Correlation between neutrons detected outside the reactor building and fuel melting

1. Introduction

The Fukushima Daiichi Nuclear Power Station (hereinafter referred to as “Fukushima Daiichi NPS”) Unit-1 to Unit-4 experienced a station blackout (SBO) due to the loss of all power sources: loss of external sources damaged in the wake of the Tohoku–Chihou-Taiheiyou-Oki (Great East Japan) Earthquake that had struck at 14:46 on March 11th, 2011; and loss of emergency diesel generators because of the ensuing tsunami. All radiation monitoring posts lost their ability to function due to the power source losses. Environmental radiation monitoring on the site depended solely on mobile radiation monitors mounted on monitoring cars.

These mobile radiation monitors detected neutrons twice, in the early morning of March 13th, and during the night of March 14th to the dawn of March 15th. The neutron dose rates measured were minimal, 0.01μSv/h, which is the detectable limit of the neutron detectors, and 0.02μSv/h, which is twice the detection limit. These neutrons are not considered to have come directly from the reactors, because they were detected near the station main gate, far from the reactor buildings. The origin of these neutrons has been considered as unknown, since their detection was not consistent with the timing of increased gamma radiation doses on the site due to release of radioactive materials. Furthermore, the leak timing and path of uranium, plutonium, etc. detected from the soil of the site were also considered unknown.

This document attempts to account for the neutrons detected, based on the findings of the accident progression in Unit-1 to Unit-3 as obtained so far.

2. Progression of the accident events in Unit-2 and Unit-3

2.1 Estimations of the time when fuel melting occurred in Unit-2

Figure 1 shows the correlation between the estimated processes of water injected and steam generation, as related to the progression of fuel melting in Unit-2.

- Reactor depressurization started at 18:02 on March 14th by opening the main steam safety relief valve (SRV). The reactor pressure was lowered to about 1 MPa by around 18:30.
- Accordingly, the reactor water level decreased from just below the top of active fuel (TAF) at TAF-1.1m (designated as stage 0 in Figure 1) before depressurization at 18:00 to below the bottom of active fuel (BAF) at TAF-3.9m (stage 1) at 18:30.
- After fire engines resumed water injection at 19:54, the reactor pressure started to increase at around 20:15.
- Later the reactor pressure decreased, while the PCV pressure increased from a stable level, over the time period including 21:20, when the SRV opening operation was recorded.
- Also between 22:40 and 23:25 on March 14th, and between 00:06 and 01:02 on March 15th, the reactor pressure increase and decrease were observed to have started as shown

in Figure 2.

These three occasions of reactor pressure transition can be interpreted as follows (with reference to Figure 1).

- Water injection starts at Stage 1 by fire engines, the water injected around the outside of the shroud reaches the core and steam starts to be generated (Stage 2).
- The water level increases due to continued water injection and steam increases accordingly (Stage 3).
- Steam generated contributes to the cooling of fuel, but the overheated fuel cladding leads to water-zirconium reactions, producing large amounts of hydrogen gas and energy.
- This increases the reactor pressure, exceeding the fire engine pump discharge pressure of about 1MPa, and thus the injected water fails to reach the reactor.
- As a result, the reactor water level decreases, and the steam generation is reduced (Stage 4).
- When the reactor water level drops below BAF, steam generation almost completely stops and the reactor pressure starts to decrease (Stage 5).

When the reactor pressure decreases to a level low enough to allow water injection by the fire engines, the reactor status returns to Stage 1 and the same sequence is repeated. This is considered to be the mechanism of the repeated pressure increase and decrease during the night of March 14th to the dawn of March 15th. During the transition from Stage 3 to Stage 4, water-zirconium reactions release large amounts of energy in a limited time, and fuel temperatures are considered to rise sharply. This is probably when fuel melting occurred.

