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Introduction to Criticality Accident Evaluation

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1. Introduction

For the use of nuclear energy, uranium must be processed to make a form to fit the purpose, such as a pellet. In nuclear fuel fabrication process, uranium is dissolved with nitric acid to make homogeneous solution. Uranium nitrate solution is full of water, which is the best moderator for neutron. That is a well-known reason of the criticality accident of nuclear fuel.

So far 22 criticality accidents have been reported (McLaughlin et al., 2000) including JCO accident in Japan. All cases took place with fuel solution or slurry except one case of metal fuel. It was not clear that what was actually happening during those criticality accidents except the JCO accident, for which a fission power profile was reproduced from a gamma-ray monitoring data (Tonoike et al., 2003). It is not easy to understand the phenomenon of criticality accident in detail, because it is a mixture of reactor physics, thermal dynamics and fluid dynamics.
In a criticality accident, the dose of the employee or public is the most important information. It is estimated from the amount of fission products produced during the criticality accident. The amount of fission products is proportional to the total number of fission, which is used to estimate the scale of the criticality accident as well.

Many kinetic methods have been developed for the estimation of the total number fission (Mather et al., 1984; Basoglu et al., 1998; Pain et al., 2001; Nakajima et al., 2002a, 2002b; Mitake et al., 2003). Some of them shows good agreement to experimental results (Miyoshi et al., 2009), however, Such methods requires rather high cost for the calculation and a lot of calculations are needed to find the response of the result to the variables such as temperature, input reactivity, etc.

Four simplified methods have been proposed to calculate the total fission number for a criticality accident (Tuck, 1974; Olsen et al., 1974; Barbry et al., 1987; Nomura & Okuno, 1995). Some are empirical equations and some are theoretical. Those simplified methods are easy to use, low cost and quick calculation, however, are known to overestimate the number of fission too much (Nakajima, 2003). Such overestimation could be reduced if we would focus to the detail power profile during the criticality accident.

The aim of the chapter is to introduce a concept of new method developed to evaluate the number of fission in a criticality accident, which is expected to give reasonable value, not

too much overestimated, i.e. the estimated value is in the almost same order as the actual value.

The 1st section introduces the phenomena of the criticality accident with uranyl nitrate solution based on the TRACY experiment, which has been conducted by Japan Atomic Energy Agency (Nakajima et al., 2002c, 2002d, 2002e). The power profile is divided into three parts, the 1st peak, monotonically decreasing and plateau, for them the dominant mechanism and its time scale are different to each other. In the 2nd section, the condition characterizing a criticality accident is considered, such as temperature, reactivity temperature coefficient, water, cooling, etc. In the 3rd section, a new simplified method to evaluate the total fission number is described. The estimation is done for part-by-part by using equations differently introduced based on one-point kinetics equation or thermal equation (Yamane et al., 2007, 2008, 2009). In the 4th section, the new developing method is applied to some case to see its applicability.

2. Characteristics of criticality accident

In this section, the character of criticality accidents is explained. The most description is based on the data from TRACY experiments. At the first, the image of phenomenon, what happening, and its underlying physics are briefly introduced. Then, the conditions characterizing criticality accidents are described.

2.1 Phenomenon

A criticality accident occurs if enough amount of nuclear solution fuel such as uranyl nitrate solution is pumped into a tank with a shape not designed to avoid criticality. In most cases, very high energy caused by the fission of uranium is released in a moment, which is called ”the first peak” of power profile. At the same time, the temperature of uranium and surrounding water is increased due to the released fission energy and the system becomes subcritical. After the first peak or at the same time, radiolytic gas void mainly due to the fast moving of fission product in the water is created and grows up. In any case, the system is approaching critical again, but if the system were disconnected thermally from surrounding materials, it would keep being subcritical. This phenomenon is a typical example of the system of uranyl nitrate aqueous solution. For the case of the largest total fission so far, criticality terminated after a large amount of water had been vaporized out. For the system of dilute plutonium solution, powder or metal, the phenomena may be different to each other.

2.2 Physics

Nuclear side of the phenomenon noted above is described by transport equation of neutron, which consists of neutron flux and its probability of reaction with nuclides. It can be applicable to any complicated condition but solving the equation for a complicated geometry, for example, requires a lot of computation power. Some assumptions, however, can reduce the complexity of neutron transport equation to make one-point kinetics equation (1). It has a simple form and even has the general solution for the simplest condition. It is enough for our purpose to understand the underlying physics of criticality accident.

$$\frac{dn}{dt} = \frac{\rho - \beta_{eff}}{\ell} n + \sum_{i=1}^{6} \lambda_i C_i$$
$$\frac{dC_i}{dt} = \frac{\beta_i}{\ell} n - \lambda_i C_i \tag{1}$$

where power, n, is used instead of the number of neutron, because they are proportional to each other and the equations are homogeneous. In the equation (1), $\rho = (k-1)/k$ is reactivity and k, neutron multiplication factor. $k=1$ means the number of neutron doesn't change in course of time. $k>1$ means its increase and $k<1$, decrease. They correspond to $\rho = 0$, $\rho > 0$ and $\rho < 0$ each other. When a system is critical, its $\rho = 0$ and $\rho < 0$ stands for subcritical. Neutron multiplication factor, k, is a property of material and it is defined for infinite geometry. An actual system has a finite geometry and there must be some leak of neutrons from the system depending its shape. For such finite system, effective neutron multiplication factor, k_{eff}, is used instead of k. C_i is the density of i-th delayed neutron precursor, which is a source of neutrons being released with a time constant λ_i, a decay constant of i-th delayed neutron precursor. The order of λ_i is in the range of about 0.17s and 55s.

