^{135}Xe activity measured. The ^{135}Xe activity calculated was compared to the experimental measurements accomplished by TEPCO.

The comparison between the value obtained experimentally and the value calculated in this work, for ^{135}Xe activity, shows that the spontaneous fission process of actinides, presents in the nuclear fuel, was the unique responsible for the production of xenon, since the value of the xenon activity is within the range of uncertainties calculated in this work.

It was also defined in this work a complementary indicator for the assessment of the neutronic condition of the nuclear fuel, based on the ratio between the activities of 135I and ^{135}Xe.

From this result, one can conclude that there is no evidence of recriticality in the damaged reactor.



Extracted from IAEA Report of Expert Mission - 2011

NPP	Unit	Type		Capacity (MW(e))	Status		
		CV** type	Safety system		Before earthquake	After earthquake	After tsunami
Higashi Dori	1	Mark I R	BWR-5	1,100	Outage	Cold Shutdown	Cold Shutdown
Onagawa	1	Mark I	BWR-4	524	Operating	Automatic Scram	Cold Shutdown
	2	Mark I	BWR-5	825	Reactor Start	Automatic Scram	Cold Shutdown
	3	Mark I	BWR-5	825	Operating	Automatic Scram	Cold Shutdown
Fukushima Dai-ichi	1	Mark I	BWR-3	460	Operating	Automatic Scram	Loss of Cooling
	2	Mark I	BWR-4	784	Operating	Automatic Scram	Loss of Cooling
	3	Mark I	BWR-4	784	Operating	Automatic Scram	Loss of Cooling
	4	Mark I	BWR-4	784	Outage	Cold Shutdown	Loss of SFP* cooling
	5	Mark I	BWR-4	784	Outage	Cold Shutdown	Cold Shutdown
	6	Mark II	BWR-5	1,100	Outage	Cold Shutdown	Cold Shutdown
Fukushima Dai-ni	1	Mark II	BWR-5	1,100	Operating	Automatic Scram	Cold Shutdown
	2	Mark II R	BWR-5	1,100	Operating	Automatic Scram	Cold Shutdown
	3	Mark II R	BWR-5	1,100	Operating	Automatic Scram	Cold Shutdown
	4	Mark II R	BWR-5	1,100	Operating	Automatic Scram	Cold Shutdown
Tokai Dai-ni	-	Mark II	BWR-5	1,100	Operating	Automatic Scram	Cold Shutdown

*: Spent Fuel Pool
**: Containment Vessel

Status of NPPs Affected by the Earthquake.
extracted from IAEA Report of Expert Mission - 2011

FUKUSHIMA DAIICHI SITE

Extracted from IAEA Report of Expert Mission - 2011

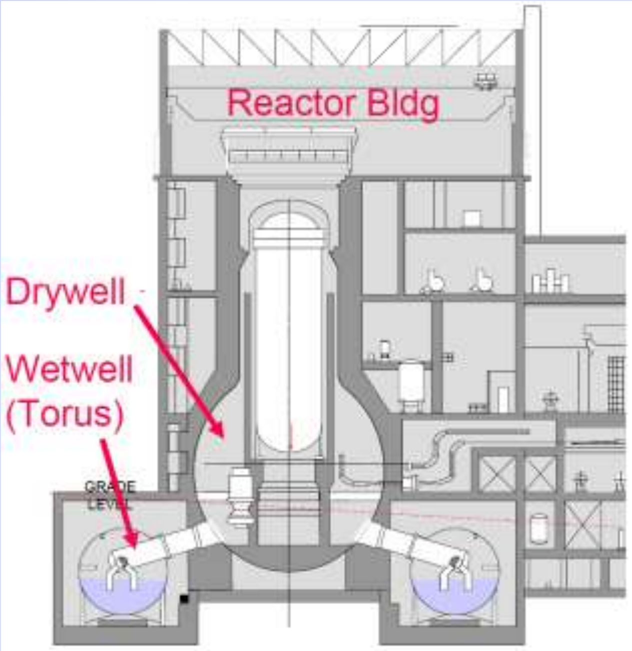
Before

After

Extracted from IAEA Report of Expert Mission - 2011

FUKUSHIMA ACCIDENT
(BRIEF DESCRIPTION)

- On March 11, 2011 a severe earthquake occurred off the north-eastern coast of Japan with the epicenter about 180 km away from the Fukushima Daiichi Nuclear Power Plants generating a tsunami which caused devastating damage over the whole nuclear site, composed by six BWR nuclear reactors.
- The reactors 1, 2 and 3 were in operation.
- The reactors 4, 5 and 6 had been in outage.
- The large acceleration earthquake caused an automatic reactor scram in all operating units.



