



Development of Criticality Accident Analysis Code AGNES

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A one-point kinetics code, AGNES2, has been developed for the evaluation of the criticality accident of nuclear solution fuel system. The code has been evaluated through the simulation of TRACY experiments and used for the study of the condition of the JCO criticality accident. A code, AGNES-P, for the criticality accident of nuclear powder system has been developed based on AGNES2. It is expected that these codes be useful for the evaluation of criticality safety for fuel reprocessing and fabrication plants.

KEYWORDS: AGNES2, AGNES-P, One-point Kinetics, Criticality Accident, Solution System, Powder System, JCO, TRACY

1. Introduction

For the aim of the evaluation of criticality accident for reprocessing plants, a numerical simulation code, AGNES2¹⁾, has been developed at the Japan Atomic Energy Research Institute, JAERI.

The efforts to understand the unique character of the criticality accident of solution fuel have yielded a number of numerical codes such as CRITEX²⁾, CREST³⁾, SKINATH⁴⁾, TRACE⁵⁾ and FETCH⁶⁾, and for such purpose experimental works have also been done using CRAC⁷⁾, SHEBA⁸⁾, SILENE⁹⁾ and TRACY¹⁰⁻¹²⁾ reactors. From the JCO criticality accident in 1999, it was found that the heat loss from the fuel by the cooling system sustained the high level of the power and there was the requirement for the method to evaluate quickly the effect of a countermeasure on the accident. And the basic mechanism and effect of radiolytic dissociation gas voids are still not clear. The AGNES2 code has been developed for the evaluation of the criticality accident of solution system and is expected to be useful for the designing of a fuel reprocessing plant, the quick evaluation of the effect of a countermeasure against the accident and the study of the nature of the criticality accident of solution system.

A numerical code, AGNES-P, for nuclear powder system based on AGNES2 has been developed in recent work. In this field, the numerical codes such as POWDER¹³⁾ have been developed for uranium

powder system. AGNES-P code has been developed for general powder system, both uranium and plutonium with the additive powder which may contain hydrogen.

This paper presents the current status of the development of AGNES2 and AGNES-P codes. In the next section, the outline of the AGNES2 code and the examples of simulation using AGNES2 are described. The description of AGNES-P is in section 3.

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2. Analysis of Solution System

2.1 Outline of AGNES2 code

2.1.1 One-point kinetic equation

The calculation geometry consists of three regions such as fuel, container and coolant. The calculation regarding neutron is done in fuel region only. The basic equation of AGNES2 is the one-point kinetics equation as follows;

$$\frac{dP}{dt} = \frac{\rho - \beta}{\Lambda} P + \sum_{i=1}^6 \lambda^i C^i + S, \quad \frac{dC^i}{dt} = \frac{\beta^i}{\Lambda} P - \lambda^i C^i.$$

The following equation was derived from the above, and is used to calculate the power density, P , and the delayed neutron precursor densities, C_i , in course of time;

$$P_{k+1} = e^{\omega_k \Delta t} \left[P_k \left\{ 1 + \frac{\Delta t}{2\Lambda} (\rho_{k+1} - \rho_k) \right\} + \sum_{m=1}^G \lambda^{(m)} C_k^{(m)} \left(\frac{e^{\Delta t (\gamma_k^{(m)} - \omega_k)} - 1}{\gamma_k^{(m)} - \omega_k} - \Delta t \right) + S \left(\frac{e^{-\Delta t \omega_k} - 1}{-\omega_k} - \Delta t \right) \right],$$

$$C_{k+1}^{(m)} = e^{\gamma_k^{(m)} \Delta t} \left[C_k^{(m)} + \frac{P_k \beta^{(m)}}{\Lambda} \left(\frac{e^{\Delta t (\omega_k - \gamma_k^{(m)})} - 1}{\omega_k - \gamma_k^{(m)}} - \Delta t \right) \right],$$

where

$$\omega_k = \frac{\rho_k - \beta}{\Lambda} + \frac{1}{P_k} \sum_{m=1}^G \lambda^{(m)} C_k^{(m)} + \frac{S}{P_k}, \quad \gamma_k^{(m)} = \frac{\beta^{(m)} P_k}{\Lambda C_k^{(m)}} - \lambda^{(m)}.$$

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P_k is the abbreviated form of $P(t_k)$ and $\Delta t = t_{k+1} - t_k$. (m) denotes the number of the group of delayed neutron.