The ratio of the number of delayed neutrons to the total neutrons is denoted as b_{eff}, where suffix *eff* means that the leak of precursors and delayed neutrons from the fuel solution is considered. The system is delayed critical if $0 < \rho < b_{eff}$, the system needs a help of delayed neutron to keep fission chain reaction and the fission power grows linearly. If $\rho > b_{eff}$, the system is prompt critical; fission chain reaction continues only with prompt neutrons and the power increases exponentially. ρ divided by b_{eff} is used for convenience and comes with unit "$"; i.e. more than 1$ excess reactivity corresponds to prompt critical. ℓ is prompt neutron life time. The time scale of neutronics is very small because ℓ is small. For example, ℓ is about 5×10^{-5}s for TRACY.

During a criticality accident, most of released fission energy is consumed to increase the temperature of uranium and other materials. Some equations such as thermal conduction equation, heat diffusion equation or Fourier's law can describe the distribution of thermal energy. The temperature of uranium is important because it has reactivity effect well-known as "Doppler effect," which can change the system's total reactivity, mainly decreases it. The thermal expansion of the solution has the same effect as well.

After a fission, high energy fission products run into water, excite or break water molecules and the overlapping of those exciting atoms and ions make voids in the fuel solution. Such radiolytic gas void usually has a negative reactivity effect; a lot of void can make the system subcritical. Boiling void has the same reactivity effect. The motion of void is a matter of fluid dynamics and can be described as multi phase flow. The knowledge of the elementary step of creating void is not enough, however, some models are used tentatively (Nakajima et al., 2002).

2.3 Power

For criticality accident, in other words, transient criticality, power profile has its unique pattern depending on the condition of the system. For an example, a typical power profile is explained based on TRACY experiment.

Figure 1 shows a power profile data obtained from a transient criticality experiment with 1.5$ excess reactivity inserted by pumping fuel solution into the TRACY core tank. At the first, the power increases exponentially and suddenly decreases to make a peak in the power profile. Then, the power decreases monotonically. Finally it recovers and keeps some value as plateau shown in Fig.1.

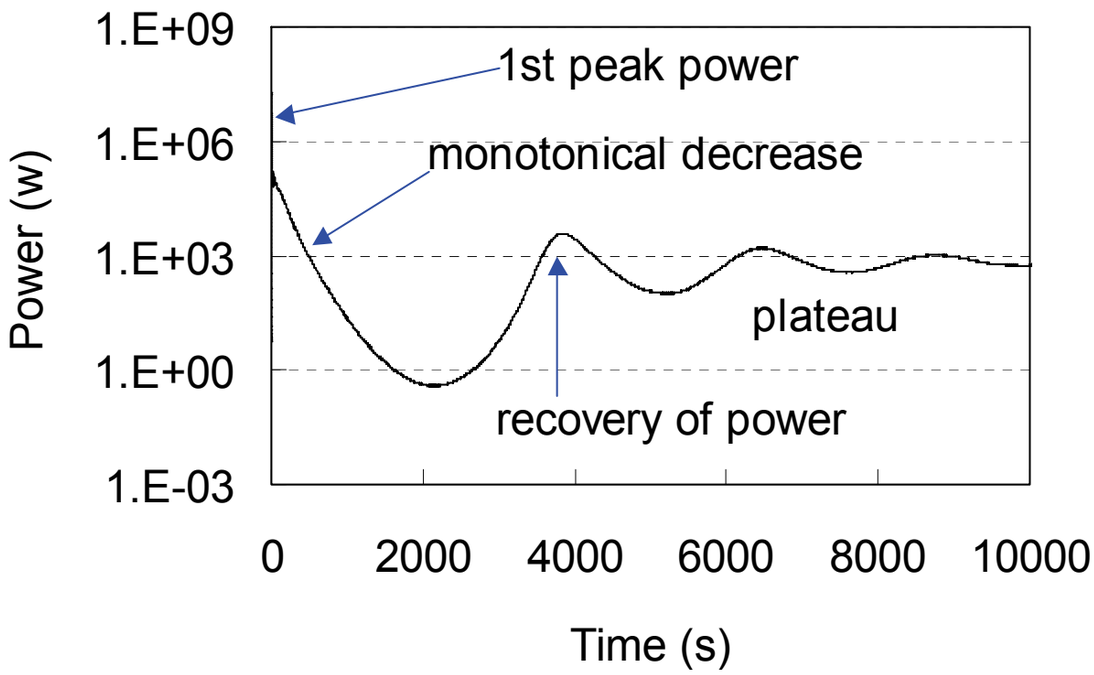

Fig. 1. Example of power profile observed by using TRACY for the case of 1.5$ inserted by feed of nuclear fuel solution.

Around the first peak for the case more than 1$ excess reactivity inserted instantaneously, the power profile is described as follows;

$$\frac{dn}{dt} = \frac{\rho - \beta_{eff}}{\ell} n \tag{2}$$

Because the width of the 1st peak is less than 0.1s, which is less than the smallest l_i, the change in C can be ignored.

Monotonic decrease of the power is driven by the decay of neutron precursors and described as follows;

$$n = n_{pe} \exp(-\lambda_1 t) \tag{3}$$

where λ_1 is the smallest decay constant.

The power at plateau corresponds to the cooling rate of the fuel solution. For TRACY, during several hours from the first, the thermal conduction from the fuel solution to the stainless steel tank and its support plays a dominant role.

It should be mentioned that another pattern is observed for another condition such as initially boiling, dilute plutonium solution, etc.

2.4 Energy-fission

Released fission energy is consumed to increase the temperature of uranium and other nuclides except some loss. one fission gives rise to about 200MeV energy and 10^{18} fission is almost equal to 32MJ. The number of fission at the largest criticality accident so far is $4x10^{19}$. That is, however, a rare case. For the second largest and other cases, the number of fission is $3x10^{18}$ or less.

The number of fission is used to evaluate the public dose around a nuclear facility. For example, for the design of Rokkasyo fuel reprocessing facility, 10^{19} fission is used for DBE, Design Basis Event, which is a postulated event to evaluate the safety of the design of the facility, 10^{20} for SEA, Siting Evaluation Accident, which is an postulated accident to evaluate the safety of the public around the facility (Working Group on Nuclear Criticality Safety Data, 1999).