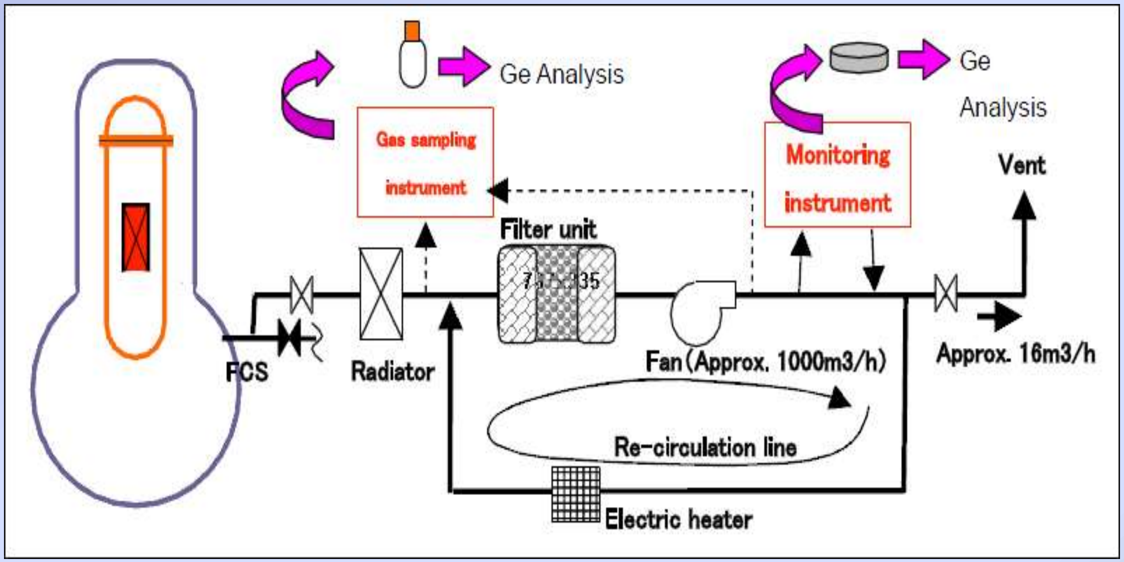
BWR – MK 1

- The external electrical power sources have been lost.
- The emergency diesel generators automatically started, providing energy to the main emergency systems.
- The AC power was lost when a series of tsunamis flooded the emergency diesel generators. (11/12)
- All DC power was lost on Units 1 and 2, while some DC power from batteries remained available on Unit 3.
- Without operational decay heat removal systems, the fuel of nuclear reactors 1, 2 and 3 suffered total/partial melting.
- A large amount of hydrogen was released, in consequence of zirconium and steam water reaction.
- Strong hydrogen explosions damaged the buildings of the Units 1, 3 and 4.
- No evidence of hydrogen explosion was verified in Unit 2.

Source : TEPCO

THE XENON DETECTION IN THE UNIT 2

In order to evaluate the radioactivity into the Unit 2, TEPCO has held experimental measurements, in November, using the PCV Gas Management System. The experimental procedure was based on gas injection inside the primary containment vessel through Flammability Control System (FCS). The gas sampling was collected and the gamma ray analysis was done using Germanium semiconducting detector.



Source : TEPCO

EXPERIMENTAL MEASUREMENTS

- Using the experimental setup, TEPCO obtained the value of 2.7 x 10^{-2} Bq/cm^3 for ^{135}Xe radioactive density (activity concentration).

- The experimental uncertainty of this measurement, however, was not available in the literature.

RECRITICALITY SUSPECTION

- Since the ^{135}Xe has a half-life of about 9 hours, its presence could be interpreted as an eventual occurrence of neutron induced fissions into the nuclear reactor.
- The detection of ^{135}Xe in November opens doubts about the evidence of nuclear recriticality in Fukushima Daiichi Nuclear Power Plant - Unit 2.
- However, even after the injection of borate acid, the same activity of ^{135}Xe was measured. It is discarding, in this way, the possibility of recriticality in Unit 2.
- TEPCO informed to IAEA that the detected xenon was produced from spontaneous fissions of ^{242}Cm and ^{244}Cm.

OBJECTIVE OF THE WORK

- The aim of this work was to perform an independent calculation to confirm the origin of ^{135}Xe activity detected in the Unit 2.

HYPOTHESIS

(i) The contribution of twelve actinides (the more representatives) in the fuel, for the calculation of the fission rate;

(ii) Neutrons reactions in two energy groups (thermal and fast);

(iii) The occurrence of neutron induced fission and spontaneous fissions as well;