Nuclear solution fuel region is assumed to be homogeneous and only cylindrical shape can be calculated by using R-Z coordinate. Total reactivity, ρ , is the sum of initial reactivity, ρ_0 , added reactivity, $\Delta\rho_a$, temperature feedback reactivity, $\Delta\rho_t$, and reactivity due to radiolytic dissociation gas voids, $\Delta\rho_v$, and is represented as follows;

$$\rho = \rho_0 + \Delta\rho_a + \Delta\rho_t + \Delta\rho_v.$$

As temperature reactivity feed back, many effects such as Doppler effect, scattering cross section effect, density effect, and volume expansion effect can be taken into account, if the reactivity coefficients of those are evaluated beforehand.

2.1.2 Thermal Calculation

Temperature is considered in fuel, container and coolant region. The fuel and coolant regions connect to only the container region, and the energy released by fission in the fuel region goes to the coolant region through the container region. The temperatures denoted by T_i are calculated as follows;

$$V_i(\rho C_p)_i \frac{\partial T_i}{\partial t} = \gamma_i P V_i + (hA)_{i-1}(T_{i-1} - T_i) - (hA)_i(T_i - T_{i+1}),$$

where i denotes region number; 1 is fuel, 2, container and 3, coolant region. The first term of the right hand side is the energy released in the region i , and the second and third are the energy transferred from or to the next region. In the coolant region, this equation is written in the following form;

$$V_3(\rho C_p)_3 \frac{\partial T_3}{\partial t} = \gamma_3 P V_3 + (hA)_2(T_2 - T_3) - \omega C_{p3}(T_{out} - T_{in}),$$

where ω is the flow rate of the coolant and C_{p3} is the specific heat of the coolant. T_{out} and T_{in} represent the temperatures of the coolants at the outlet and inlet, respectively. The temperature T_3 of the coolant region is assumed to be the average of T_{out} and T_{in} as follows;

$$T_3 = \frac{1}{2}(T_{out} + T_{in}).$$

2.1.3 Cooling by Conduction and Natural Convection

From the JCO accident, it was observed that the high power was sustained by the heat loss from solution by the cooling system. That implies the continuous removing of heat energy from the fuel gives rise to large amount of radiation, which is important parameter for the planning of the counter action. Therefore cooling is important problem.

Adding to the original cooling model described in the previous section, a new model of cooling has been implemented in recent development work. In the new model, the heat energy is removed from the container region by the structural materials connected directly to the container and by the natural convection of air.

A virtual structural material is considered to take into account the heat loss by many structural

materials. The cooling through this virtual region is modeled as the Newton's law of cooling;

$$q = kA \frac{t_1 - t_0}{x},$$

where q is conduction heat transfer rate, k , thermal conductivity, A , the area of heat exchange, t_1 , the temperature of the container, t_0 , the temperature of the atmosphere and x , the distance between the container and the atmosphere.

The heat exchange between the container and coolant regions is calculated using the model of the natural convection of air near vertical plane.