2.5 Temperature

The temperature of nuclear fuel solution increases when a criticality accident occurs. Figure 2 shows a typical example of the temperature profile measured at TRACY. The difference value from the initial temperature is determined the released fission energy and the specific

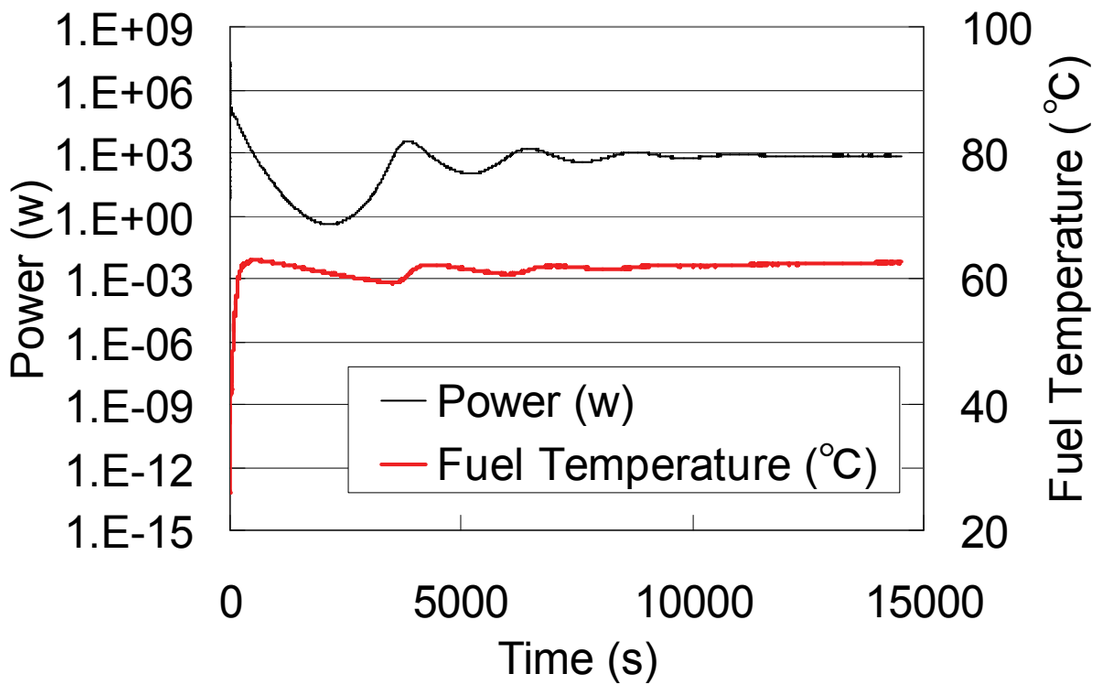

Fig. 2. Example of fuel temperature profile with power profile for the case of 1.5$ inserted by feed of nuclear solution fuel.

heat of the fuel solution and it gives rise to reactivity feedback. The ratio of the feedback reactivity to the temperature difference is called "reactivity temperature coefficient." More precisely, feedback reactivity is a function of temperature difference and described as follows;

$$\rho = \sum_i \alpha_i \Delta T^i \tag{4}$$

where α_i denotes i-th order reactivity temperature coefficient. For uranyl nitrate solution, the temperature feedback reactivity has negative value and is a non-linear function of temperature difference usually.

2.6 Pressure

In the fuel solution, rapid increase of temperature gives rise to rapid expansion of the solution. That is observed as pressure. Some pressure can be observed and measured at pulse withdrawal mode experiment of TRACY, for that the neutron absorber rod in the centre of the core tank is instantaneously withdrawn. Pressure can observed clearly at slow transient experiment as well.

3. Simplified evaluation of fission yield

In this section, a new simplified evaluation method for the number of fission released at a criticality accident is proposed, which is consist of three parts.

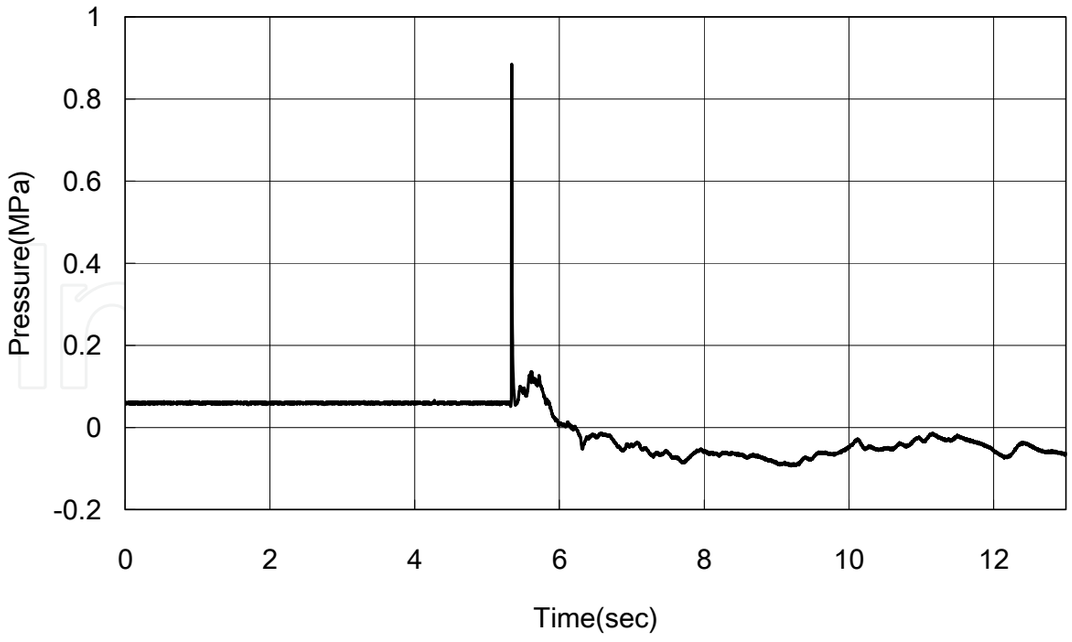

Fig. 3. Example of pressure profile measured at TRACY experiment for the case of 2.9$ inserted instantaneously by pulsed withdrawal of the transient rod (Nakajima et al., 2002a).