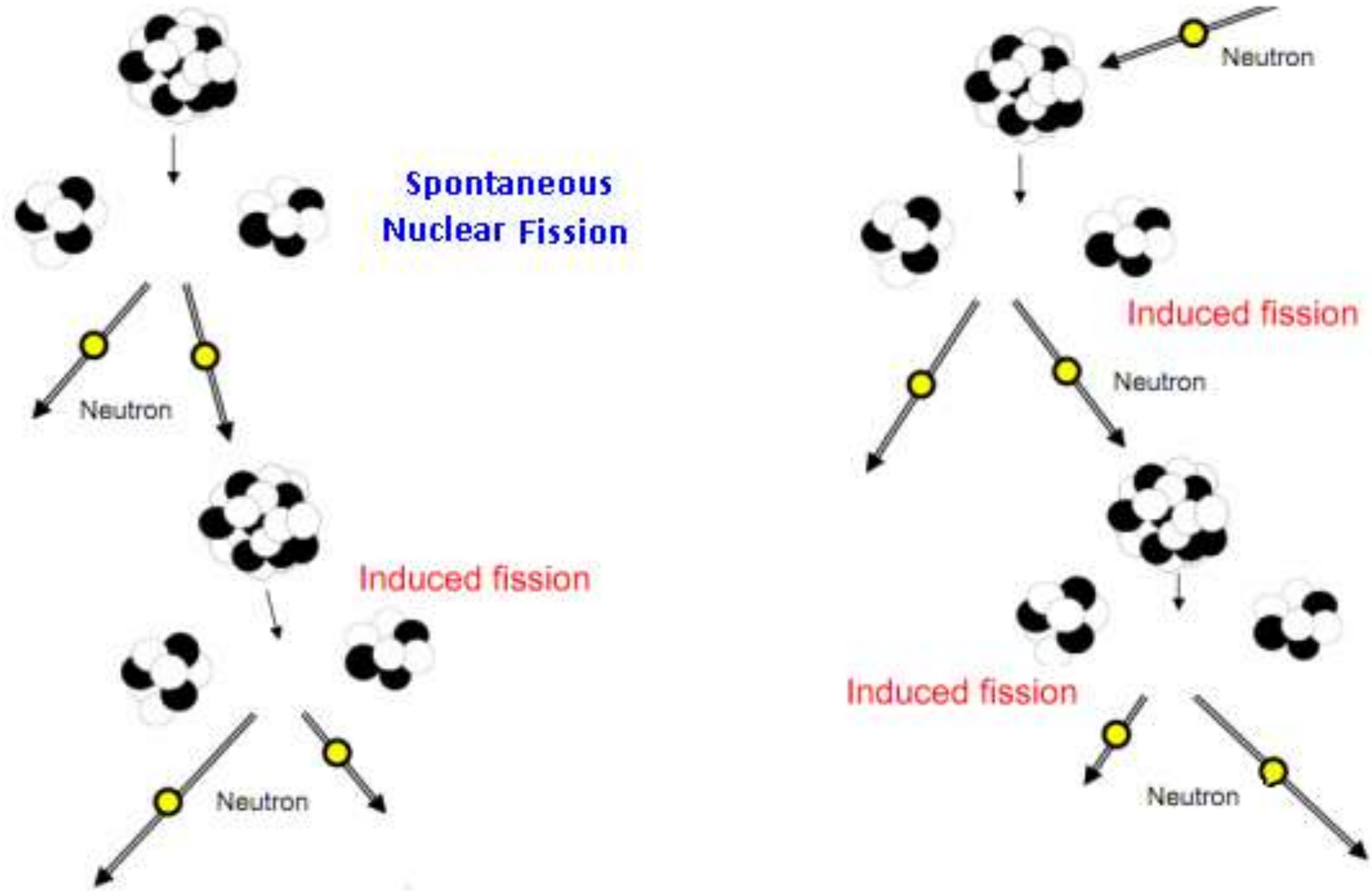
(iv) The occurrence of neutron induced fission due to neutrons coming from spontaneous fission;

(v) The contribution of neutron capture process;

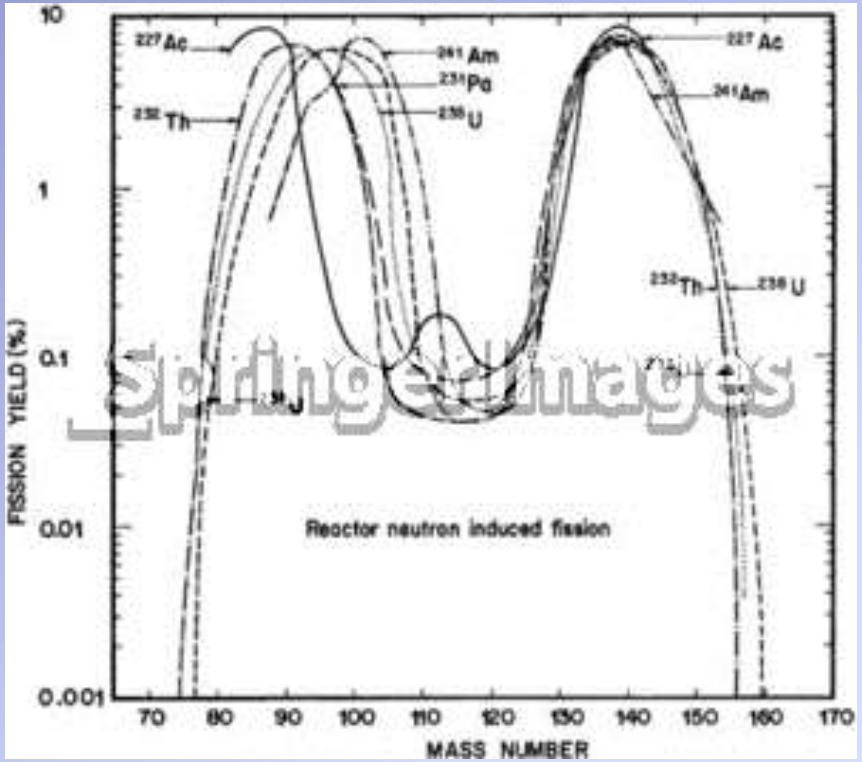
(vi) The contribution of ^{135}I decay for ^{135}Xe production;