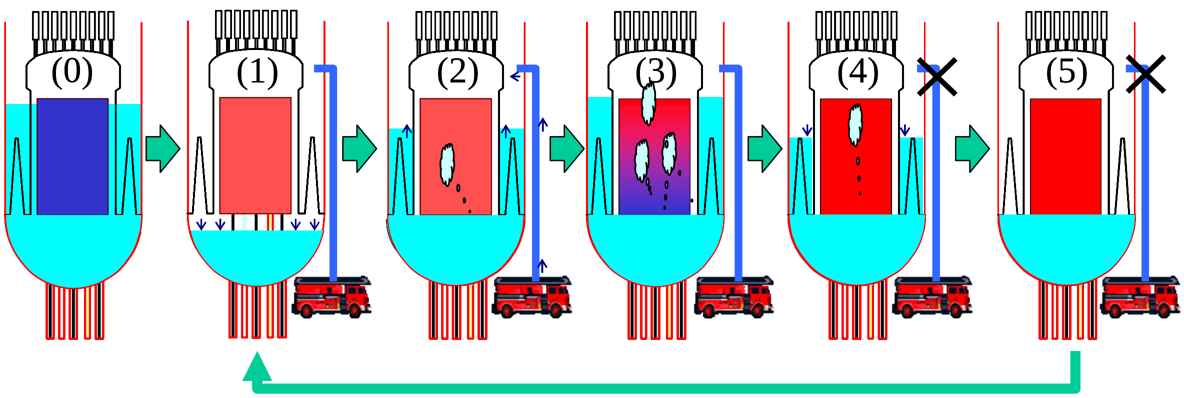

Figure 1 Steam generation and pressure increase after the water injection

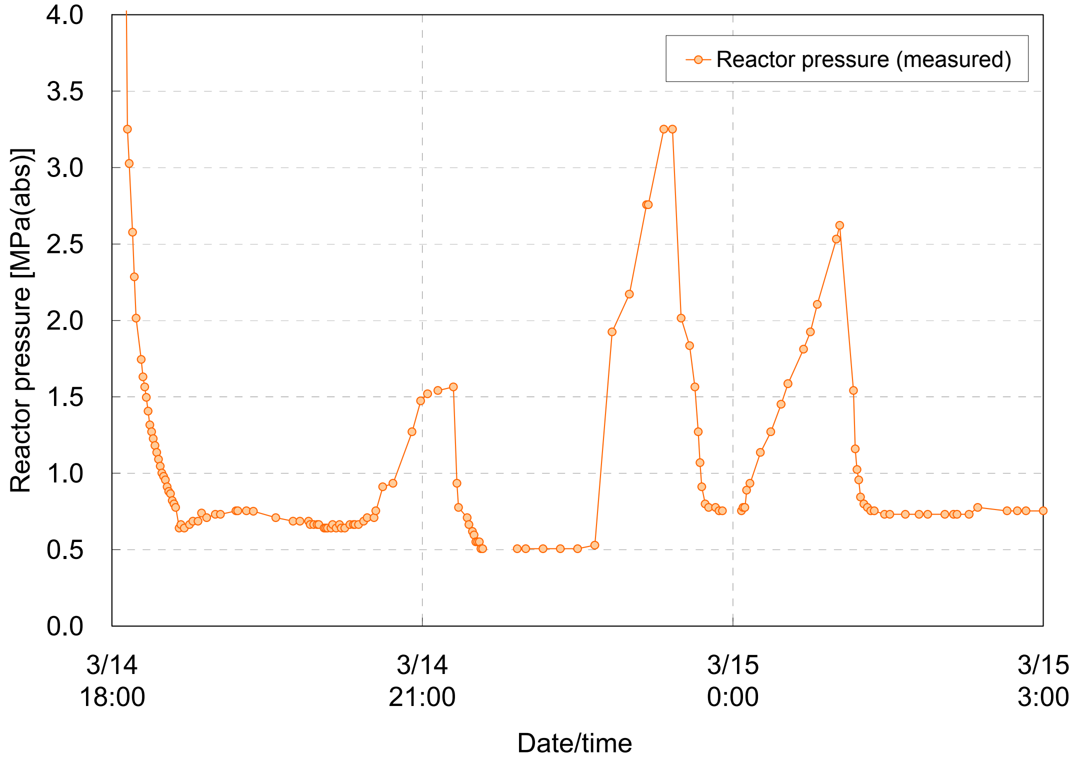

Figure 2 Observed intermittent increase in reactor pressure in Unit-2.