3.1 Around first peak power

If all of the excess reactivity is given to the system in pseudo-steady manner, a power peak like the one shown in section 2.3 cannot be observed. In most actual cases, the excess reactivity was inserted in a rate and that caused an exponential growth of power. It is known that if the excess reactivity is given instantaneously the value of the 1st peak power, n_p, and the energy released in the 1st peak, E_p, are denoted as follows;

$$n_p = \frac{(\rho-\beta)^2}{2\alpha K\ell} \tag{5}$$

$$E_p = \frac{2(\rho-\beta)}{\alpha K} \tag{6}$$

where α is reactivity temperature coefficient, K reciprocal heat capacity. These expressions were introduced analytically by Nordheim and Fuchs (Fuchs, 1946; Nordheim, 1946). The estimated value for the released energy during the 1st power peak using equation (6) was compared to the experimental data of TRACY experiment and it was found that they showed good agreement to each other for the excess reactivity greater than 2$ as shown in Fig. 4.

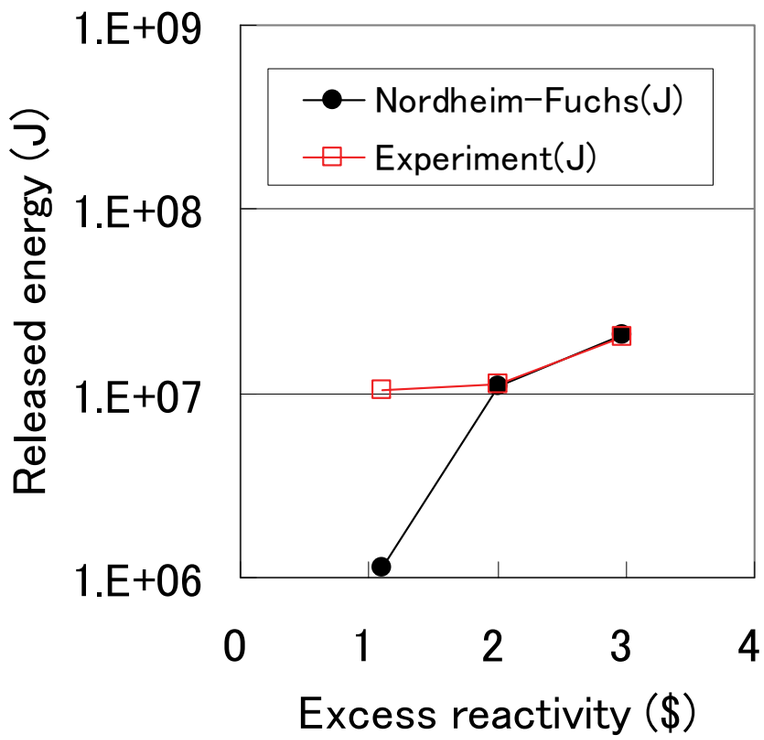

Fig. 4. Estimated value based on Nordheim-Fuchs model compared to TRACY experimental data for released energy during the 1st power peak (Yamane, 2009).

Because the instantaneous reactivity insertion corresponds to the largest insertion rate, the expression (6) gives the largest energy for the same excess reactivity case. And for our purpose, to evaluate the number of fission in the simplest manner, it is enough useful.

3.2 Monotonically decreasing

After the 1st peak, the power decreases monotonically. Based on one-point kinetics, the simple expression for the power n during the monotonically decreasing can be introduced.

Let us start with the following equations (Yamane, 2009);

$$\begin{aligned} \frac{dn}{dt} &= \frac{\rho - \beta_{eff}}{\ell} n + \lambda C \\ \frac{dC}{dt} &= \frac{\beta_{eff}}{\ell} n - \lambda C \end{aligned} \tag{7}$$

where λ is the average of λ_i in equation (1);

$$\lambda = \left[\frac{1}{\beta} \sum_{i=1}^{6} \frac{\beta_i}{\lambda_i} \right]^{-1} \tag{8}$$

For the excess reactivity larger than 1$, integrating the equations (7) gives

$$n = n_{pe} \exp\left(\frac{\lambda}{\beta / \rho - 1} t \right) \tag{9}$$

for the power after the 1st peak, where n_{pe} denotes the power at the end of the 1st peak. For the uranyl nitrate solution used for TRACY experiment, the value of λ is 0.08 (1/s), β, 0.0076. During monotonically decreasing of the power, the total reactivity is negative and its nominal value is large, for example, minus several ten $. So, the equation (8) can be denoted in simpler form as follows;

$$n = n_{pe} \exp(-\lambda t) \tag{10}$$

Monotonically decreasing of the power continues for long time, hundreds or more, and in such time scale, the smallest decay constant λ_1 is dominant. So, released energy during monotonically decreasing, E_d, is denoted as follows;

$$E_d = n_{pe} / \lambda_1 \left(1 - \exp(-\lambda_1 t_2)\right) \tag{11}$$

where t_2 is the time for monotonically decreasing of the power, λ_1, the smallest decay constant, 0.0127s for TRACY condition. Because the contribution of the power to the released energy is negligibly small for large t_2, t_2 can be ∞ and we have;

$$E_d = \frac{n_{pe}}{\lambda_1} \tag{12}$$

The power profile calculated by using the equation (10) is compared to TRACY experimental data as shown in Fig. 5. The calculation reproduces experimental power profile well. The difference after 400 seconds is due to cooling effect. For the released energy, as shown in Fig. 5, the value estimated by using the equation (12) shows good agreement to the experimental value.