(vii) The use of specific yields for each kind of fission (neutron induced and spontaneous);

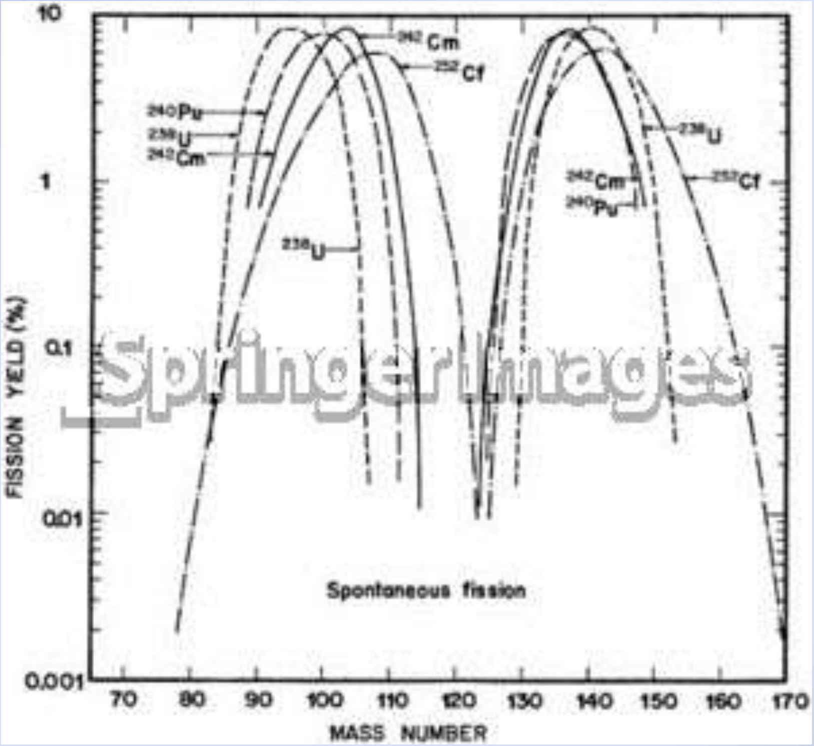
(viii) The total release of ^{135}Xe produced in the damaged fuel (Phébus Project – 2006).


Spontaneous
Nuclear Fission
Neutron
Induced fission
Neutron
Neutron
Induced fission
Neutron
Induced fission
Neutron

SPECIFIC YIELDS
(NEUTRON INDUCED AND SPONTANEOUS FISSIONS)



Mass-yield curves in the reactor-neutron induced fission of ^{227}Ac, ^{232}Th, ^{231}Pa, ^{235}U, ^{238}U, and ^{241}Am; from (von Gunten 1969).



Mass-yield curves for the spontaneous fission of ^{238}U, ^{240}Pu, ^{242}Cm, and ^{252}Cf; from (von Gunten 1969).

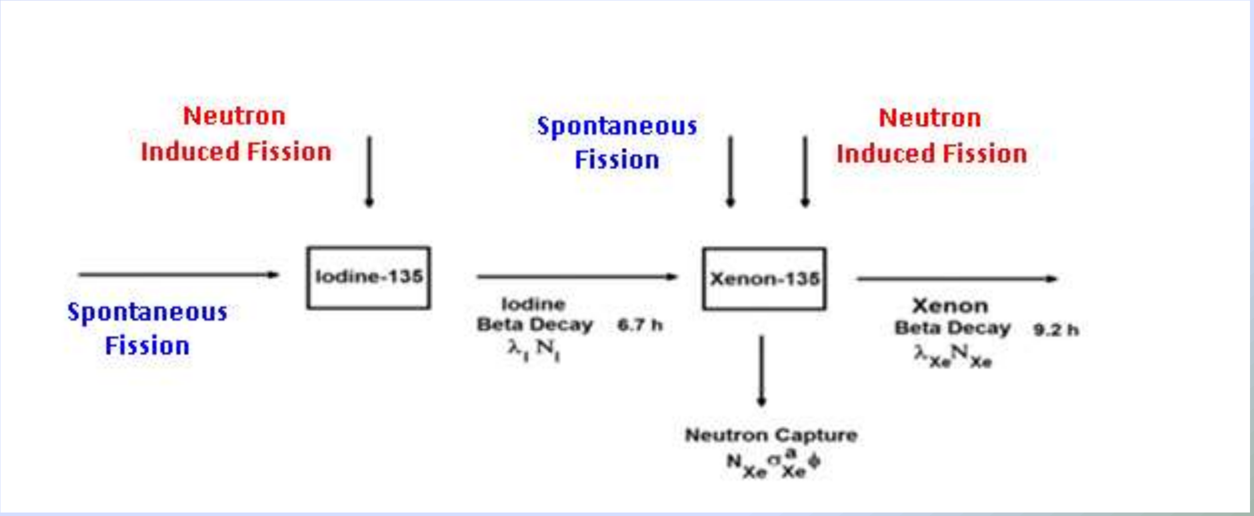
^{135}Xe PRODUCTION AND BURN-UP MODES

The ^{135}Xe production in nuclear reactors is due to the following modes:

- Spontaneous fission;
- Neutron induced fission;
- Decay of ^{135}I.