The conduction heat transfer rate q is denoted as follows;

$$q = hA(t_2 - t_\infty),$$

where h is convection heat transfer coefficient, t_2 , the temperature of the container, t_∞ , the temperature of atmosphere. The symbol h is denoted as a function of Nusselt number as follows;

$$h = \frac{Nu \cdot k'}{H},$$

where Nu is Nusselt number, k' , the heat conductivity between the container and air, and H , the height of the heat exchange area. For the expression of Nu , the following model recommended by Churchill¹⁵⁾ is used;

$$Nu = \left\{ 0.825 + \frac{0.387 Ra^{1/6}}{\left[1 + (0.492/Pr)^{9/16} \right]^{1/4}} \right\}^2,$$

where Ra is Rayleigh number and Pr , Prandtl number. They are denoted as follows;

$$Ra = \frac{g\beta_e(T_2 - T_\infty)H^3}{\alpha\nu}, \quad Pr = \frac{\nu}{\alpha}.$$

g is gravitational acceleration, β_e , volumetric thermal expansion coefficient and ν , the kinematic viscosity of air. α is thermal diffusivity and is denoted as follows;

$$\alpha = \frac{k}{\rho C_p}.$$

2.1.3 Radiolytic dissociation gas void

It is known for the criticality accident of solution that at some level of the power, radiolytic gas voids are formed along the tracks of fission products and they grow enough to give some feedback effect to the criticality of the solution system. The feedback of such gas voids is treated as follows; the fuel region is divided into some meshes in R-Z coordinate. The balance equation of the void fraction F_{ij} in a mesh (i,j) is denoted as follows;

$$\frac{\partial F_{i,j}}{\partial t} = \nu_i \cdot \gamma_{i,j} \cdot P \cdot (C_{i,j} - C_0) \cdot \theta(C_{i,j} - C_0) - \nu_{i,j} \frac{\partial F_{i,j}}{\partial z},$$

where ν_i is void-energy transfer coefficient, γ_{ij} , power fraction and C_0 , the saturation mol density of dissociation gas. θ is Heviside function. $C_{i,j}$ is the mol density of dissociation gas, which satisfies the following equation;

$$\frac{\partial C_{i,j}}{\partial t} = G \cdot \gamma_{i,j} \cdot P - \frac{C_{i,j}}{F_{i,j}} v_{i,j} \frac{\partial F_{i,j}}{\partial z} - \frac{\partial C_{i,j}}{\partial \tau}$$

where G is gas production rate and τ , the dissolution time of dissociation gas.

2.2 Examples of Simulation

2.2.1 Simulation of TRACY experiment

From the view point of the analysis of the criticality accident of nuclear fuel solution, there are two important time scales. The power profile during the first several minutes affects the dose of workers until their evacuation. The power during the following several hours is important parameter for the planning of the counter action against the accident. In this section, two corresponding cases of TRACY experiments and the simulations using AGNES2 code are presented.

TRACY is a reactor which uses 10% enriched uranyl nitrate solution as fuel. Its core tank has a cylindrical shape. There is a transient rod for reactivity insertion in the center of the core tank which contains B_4C inside it.

An example of the measured power profile of a TRACY experiment and the result of the simulation for it using AGNES2 code are shown in Fig.1. In the experiment, the fuel solution was pumped into the core tank from its bottom to insert 2.6\$ of total excess reactivity. Uranium concentration was 422 gU/Lit. Acid molarity was 0.77 mol/Lit. The feed rate of the fuel solution was 60 Lit./min, and corresponding average reactivity insertion rate was 0.18 \$/sec. No external neutron source was used.

In Fig.1, after the large first peak, small oscillation due to some feedback effect due to radiolytic gas voids is observed in the power profile.

The result of the simulation shows good agreement with the experiment until the second peak of the power. After the second peak, the result of simulation is longer in the period of the oscillation than the experimental result. For the simulation, a weight for the reactivity temperature coefficient was used to take into account the temperature distribution due to the distribution of power at the first power peak. The value of the weight was 1.5 at the first peak power, then decreases exponentially to be 1.0 in five seconds after the first peak, because the solution was stirred by convectional flow due to temperature difference and radiolytic gas voids after the first power peak. Since the heat loss from the core tank during the first several minutes is negligibly small for this case, an assumption is possible such that the surface of the tank is adiabatic.