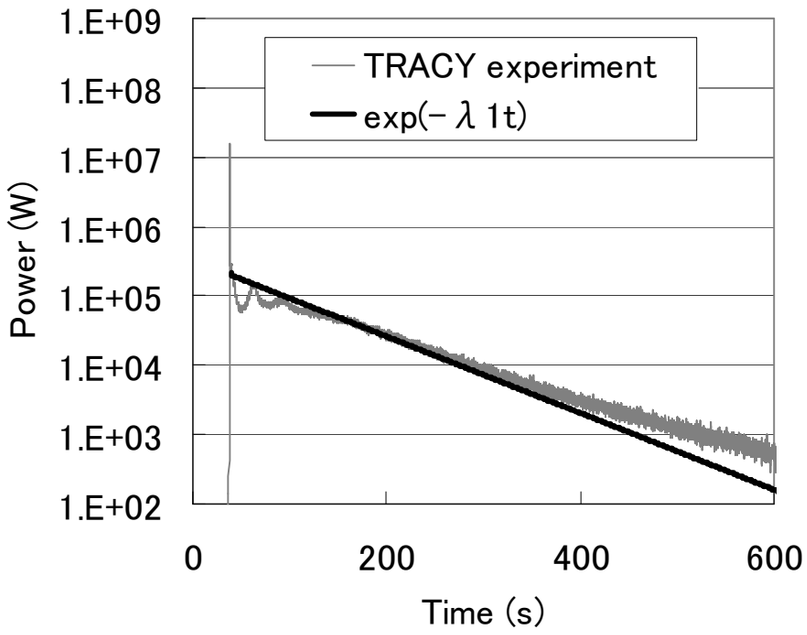

Fig. 5. Estimated power profile based on equation (10) compared to TRACY experimental data during monotonically decreasing of the power (Yamane, 2009).

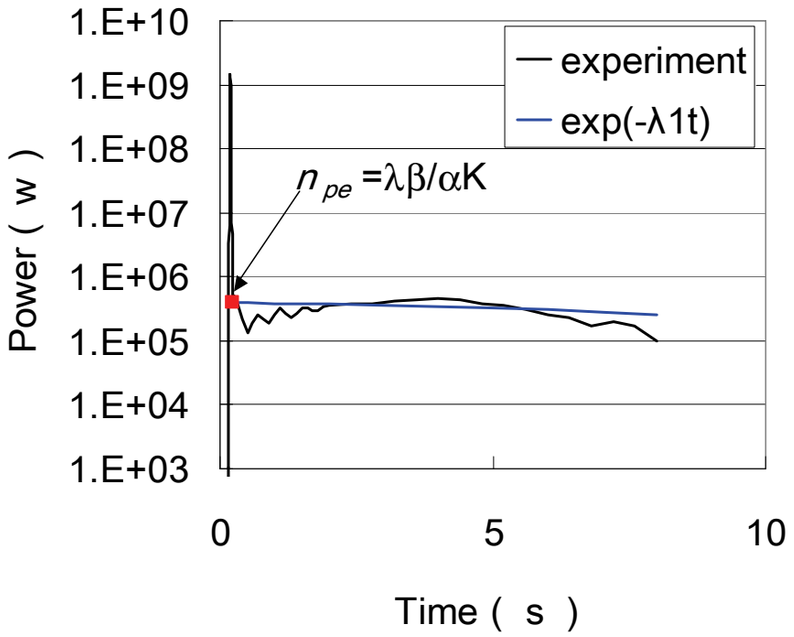

Fig. 6. Good case for estimated power profile based on equations (10) and (13) compared to TRACY experimental data during monotonically decreasing of the power (Yamane, 2009).

A candidate of the theoretical expression of n_{pe} has been introduced as follows;

$$n_{pe} \approx \frac{\lambda\beta}{\alpha K} \tag{13}$$

The calculated value using the equation (13) shows good agreement to the experimental value for some cases as shown in Fig.6, however, that is not always. As shown in Fig.7, there is some clear difference between those values. The expression of n_{pe} can be much improved.

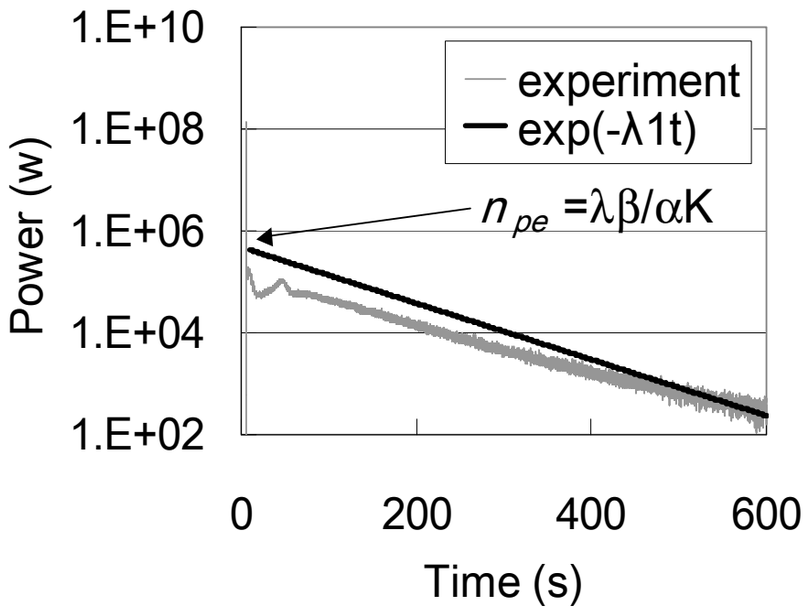

Fig. 7. Not good case for estimated power profile based on equations (10) and (13) compared to TRACY experimental data during monotonically decreasing of the power (Yamane, 2009).