The ^{135}Xe burn-up occurs through the following modes:

- Decay, with a half-life of approximately 9.2 h;
- Transmutation in ^{136}Xe by neutron capture.



Xe^{135} - Production and burn-up modes.

DEPLETION EQUATIONS

Using these assumptions and considering neutron induced fissions and spontaneous fissions, the depletion equations of ^{135}I and ^{135}Xe to two energy groups are the following:

$$\frac{d}{dt}N_I(t) = \sum_i \gamma_I^i \sum_{g=1}^{2} \Sigma_{fg}^i \bar{\phi}_g + \sum_i \left(\frac{\tilde{\gamma}_I^i T_{SF}^i}{V} \right) - \lambda_I N_I(t)$$

$$\frac{d}{dt}N_{Xe}(t) = \sum_i \gamma_{Xe}^i \sum_{g=1}^{2} \Sigma_{fg}^i \bar{\phi}_g + \sum_i \left(\frac{\tilde{\gamma}_{Xe}^i T_{SF}^i}{V} \right) + \lambda_I N_I(t) - \left(\lambda_{Xe} + 10^{-24} \sum_{g=1}^{2} \sigma_{cg}^{Xe} \bar{\phi}_g \right) N_{Xe}(t)$$

^{135}Xe ACTIVITY CALCULATION

(Equilibrium Condition)

$$A_{Xe} = \frac{\sum_{i}(\gamma_I^i + \gamma_{Xe}^i)\sum_{g=1}^{2}\Sigma_{fg}^i\,\bar{\phi}_g + \sum_{i}\left(\frac{(\tilde{\gamma}_I^i + \tilde{\gamma}_{Xe}^i)T_{SF}^i}{V}\right)}{1+10^{-24}\sum_{g=1}^{2}\left(\frac{\sigma_{cg}^{Xe}\bar{\phi}_g}{\lambda_{Xe}}\right)}$$

COMPUTATIONAL TOOL

In order to obtain the radionuclide inventory in Unit 2, at the moment of the accident, it was used the ORIGEN code, from SCALE 6.0.

Input parameters utilized in ORIGEN.

Assembly Library	BWR GE 9X9-7
U-235 Initial Enrichment	3.8%
Power Rated Output	2381 GW
Specific Power	25.52 MW/T
Average Reactor Burn-up†	23.2 GWd/T
Water Density	0.7332 g/cm^3

† TEPCO estimative (TEPCO, 2011b).

Using the ORIGEN, it was obtained the isotopic composition of the nuclear fuel of Unit 2, at the moment of the accident.

UNCERTAINTY IN FUEL ISOTOPIC COMPOSITION

- The evaluation of the computational uncertainties in ORIGEN is an essential step to predict the isotopic concentration in the damaged fuel.

- To validate the code and data used in depletion approaches, experimental measurements are compared with ORIGEN results for relevant BWR spent fuel samples.

- The results of Hermann and DeHart (1988) for percentage difference between measured and computed nuclide composition for Cooper BWR samples (nuclear reactor similar to Fukushima Daiichi Unit 2, fuel assembly GE 7 X 7) was used in this work to obtain the evaluation of ORIGEN uncertainty for each nuclide.

FUKUSHIMA UNIT 2

FUEL ISOTOPIC COMPOSITION CALCULATED AT THE MOMENT OF THE ACCIDENT - ORIGEN RESULTS

Nuclide	Calculated Mass (g/ton U)	Uncertainty (g/ton U)
U-235	1.722×10^{4}	8.094×10^{2}
U-238	9.559×10^{5}	5.735×10^{3}
Np-237	2.177×10^{2}	2.329×10^{1}
Pu-238	4.395×10^{1}	1.538×10^{0}
Pu-239	3.811×10^{3}	2.667×10^{2}
Pu-240	1.301×10^{3}	5.983×10^{2}
Pu-241	6.256×10^{2}	4.442×10^{1}
Pu-242	1.358×10^{2}	1.059×10^{1}
Am-241	1.542×10^{1}	1.234×10^{0}
Am-243	1.543×10^{1}	-
Cm-242	2.791×10^{0}	4.885×10^{-1}
Cm-244	1.751×10^{0}	2.714×10^{-1}