For the same experiment, the temperature was also measured and plotted in Fig.2. The figure shows the values measured at two different levels in the solution. The temperature measured near the surface of the solution is higher than one near the bottom of the core tank, because a hot part of the solution moved up to the surface. Simulated value of average

temperature plotted in Fig.2 is almost between two experimental values. The temperature increases like a step, and each step corresponds to each peak in power profile. Experimental value measured at 45cm from the bottom increases about 2 seconds after other data at the first step. Since convectional flow of solution arise at the first power peak, a hot part of the solution ascends to the solution surface. Such effect seems small at the bottom.

For another experiment, the power profile during much longer term was also measured and plotted in Fig.3. In this experiment, Uranium concentration was 382.3 gU/Lit. Acid molarity was 0.62 mol/Lit. The excess reactivity of 1.5\$ was inserted by feeding solution with the rate of 0.15 \$/sec. No external neutron source was used. The measurement was continued about 5 hours after the reactivity insertion. It was observed that a deep bottom in the power profile at about 2000 seconds and the following oscillation. In this time scale, the heat loss from the core tank is dominant for the change of the power.

The simulated value shows good agreement with experiment until 4000 seconds. However, as shown in Fig.4, the simulated value of temperature is higher than the experimental value. Such difference may be due to the problem of the new cooling model.

2.2.2 JCO criticality accident

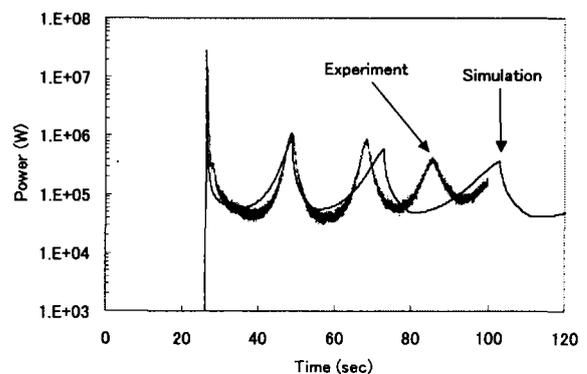


Fig.1 Power profiles of a TRACY experiment and simulation in the first 2 minutes.

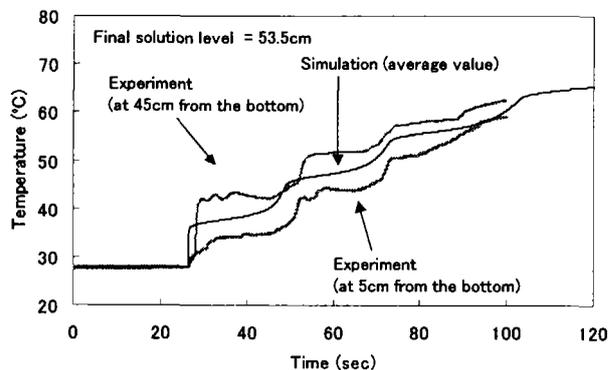


Fig.2 Temperature profiles of a TRACY experiment and simulation in the first 2 minutes.

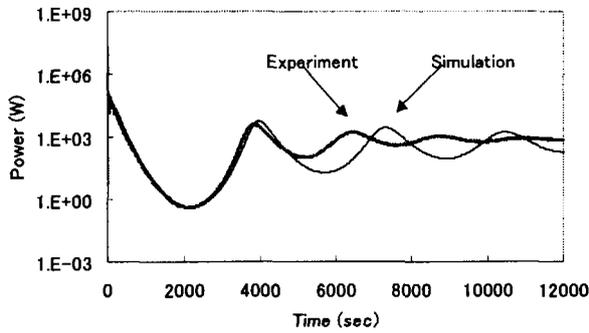


Fig.3 Power profiles of a TRACY experiment and simulation during 200 minutes.

A criticality accident which occurred at a precipitation tank in the JCO company Tokai-works in 1999 was simulated using AGNES2 code for the investigation of the condition which initiated the accident. In this section, an example of the simulation is presented.