3.3 Recovery and plateau

When the power decreases and it is very low, the fuel solution becomes being cooled and as its temperature decreasing the system becomes criticality again. Such cooling effect is mainly due to thermal conduction between the fuel solution and the container or surrounding materials. The fission power reaches such cooling power in course of time. As for TRACY, such cooling power is about 1kW as seen in Fig.6. If the cooling power is calculated by using CFD code in advance, the fission energy being released can be estimated easily.

It has been confirmed by calculation that the cooling effect is mainly due to thermal conduction between the fuel solution and the container. A kinetics calculation by using AGNES code into which a thermal conduction model based on Fourier's law has been implemented is compared to an experiment by using TRACY as shown in Fig.8. And it can be seen that the calculation reproduced the power profile obtained experimentally very well.

If the tank which contains the fuel solution is cooled forcibly, the fission power is the same as its cooling power. For example, the tank into which the fuel solution was poured had a water jacket in the JCO accident. The fission power kept high value because the cooling system kept working. After cutting the pipe between the jacket and the water supplier, the fission power got down.

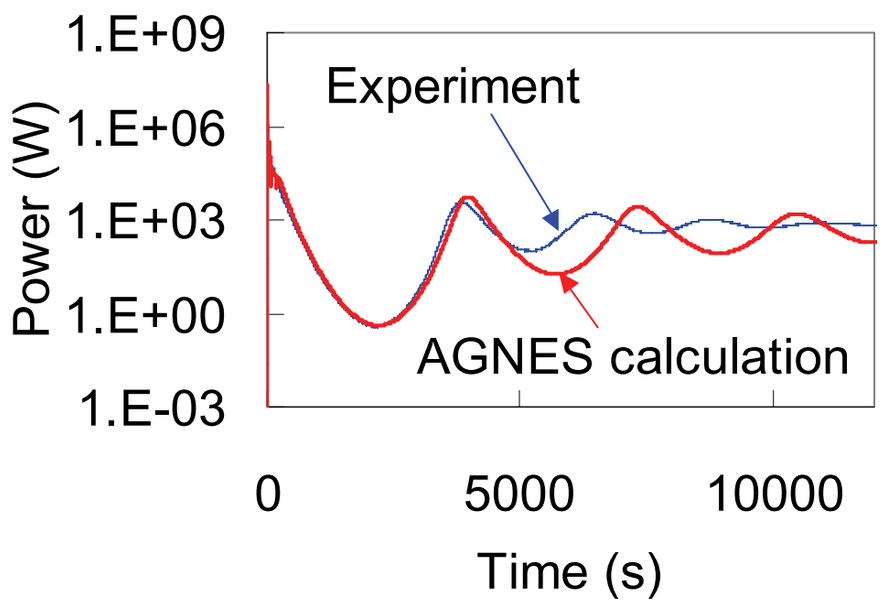

Fig. 8. Not good case for estimated power profile based on equations (10) and (13) compared to TRACY experimental data during monotonically decreasing of the power (Yamane, 2008).

3.4 New method

The total number of fission can be estimated as the sum of three values described above. Table 1 shows the whole expressions. To use these expressions, the excess reactivity, ρ, must be known in advance. In the case of planning the nuclear facility, it is common that credible maximum excess reactivity is calculated by using a montecarlo code or a deterministic code. In the case of criticality accident, such calculation should be done to confirm the effect of counter action or to understand what is happening. Anyway, it can be expected that we can obtain an estimated excess reactivity.

Range	Expression
Around 1st peak	$E_p = \frac{2(\rho_\$ - 1)}{\alpha K}$
Monotonically decreasing	$E_d = \frac{n_{pe}}{\lambda_1} \quad n_{pe} \approx \frac{\lambda\beta}{\alpha K}$
Plateau	E_c = Cooling power x duration time
Total	E_p+E_d+E_c

Table 1. New expression for simplified estimation of fission yield. $\rho_\$$ is excess reactivity in the unit \$.

4. Example of application

The method proposed in the previous section was applied to a TRACY experiment to evaluate it. Here considered are two cases, fast and slow transient cases. Before going to evaluation, the outline of TRACY experiment is explained.

4.1 TRACY experiment

TRACY, transient experiment critical facility, is a critical assembly which fuel is uranyl nitrate solution (Nakajima et al., 2002a, 2002b, 2002c). Its enrichment of ^{235}U is 98.9% and uranium concentration is in the range of 375 to 433 g/Lit. Its free nitric acid molarity is in the range of 0.6 to 0.9 mol/Lit. Such solution is contained in a cylindrical tank of SUS and its inner diameter is 52cm as shown in Fig. 9. A guide tube for a neutron absorber rod, "transient rod," is in its centre and its outer diameter is 7.6cm. For an transient experiment, reactivity is given by three ways such as (1)pulsed withdrawal of the transient rod, Pulse Withdrawal mode, (2)slow withdrawal of the transient rod, Ramp Withdrawal mode, (3)pumping the fuel solution from the bottom of the tank, Ramp Feed mode.