2.2 Estimations of the time when fuel melting occurred at Unit-3

Figure 3 shows the correlation between the measured reactor water level in Unit-3 and that obtained by the MAAP analysis which TEPCO published in March 2012. As already reported in "Unit-3/Issue-3," the estimated water injection by HPCI could not have provided sufficient water into the reactor before the operator manually shut down the HPCI. As a consequence, the reactor water level might have been at or near the TAF level at the time when the HPCI was manually shutdown. The reactor water level was almost constant at BAF+0.7m from 07:35 on March 13th to the time of reactor depressurization, as measured by the fuel range water level indicator (Figure 4). It is unlikely, though, that the reactor water level would remain constant when decay heat was continually releasing energy. The actual water level can be estimated as below BAF, since the apparently constant water level is likely to have overestimated the actual value due to the evaporated water head in the reference condensing water chamber side piping.

As seen above for Unit-3, in the accident progression the reactor water level can be estimated to have gradually decreased due to boiling, while its pressure remained high, down to the BAF level and then the fuel was completely uncovered, as had occurred in Unit-1. While the water level is gradually decreasing, the fuel cladding temperature rises remain more or less limited by the steam flow from below (steam cooling). But as the water level decreases, the area of the exposed fuel surface to be cooled increases and the amount of steam for cooling decreases. Thus the fuel cladding temperatures are considered to increase gradually. The cladding material Zr reacts with steam and forms oxides. The oxidation progresses rapidly due to positive feedback once the cladding temperature exceeds 1200 deg C. Therefore, if this rapid oxidation had progressed during the reactor water level decrease, fuel might have started melting, due to the large amounts of energy released during the time period of 05:00 to 07:00 on March 13th, depending on the oxidation history before then and the decay heat.