The geometry of the precipitation tank was very complicated¹⁴⁾, however, a cylindrical shape was used for the simulation. It contained 18.8% enriched uranyl nitrate solution at that time. The uranium concentration was 370gU/Lit. and the acid molarity was 0.5mol/Lit. The solution was poured into the tank using a bucket when the criticality achieved.

Since there were no information for the inserted reactivity and its insertion rate, parameter survey was performed. The range of the parameters is as follows; the excess reactivity between 1.5 and 4.5 \$, reactivity insertion rate between 15 and 70 \$/sec and total thermal conduction rate from the solution to the coolant between 20 and 700 J/(s·m²·K). An example of the result of such simulation is shown in Fig.5.

Figure 5 shows the two different reactivity insertion cases and the estimated value of the power based on the record of a gamma-ray area monitor of the JCO¹⁴⁾. The simulated values show good agreement with the value of the gamma-ray area monitor except the detail fluctuation. From this result, the inserted reactivity is estimated to be close to 1.5 – 3.0\$.

Another example of the power profile during

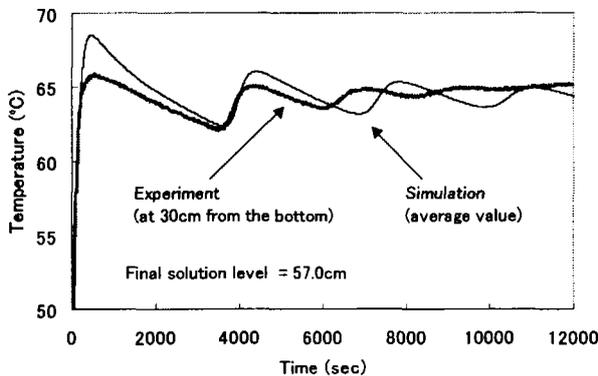


Fig.4 Temperature profiles of a TRACY experiment and simulation during 200 minutes.

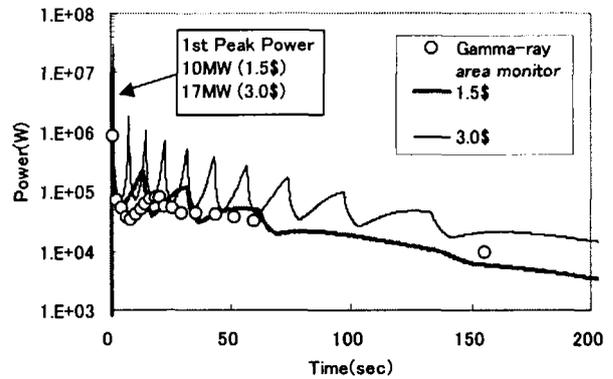


Fig.5 Simulated power profile and the record of a gamma-ray area monitor during 200 seconds.

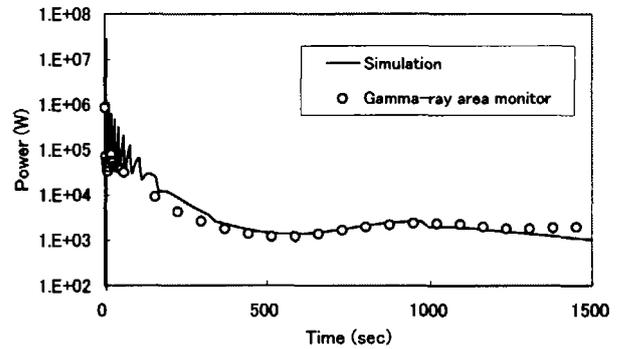


Fig.6 Simulated power profile and the record of a gamma-ray area monitor during 25 minutes.

1500seconds is shown in Fig.6. The value of total thermal conduction rate for the simulation was 344 J/(s·m²·K) for this case. The result of simulation shows good agreement with the value of the gamma-ray area monitor. From this result, the total thermal conduction rate is estimated close to such value. But further investigation is needed for much better estimation.

3. Analysis of Powder System

3.1 Development of AGNES-P code

For the criticality accident of MOX powder system, a one-point kinetics code, AGNES-P, has been developed at JAERI.