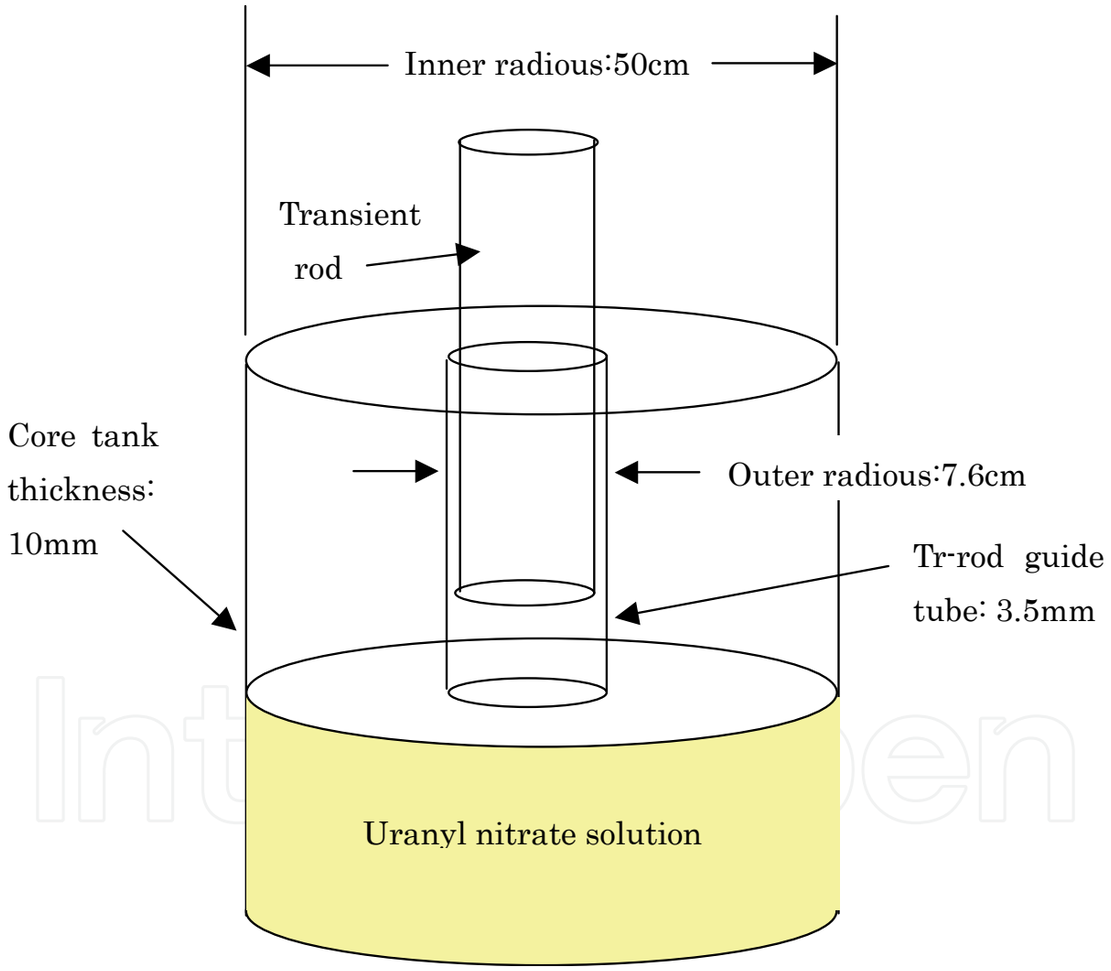

Fig. 9. Schematic view of TRACY

4.2 Fast transient case

For TRACY experiment R203, 2.97$ was inserted by pulsed withdrawal of the transient rod. The estimated values to use the simplest method are plotted in Table 2.

Parameter	Estimated value
ρ	2.97\$
β_{eff}	7.6×10^{-3}
$-\alpha$	-6.3×10^{-2} \$/°C
K	2.0×10^{-6} °C/J
λ	8.0×10^{-2} 1/s
λ_1	1.27×10^{-3} 1/s

Table 2. Parameters used to apply new method to R203.

For R203, free excursion was terminated 8 seconds after the insertion of reactivity as shown in Fig. 10. To consider the effect of the termination in short time, the equation (11) should be used instead of the equation (12). For TRACY experiment, reactivity temperature coefficient, α, should be multiplied by 1.5 to consider the effect of the temperature distribution in the solution at the 1st peak of power. For this case, E_c is zero, because the effect of cooling didn't appear. The result of calculation and the experiment are shown in Table 3. For the total number of fission, new method provides almost same value as the experiment. If we use the equation (12), $E_d = 1.2 \times 10^{17}$ and the total number of fission is estimated to be 7.7×10^{17}. That is enough close to the experimental value for our purpose.

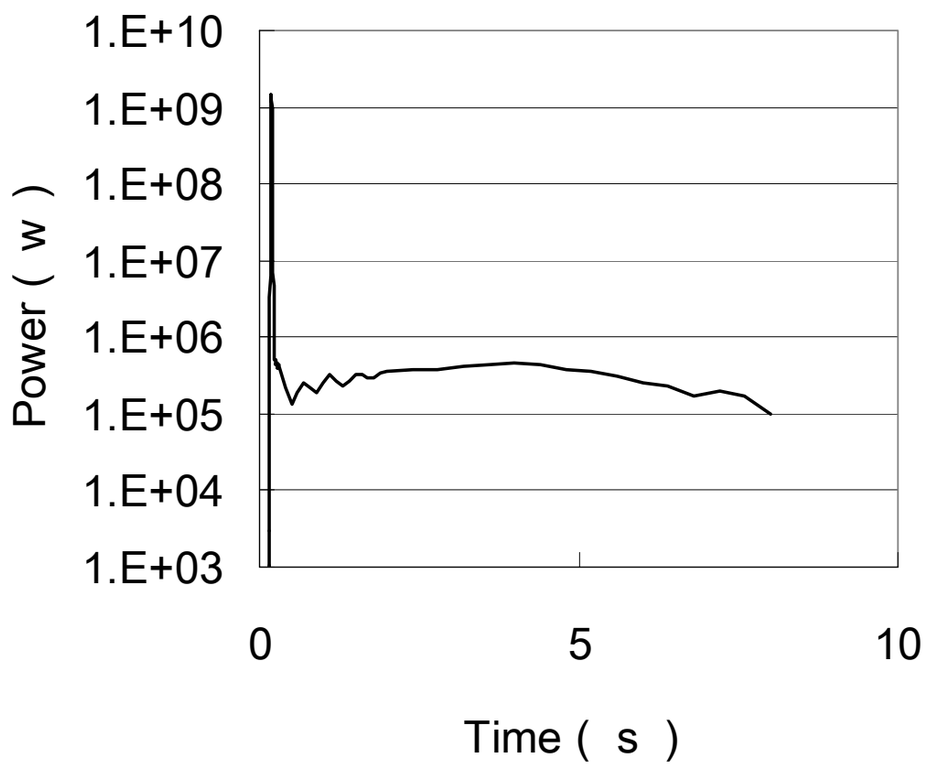

Fig. 10. Power profile of R203 (Nakajima et al., 2002a).