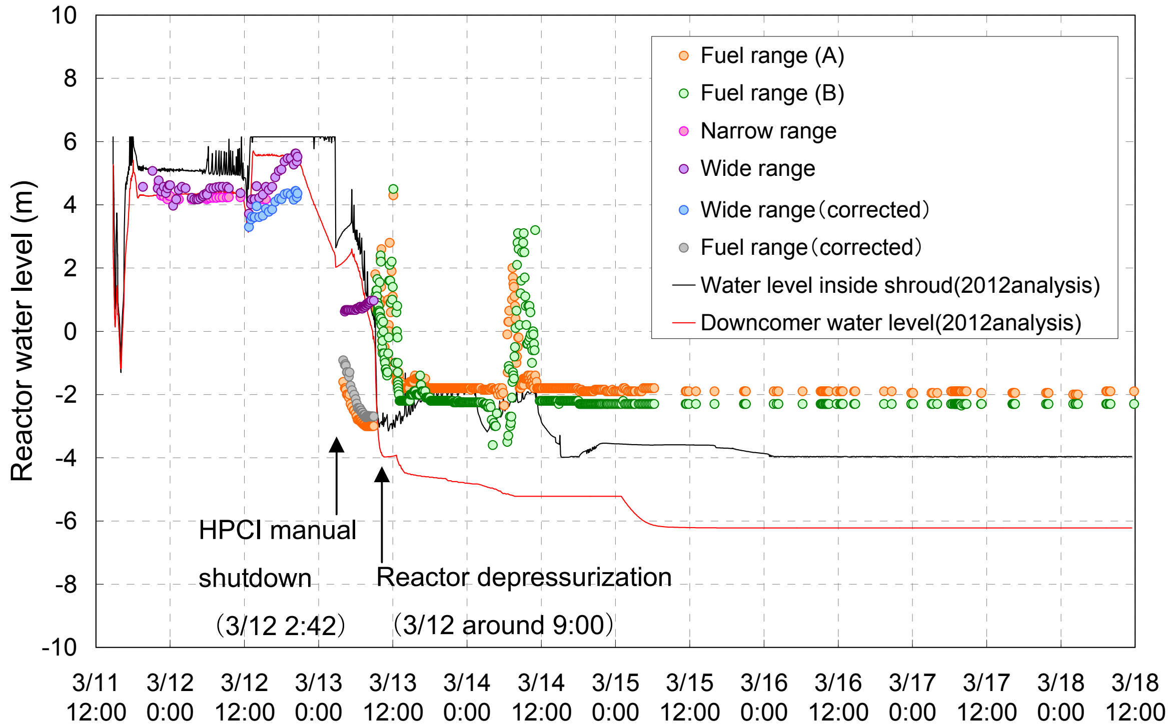

Figure 3　Reactor water level changes in Unit 3

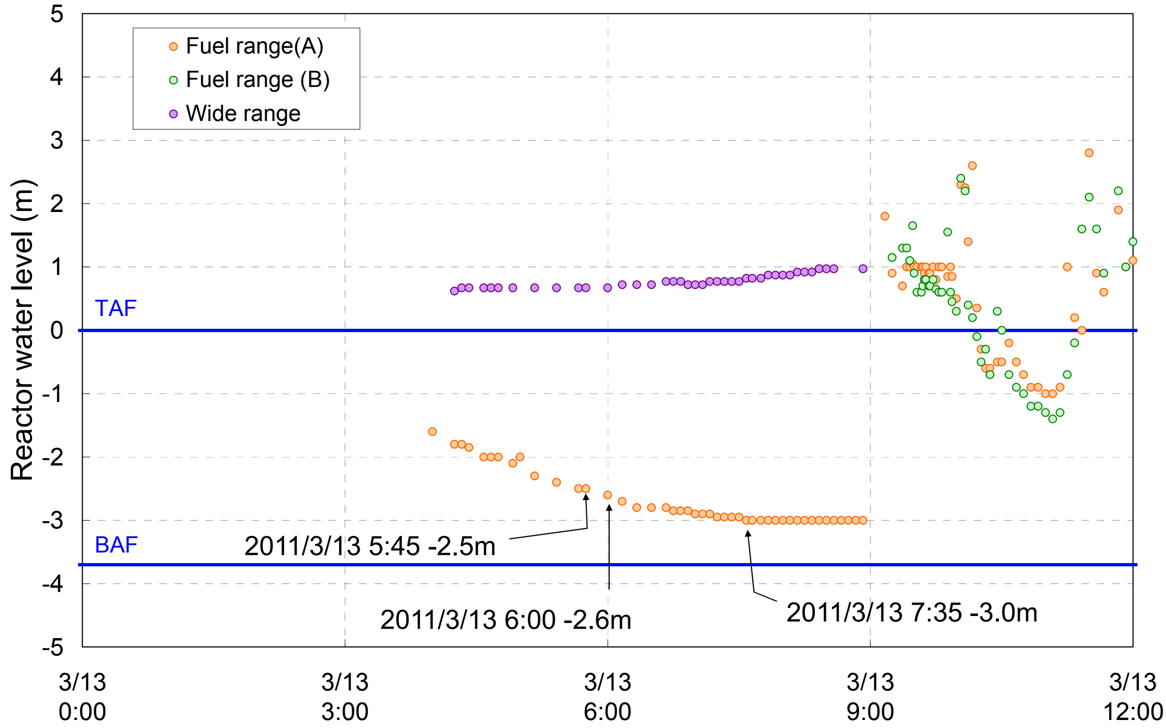

Figure 4 Reactor water changes in detail in Unit-3

3. Timing of neutron detections

3.1 Neutron detections on March 13th, 2011

Table 1 gives the times when neutrons were detected on March 13th. Figure 5 shows the measured gamma dose rates near the main gate, in which blue points are the times when the neutron dose rate of 0.01μSv/h was observed, while red points show the times of 0.02μSv/h. In the morning of March 13th, gamma dose rates increased due to reactor depressurization at around 09:00 and venting from S/C accordingly, but no correlations can be observed between neutron detections and gamma dose rate changes. This indicates the origin of neutrons is not related to the FP release which caused the gamma dose rate increase.