Spontaneous Fission Rate

Nuclide	Branching Spontaneous Fission (%)	Spontaneous Fission Rate (s^{-1}) Nov./2011	Uncertainty of Spontaneous Fission Rate (s^{-1})
U-235	7.0×10^{-9}	9.052×10^{0}	4.254×10^{-1}
U-238	5.5×10^{-5}	6.093×10^{5}	3.656×10^{3}
Np-237	2.0×10^{-10}	1.073×10^{0}	1.148×10^{-1}
Pu-238	1.9×10^{-7}	4.957×10^{6}	1.735×10^{5}
Pu-239	3.1×10^{-10}	2.547×10^{3}	1.783×10^{2}
Pu-240	5.7×10^{-6}	5.858×10^{7}	2.695×10^{6}
Pu-241	2.4×10^{-14}	5.250×10^{1}	3.728×10^{0}
Pu-242	5.5×10^{-4}	1.021×10^{7}	7.964×10^{5}
Am-241	4.3×10^{-10}	7.899×10^{2}	6.635×10^{1}
Am-243	3.7×10^{-9}	3.961×10^{2}	-
Cm-242	6.2×10^{-6}	6.881×10^{8}	1.204×10^{8}
Cm-244	1.4×10^{-4}	6.556×10^{8}	1.016×10^{8}

It was considered that the fuel of the Unit 2 contains 94 tonnes of uranium.

NUCLEAR DATA

FISSION PRODUCTS YIELDS (%) AND CROSS SECTIONS

Nuclide	γ_I^i	$\tilde{\gamma}_I^i$	γ_{Xe}^i	$\tilde{\gamma}_I^i$	FAST (barn)	THERMAL (barn)
U-235	6.29	5.71	0.268	0.059	8.050	2.55×10^{2}
U-238	7.02	5.21	0.027	0.005	0.118	1.27×10^{-5}
Np-237	6.72	5.45	0.553	0.156	0.594	8.99×10^{-3}
Pu-238	5.74	5.16	0.994	0.661	1.730	6.10×10^{0}
Pu-239	6.54	5.23	1.080	0.297	8.880	7.28×10^{2}
Pu-240	6.85	5.17	0.531	0.129	0.684	3.82×10^{-2}
Pu-241	6.95	5.03	0.227	0.049	16.000	6.93×10^{2}
Pu-242	6.95	4.88	0.144	0.018	0.501	4.97×10^{-4}
Am-241	5.12	5.10	1.610	0.537	0.758	3.49×10^{0}
Am-243	6.04	4.97	0.727	0.101	0.501	4.67×10^{-2}
Cm-242	3.90	4.59	2.660	1.440	0.409	1.36×10^{0}
Cm-244	4.42	6.52	1.220	0.962	0.974	2.66×10^{-1}

CALCULATION OF K_{eff}

Neutrons emitted by spontaneous fissions can induce fissions (thermal and fast), in the nuclear fuel.

In this work it was also evaluated the effective neutron multiplication factor (K_{eff}), in function of neutron flux intensity, defined by:

$$K_{eff} = \frac{\sum_i \left(\sum_{g=1}^{2} \Sigma^i_{fg} \, \bar{\phi}_g \right)}{\sum_i \left(\sum_{g=1}^{2} \Sigma^i_{fg} \bar{\phi}_g \right) + \sum_i \left(\frac{T^i_{SF}}{V} \right)}$$

If and only if $K_{eff} < 1$
i. e., the system is subcritical

^{135}Xe ACTIVITY AND K_{eff}

(calculated in function of neutron flux intensity)