One of the main targets of AGNES-P is the homogenizing process of oxide uranium and plutonium powders in a MOX fabrication plant. Since the delayed neutron fraction of Pu-239 is smaller than that of U-235, the margin for prompt critical becomes smaller. The additive powder such as zinc stearate, used as a lubricant, contains hydrogen and acts as a moderator. The behavior of particles of MOX powder and additives at criticality accident is not known well, because there is no experimental result. Zinc stearate is solid at room temperature, melts at about 120-130°C, which is higher than the boiling point of water, and dissociates

into gas at about 400-420°C¹⁶⁻¹⁸). Such character could give quite different result in a criticality accident from the wet powder system.

AGNES-P has the same feature in neutronic and thermal calculations as AGNES2, and it implies the assumption of homogeneity for fuel region. Part of radiolytic gas void calculation is omitted, while the dissociation of additive powder is taken into account.

Dissociation is different phenomena from phase change such as melt or vaporization. In AGNES-P, however, a simple assumption is adopted that the excess energy is consumed for a process such as melt, vaporization or dissociation at the specific temperature point of the process. According to this assumption, the energy balance equation in the fuel region introduced above is re-written in the following form;

$$He = \gamma_1 P V_1 - (hA)_1 (T_1 - T_2),$$

$$V_1 (\rho C_p)_1 \frac{\partial T_1}{\partial t} = He \quad \text{for } He < 0,$$

$$E_i = He \cdot \Delta t \quad \text{for } He \geq 0,$$

where E_i represents the latent heat. In this model, the rate of phase change, or dissociation, is determined by the combination of the rates of the fission release and the cooling. The mass of each phase and the feedback reactivity are calculated from E_i .

3.2 Example of Simulation

3.2.1 Uranium Powder System

A uranium powder system was calculated using AGNES-P and POWDER code to evaluate the power, the number of fissions and the achieved temperature. The calculation condition was as follows; The mixture of uranium oxide powder and water was contained in a cylindrical vessel of which both of the radius and height was 40cm. The temperature of the mixture was 20°C and its density was 1.4g/cm³. It was assumed that a small amount of water flew into the mixture and it became critical.

The reactivity insertion continued until the termination of the calculation. The insertion rate was 0.01\$/sec. This case was calculated using AGNES-P and POWDER codes. Figure 7 shows the result of the power, the number of fissions and the temperature of the powder. In Fig.7, the temperature calculated using AGNES-P code is the average value of the fuel region and the result of POWDER code is the value of the center of the representative particle of the powder. The power increases up to the order of 10¹⁷ fission/sec. The calculation by POWDER code terminated at the beginning of nucleate boiling soon after the peak of the power. The inserted reactivity until the peak power was about 0.9\$. While no discrepancy is seen between the results of both codes, the comparison to experimental results is needed for the evaluation of AGNES-P code. Figure 8 shows the reactivity change calculated using AGNES-P. The

total reactivity decreases after the peak of the power due to the feedbacks of temperature and boiling.

3.2.2 MOX Powder System

The criticality accident with the homogeneous mixture of MOX and zinc stearate powder was considered and calculated using AGNES-P code to evaluate the power, the number of fission and the achieved temperature. A typical result of preliminary calculations is shown in this section.

The calculation conditions were as follows; The mixture of MOX and zinc stearate powder was contained in a cylindrical vessel of which both of the radius and height was 40cm. The initial temperature of the mixture was 20°C and its density was 1.4g/cm³. It was assumed that water flew into the mixture and criticality was achieved. In this assumption, the rate of reactivity insertion was 0.01\$/sec, the duration time was 10 sec and thus the total inserted reactivity in ten seconds was 0.1\$.

Figure 9 shows the result of the power, the number of fission and the temperature calculated using AGNES-P. Because of the smallness of the inserted reactivity, the power increases slowly to the peak, which corresponds to the rise of temperature. It is known that the density of zinc stearate is smaller than water, but there is no data of its specific heat. In this calculation, it is assumed that the specific heat of zinc stearate is the same as water. This assumption gives smaller value of heat capacity to zinc stearate than that of the water of the same volume. Thus zinc stearate is easy to be warmed rather than water in this calculation.