In the meantime, there is a hint of some connection between the neutron detections and fuel melting when the time period of possible fuel melting estimated from the reactor water level changes is considered, although they do not coincide exactly. That is, some of the actinide elements such as uranium, plutonium, etc. released into the gaseous phase by fuel melting may have leaked out of the reactor building through a path, different from that of the FPs which caused gamma dose rate increases, and the neutrons emitted by spontaneous fissions of plutonium, curium, etc. may have been detected. Plutonium isotopes, which seem to have clearly originated from the accident of the Fukushima Daiichi NPS, have been detected in soil sampled on the site, although the amounts are on the same level as that of plutonium isotopes accumulated in the past, originating from the fallout of nuclear bomb tests.

The main gate where neutrons were detected is, as shown in Figure 6, about 1km from the reactor buildings. Therefore, it is unlikely that neutrons from the reactors were directly detected. Furthermore, BWRs are designed to maintain their chain reactions by neutrons moderated by water. Therefore, it is considered that the neutrons detected are not the neutrons, which originated from those neutrons generated in the chain reactions for criticality in the reactor, in view of the following conditions.

- The reactor would be unlikely to go critical, because almost no water was present in the core and neutrons were not moderated.
- The reactor would be unlikely to become critical due to negative reactivity feedback by the Doppler effect, because the core was at elevated temperatures.
- Control rods would be likely to melt first and lose their function to absorb neutrons in the core region. However, control rods are designed to absorb moderated neutrons and have little effect for absorbing hard neutrons in waterless conditions (neutrons with no moderation).

Table 1 Times when neutrons were detected on March 13th, 2011

Times when neutrons were detected
2011/3/13 5:30
2011/3/13 5:40
2011/3/13 5:50
2011/3/13 6:30
2011/3/13 6:40
2011/3/13 7:10
2011/3/13 7:40
2011/3/13 8:00
2011/3/13 8:40
2011/3/13 9:30
2011/3/13 10:50