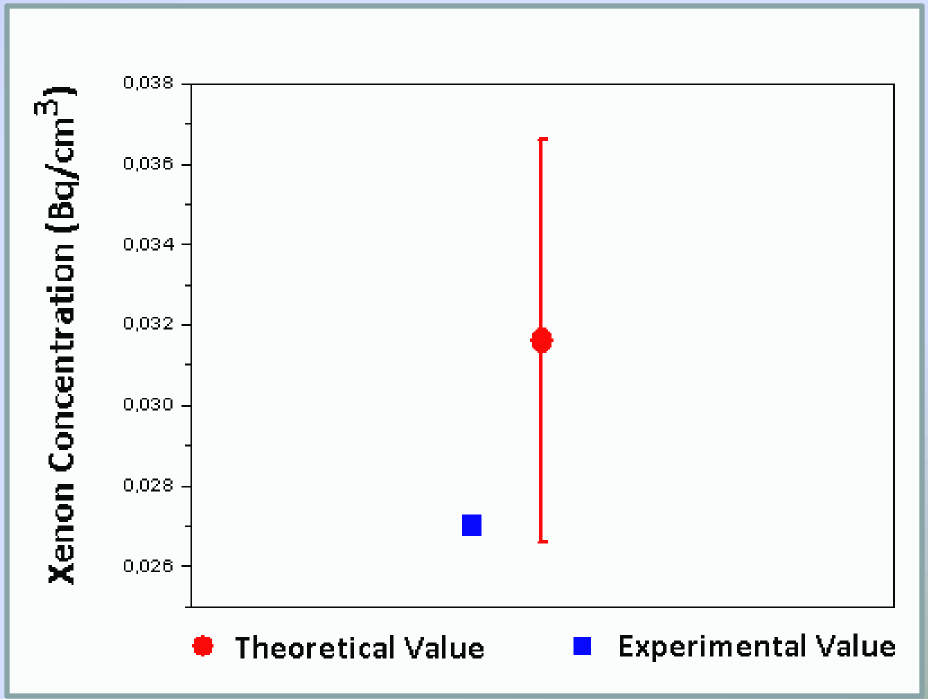
Neutron Flux ($cm^{-2}s^{-1}$)	^{135}Xe Activity (Bq/cm^3)	K_{eff}
0.00	$3.15 \times 10^{-2} \pm 5.0 \times 10^{-3}$	0.00
1.00×10^{1}	$3.19 \times 10^{-2} \pm 5.0 \times 10^{-3}$	1.07×10^{-2}
1.00×10^{2}	$3.56 \times 10^{-2} \pm 5.2 \times 10^{-3}$	9.79×10^{-2}
1.00×10^{3}	$7.30 \times 10^{-2} \pm 7.2 \times 10^{-3}$	5.20×10^{-1}
1.00×10^{4}	$4.47 \times 10^{-1} \pm 2.7 \times 10^{-2}$	9.16×10^{-1}
1.00×10^{5}	$4.19 \times 10^{0} \pm 2.3 \times 10^{-1}$	9.91×10^{-1}
1.00×10^{6}	$4.16 \times 10^{1} \pm 2.2 \times 10^{0}$	9.99×10^{-1}
1.00×10^{7}	$4.16 \times 10^{2} \pm 2.2 \times 10^{1}$	9.999×10^{-1}
1.00×10^{8}	$4.16 \times 10^{3} \pm 2.2 \times 10^{2}$	9.9999×10^{-1}
1.00×10^{9}	$4.16 \times 10^{4} \pm 2.2 \times 10^{3}$	1.00
1.00×10^{10}	$4.15 \times 10^{5} \pm 2.2 \times 10^{4}$	1.00
1.00×10^{11}	$4.14 \times 10^{6} \pm 2.2 \times 10^{5}$	1.00
1.00×10^{12}	$3.95 \times 10^{7} \pm 2.1 \times 10^{6}$	1.00
1.00×10^{13}	$2.71 \times 10^{8} \pm 1.5 \times 10^{7}$	1.00

FINAL COMMENTS

- The calculated value of xenon activity can be used to infer the sensitivity of this variable concerning the neutron multiplication factor. The magnitude of K_{eff} serves to point out how much the core composition is away from the criticality condition.

- In the case of zero neutron flux, the ^{135}Xe activity represents, uniquely, the contribution due to spontaneous fission.

- The value of ^{135}Xe activity measured is within the range of uncertainties for the maximum expected activity in condition of zero neutron flux. So, only spontaneous fissions of actinides have contributed for the xenon detected in Unit 2.