Figure 10 shows the reactivity change calculated

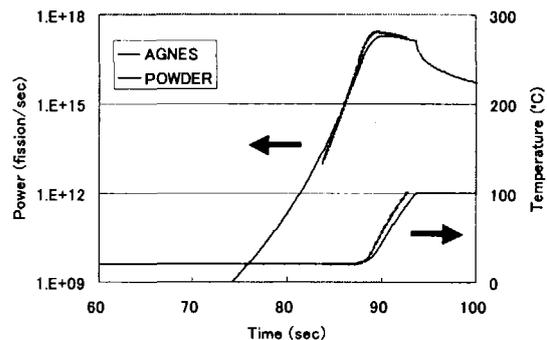


Fig.7 Power and temperature profiles calculated using AGNES-P and POWDER codes.

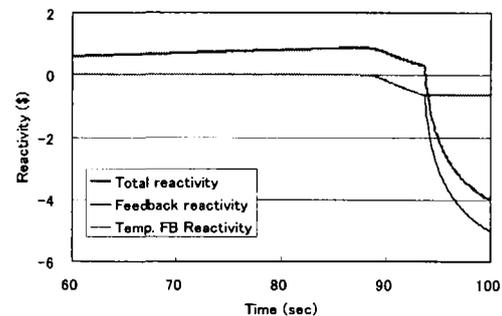


Fig.8 Reactivity profiles calculated using AGNES-P code.

using AGNES-P code. The total reactivity becomes 0.1\$ in the first 10 seconds, then decreases to be negative due to the reactivity of temperature feedback. As seen in Fig.9, the finally achieved temperature is less than 40°C, so zinc stearate keeps in solid state and there is no reactivity feed back due to phase change.

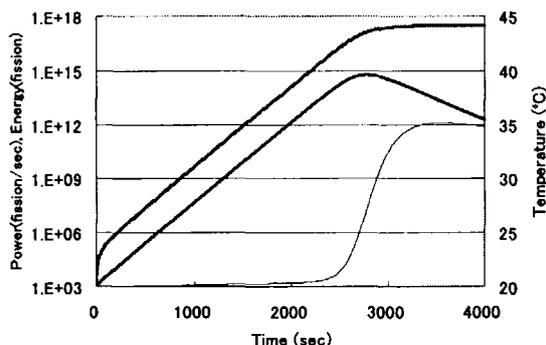


Fig.9 Power, energy and temperature profiles calculated using AGNES-P code.

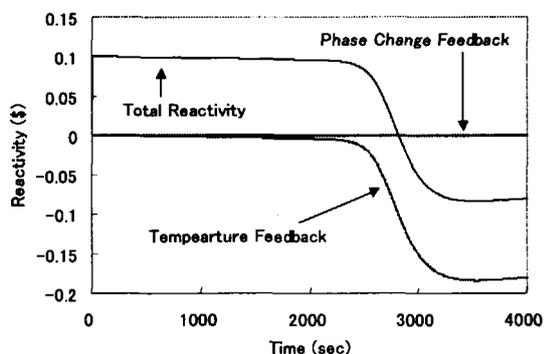


Fig.10 Reactivity profiles calculated using AGNES-P code.

4. Summary

A numerical simulation code, AGNES2, has been developed for the evaluation of the criticality accident of solution fuel system. Through the simulation of TRACY experiments and the JCO accident, it showed the applicability of the code to both short and long time phenomena. Still, part of cooling and void calculation should be improved.

AGNES-P code has also been developed for nuclear powder system and some preliminary calculations have been performed. The data output by these codes such as the power, fission, temperature, feedback reactivity, etc. are useful for the designing of a fuel processing process and the evaluation of the effect of a countermeasure against the criticality accident.

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