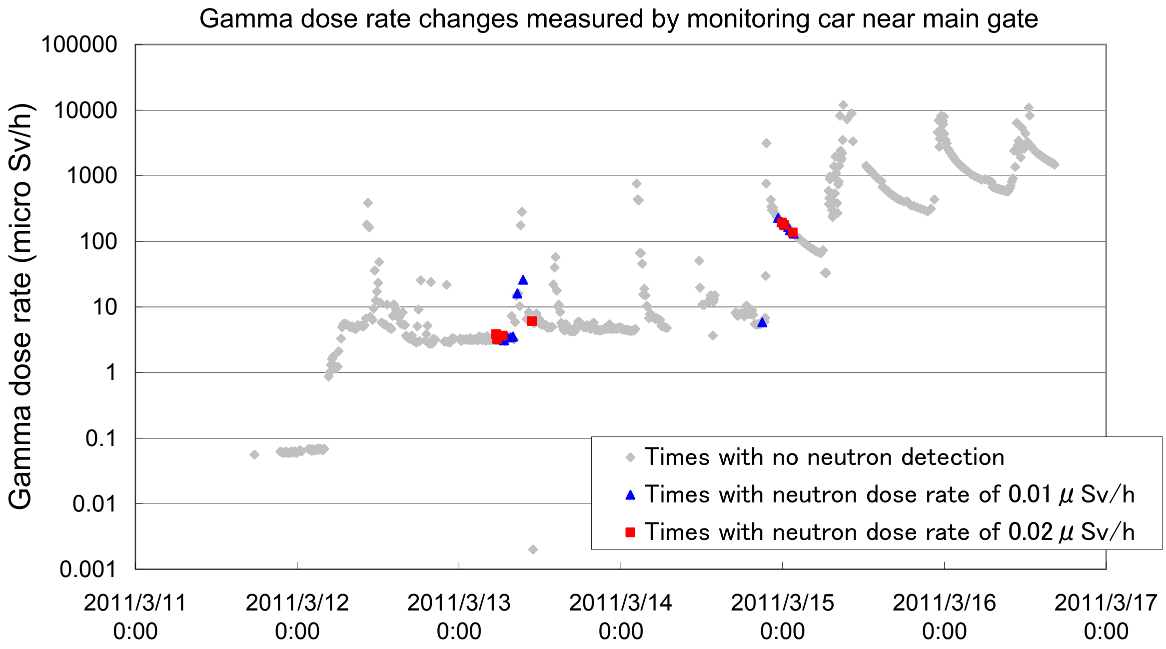

Figure 5　Gamma dose rates near the main gate

(Colored symbols show the time when neutrons were detected)

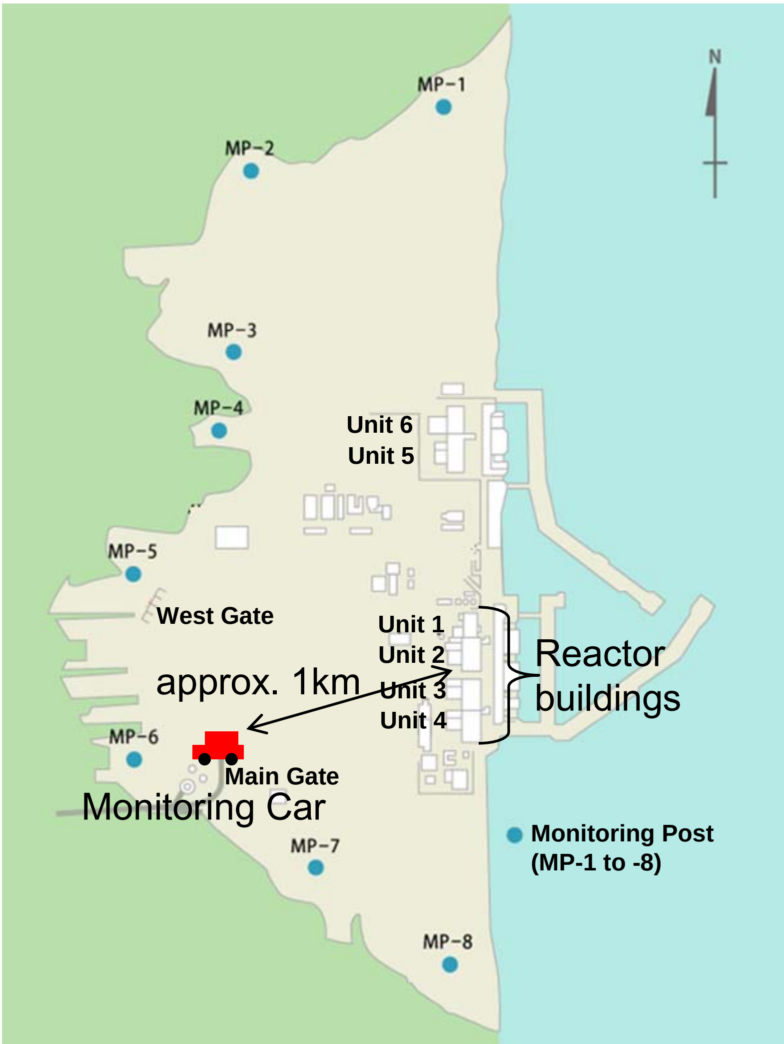

Figure 6　Locations of deployed monitoring car and reactor buildings of Unit-1 to Unit-3

3.2 Neutron detections on March 14th and 15th, 2011

Table 2 gives the times when neutrons were detected on March 14th and 15th. When compared with the gamma dose rate changes in Figure 5, it can be seen that the first detection of neutrons was at 21:00 on March 14th and before 21:30 when the gamma dose rate sharply increased and that the next detection was after 23:30. Again, no correlations are noticed between neutron detections and gamma dose rate changes.