	New method	Experiment
Total number of fission	6.5×10^{17}	6.3×10^{17}
E_p	6.5×10^{17}	-
E_d	1.2×10^{15}	-
E_c	0	-

Table 3. Simplified method applied to fast transient case of TRACY experiment R203.

4.3 Slow transient case

For TRACY experiment R164, 1.52$ was inserted by ramp feed of fuel solution. The estimated values to use the simplest method are plotted in Table 4.

Parameter	Estimated value
ρ	1.52$
β_{eff}	7.6×10^{-3}
$-\alpha$	-6.3×10^{-2} \$/°C
K	1.8×10^{-6} °C/J
λ	8.0×10^{-2} 1/s
λ_1	1.27×10^{-3} 1/s

Table 4. Parameters used to apply new method to R164.

For R164, free excursion continued about 15000s as shown in Fig. 11. In Fig. 11, the power decreases monotonically until about 2500s and its recovery and plateau can be seen between 4000s and 15000s. To estimate E_p, reactivity temperature coefficient, α, should be multiplied by 1.5 to consider the effect of the temperature distribution in the solution at the 1st peak of power for TRACY. For this case, E_c is about 1kw as seen in Fig. 11. The result of calculation and the experiment are shown in Table 5. For the total number of fission and E_p+E_d, new method provides a value close to the experiment. That is enough close to the experimental value for our purpose.

	New method	Experiment
Total number of fission	6.0×10^{17}	6.9×10^{17} *
$E_p + E_d$	2.6×10^{17}	5.3×10^{17} *
E_p	1.9×10^{17}	-
E_d	6.9×10^{16}	-
E_c	3.4×10^{17}	-

*tentative value

Table 5. Simplified method applied to fast transient case of TRACY experiment R164.

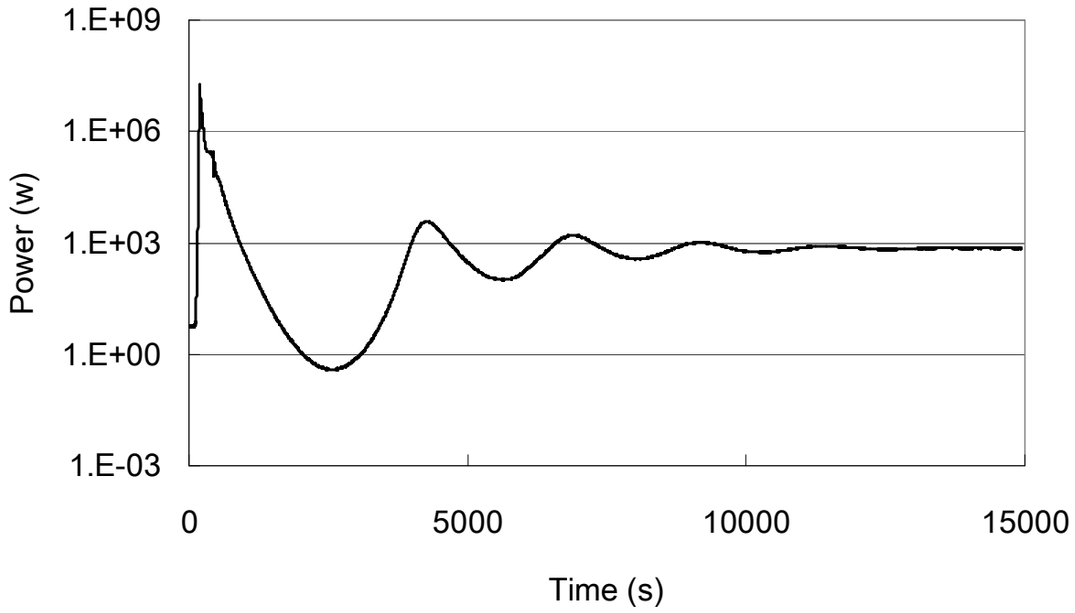

Fig. 11. Power profile of R164 (Nakajima et al., 2002c).

5. Conclusion

A concept for new simplified method to evaluate fission yield at a criticality accident was introduced and explained briefly. Two examples of its application were compared to experiments to show its applicability. The results showed the estimated values were enough close to the experimental values. There still are some points to be developed or improved; for example, n_{pe} can be much accurate, the power during plateau should be denoted in a simple form, etc.

6. Acknowledgment

The author greatly appreciate Dr. Uchiyama who prompted to write this chapter, Prof. Nakajima who is giving useful suggestion always, Mr. Futagami who is a great help to measure and record the experimental data, and staffs committed to the operation of TRACY for their dedicated efforts.

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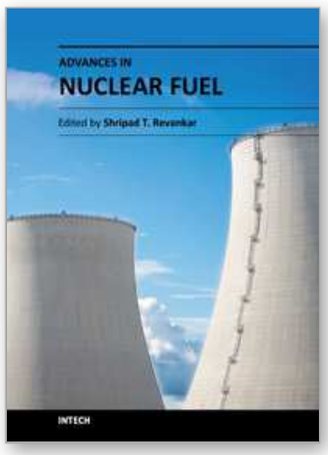

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Worldwide there are more than 430 nuclear power plants operating and more plants are being constructed or planned for construction. For nuclear power to be sustainable the nuclear fuel must be sustainable and there should be adequate nuclear fuel waste management program. Continuous technological advances will lead towards sustainable nuclear fuel through closed fuel cycles and advance fuel development. This focuses on challenges and issues that need to be addressed for better performance and safety of nuclear fuel in nuclear plants. These focused areas are on development of high conductivity new fuels, radiation induced corrosion, fuel behavior during abnormal events in reactor, and decontamination of radioactive material.

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