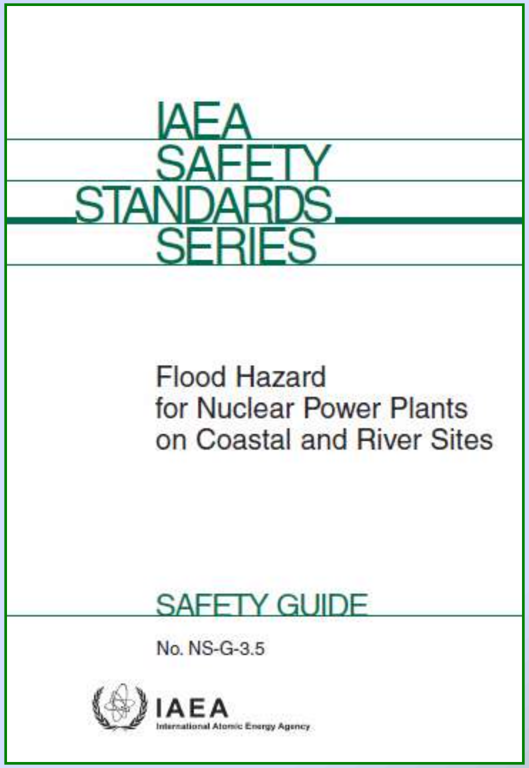


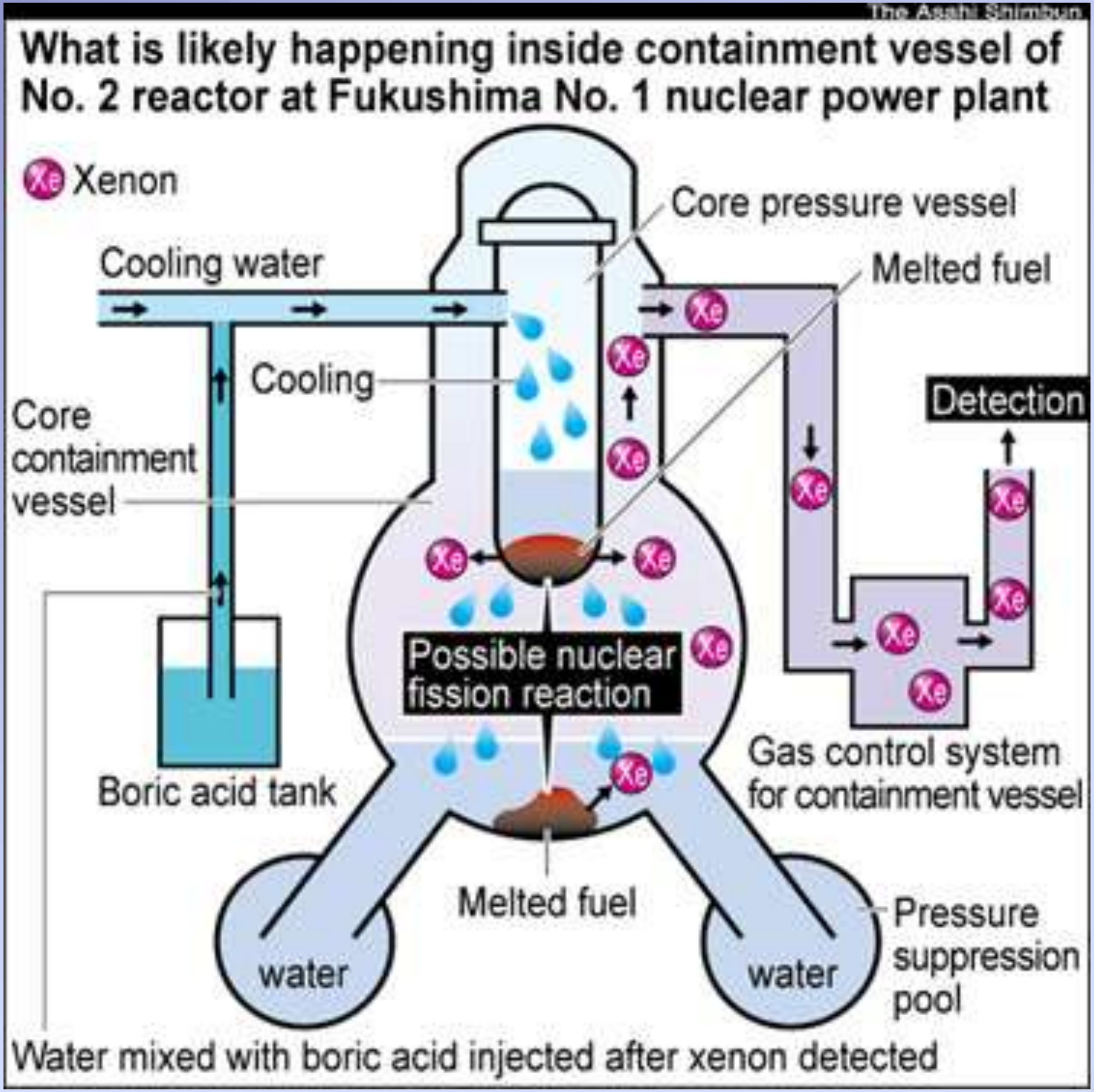
ADDITIONAL INFORMATIONS

IAEA – SAFETY GUIDE

The International Atomic Energy Agency published a Safety Guide (IAEA, 2004) with recommendations about the evaluation of the flood hazard for a nuclear power plant (on a coastal or river site), presenting guidelines for the analysis and quantification of flood effects in all the phases of the project.

The main effects of flooding on a nuclear power plant site may be a common cause of failure for safety related systems, such as the emergency power supply systems.





Extracted from The Asahi Shimbun

Glossary

$\bar{\phi}_g$	the average neutron flux for energy group *g*
V	the volume of damaged fuel
N_I	the concentrations (nucleus per cm^3) of ^{135}
N_{xe}	the concentrations (nucleus per cm^3) of ^{135}Xe,
$\tilde{\gamma}_I^i$	the spontaneous fission product yield for ^{135}I
γ_I^i	the neutron induced fission product yield for ^{135}I
$\tilde{\gamma}_{Xe}^i$	the spontaneous fission product yield for ^{135}Xe
γ_{Xe}^i	the neutron induced fission product yield for ^{135}Xe
T_{SF}^i	the spontaneous fission rate
Σ_{fg}^i	the macroscopic fission cross section for each neutron energy group, for each actinide i
λ_I	the ^{135}I decay constant
λ_{Xe}	the ^{135}Xe decay constant
σ_{cg}^{Xe}	the microscopic ^{135}Xe neutron capture cross section for each energy group g

CONCLUSIONS

- In this work was calculated a ^{135}Xe maximum activity of 0.0315 ± 0.0050 Bq/cm^3, in condition of zero neutron flux, in the Fukushima Daiichi - Unit 2.

- The comparison between the value of ^{135}Xe activity obtained experimentally by TEPCO (0.027 Bq/cm^3) and the value here calculated (condition of zero neutron flux), shows that the measured xenon activity which is inside the range of the uncertainties calculated .

- From this comparison, one can conclude that there is no evidence of occurrence of neutron induced fission in the damaged reactor (Unit 2), and therefore, there is no evidence of recriticality, as well.