Figure 7 shows the correlation between the times when neutrons were detected and the reactor/PCV pressures. Neutrons were detected after the reactor pressure started to increase. As discussed earlier, this reactor pressure increase is considered to be due to steam generated by the water injected by fire engines arriving at the core region. At the same time, large amounts of energy are considered to have been released due to zirconium-water reactions, leading to fuel melting. In other words, some of the actinide elements such as uranium and plutonium released into the gaseous phase by fuel melting during this time period may have leaked out of the reactor building through a path, different from that of the FPs which caused gamma dose rate increases, and the neutrons emitted by spontaneous fissions of plutonium, curium, etc. may have been detected.

Table 2 Times when neutrons were detected on March 14th and 15th, 2011

Times when neutrons were detected
2011/3/14 21:00
2011/3/14 23:20
2011/3/14 23:50
2011/3/14 23:55
2011/3/15 0:10
2011/3/15 0:15
2011/3/15 0:20
2011/3/15 0:50
2011/3/15 1:05
2011/3/15 1:30
2011/3/15 1:40

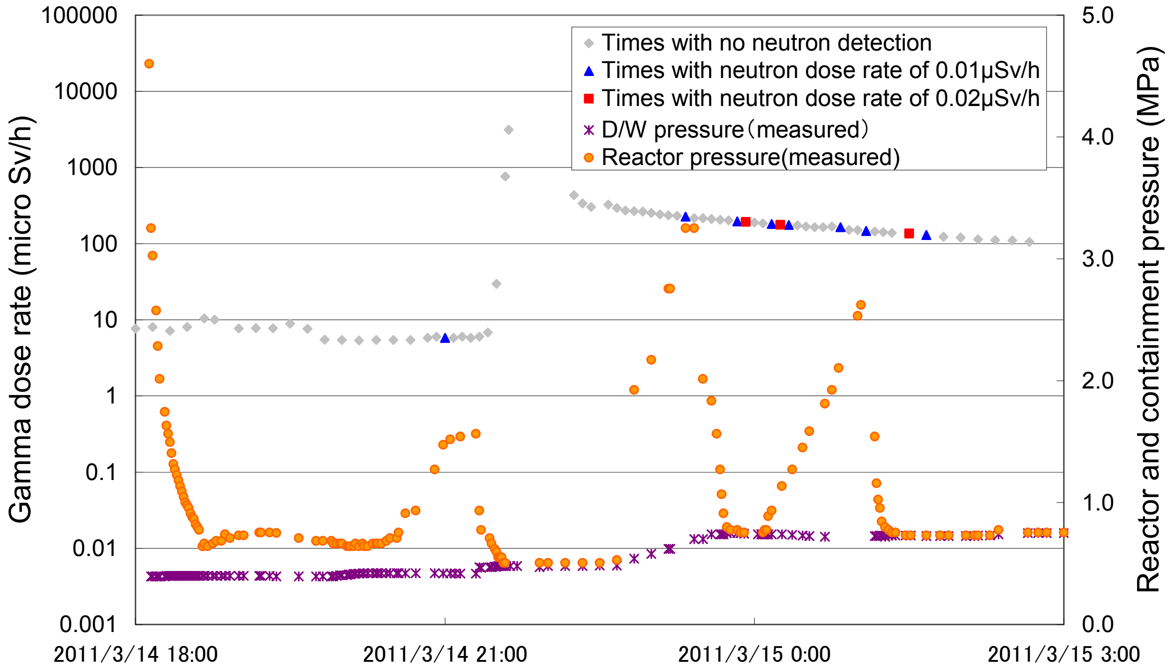

Figure 7　Correlations between times when neutrons were detected and reactor/PCV pressures

4. Summary

Examinations were conducted into the sources of neutrons detected in the early morning of March 13th, and during the night of March 14th to the dawn of March 15th. Up until now, no clear explanations had been given, since no correlations were observed between neutron detections and gamma dose rate changes. However, as the accident progression behavior could now be interpreted based on calculations, and the time period of fuel melting could be approximately identified, the possibility of a close correlation has been shown between neutron detections and fuel melting.

Concerning the release path of actinide elements, which might have led to the neutron detections, continued examination is needed, because this information is also important for ensuring safety of site personnel.

(End)