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DEVELOPMENT OF EVALUATION METHODS FOR THE TRANSFER BEHAVIOR OF CORROSION PRODUCT IN THE PRIMARY COOLING SYSTEM OF FAST BREEDER REACTOR

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ABSTRACT

Radioactive corrosion products (CPs) are a main cause of personal radiation exposure during maintenance with no breached fuel in sodium-cooled fast breeder reactor (FBR) plants. In order to establish techniques of radiation dose estimation for workers in radiation-controlled areas of the fast breeder reactor, the PSYCHE (Program System for Corrosion Hazard Evaluation) code was developed. For the inspection of the analysis model using PSYCHE code, we surveyed past reports on CP deposits in Japan Experimental Fast Reactor JOYO and sodium test loops. The SEM images of the external surface of the irradiated fuel cladding in JOYO show surface deposition of particles containing significant volumes of CP species. The CP particle deposition model (Particle model) was developed from the phenomenological considerations of the observation results for JOYO. We add the Particle model to the conventional PSYCHE analytical model. Moreover, the conventional PSYCHE code does not consider the sodium flow

of the coolant. The non-consideration of the sodium flow is a cause of reduction in calculation precision. To counter this problem we built the system that PSYCHE code linked NETFLOW++ code. This code is the one-dimensional network code which can simulate the plant transients of various types of nuclear reactors. In this study, we performed calculations of CP transfer behavior in JOYO using an improved PSYCHE code. The calculation results are consistent with the measured results for actual components in JOYO. The C/E (Calculated / Experimentally observed) value was improved by introduction of the Particle model and considered of the sodium flow.

INTRODUCTION

During the operation of sodium-cooled Fast Breeder Reactors (FBR), radioisotopes are produced from the constituent elements of activated fuel cladding and subassembly wrappers in the reactor core region. Some isotopes are released only due to the general recession of the surface "corrosion" and

some are preferentially released and then a composition gradient is established in the material, their release is limited by their diffusion in the steel. The radioisotopes generated by a corrosion reaction are called Radioactive Corrosion Product (CP). The most important CP species are ^{54}Mn and ^{60}Co ^{1,2)}. The released CP circulates along with the sodium flow and the CP is deposited on the piping and components of the primary cooling system. The deposited CP causes radiation fields near the piping and components, which greatly complicate maintenance procedures and contribute significantly to radiation exposure of the radiation worker.

In order to estimate radiation dose exposure of radiation worker in radiation-controlled areas at sodium-cooled FBR, the Program System for Corrosion Hazard Evaluation code (PSYCHE) was developed by the PNC (Power Reactor and Nuclear Fuel Development Corporation), currently JAEA (Japan Atomic Energy Agency)³⁾. The calculation of CP transfer behavior for conventional PSYCHE code based on the Solution-Precipitation model needs the fitting factor for precipitation rate of CP⁴⁾. This fitting factor must be determined based on measured values in reactors that have operating experience. Moreover, the fitting factor differs for each cooling system region and each CP species. For this reason, the inability to make accurate predictions for reactors without measured values is a major issue. In other words, it is necessary to adopt a more physically valid model for prediction of CP behavior in reactors without actual operating experience.

We surveyed past reports on CP deposits in JOYO (Experimental Reactor in Japan) and sodium test loops. The scanning electron microscope (SEM) images of the external surface of the irradiated fuel cladding in JOYO show surface deposition of particles containing significant volumes of CP species⁵⁾.

The conventional PSYCHE code is based on the solution-precipitation model^{1,2)}. We add the Particle model to the conventional PSYCHE analytical model⁶⁾. Moreover, the conventional PSYCHE code did not have the function of calculating sodium flow. Coolant temperature and flow rate are particularly important when considering CP solution, deposition and CP particle transfer phenomena. The deposition of CP species appears to be proportional to the velocity of the flow. To address this issue, in this study, along with transduction to the code of the particle model, we created a system that links the NETFLOW++ code to PSYCHE. This code is the one-dimensional network code which can simulate the plant static state and transient state of various types of nuclear reactors⁷⁾.

In this paper, we performed calculation of CP transfer behavior in the JOYO system using the improved PSYCHE code. From the results of these calculations, the ratio of particles transferred to the total CP deposition amount was estimated. Then, the effectiveness of the Particle model for the calculation of CP transfer behavior was evaluated. The concept of introduction of the calculation using the improved PSYCHE code is shown in Figure 1.

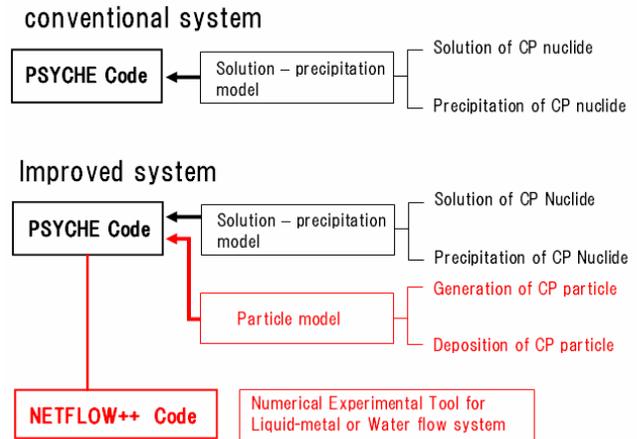


Figure 1 The concepts of the improved PSYCHE code

CALCULATIONAL METHODS

Solution-Precipitation model

The resulting expressions for the concentrations of CP species in steel and sodium, and the mass flux, for solution and precipitation, have been developed in the past¹⁾. The model is referred to as a Solution-Precipitation model. According to this model, the CP transfer behavior in a sodium loop is expressed as four steps (Figure 2):

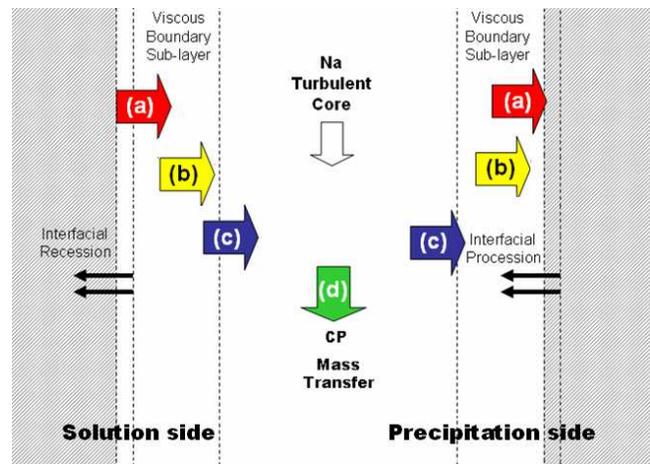


Figure 2 Solution-Precipitation model.

- (a) Solid state diffusion in the steel and surface loss or gain by solution or precipitation, respectively
- (b) Mass transfer at the sodium-steel interface
- (c) Mass transfer across the liquid viscous boundary sub-layer
- (d) Mass transfer along with the circulating sodium coolant

Solution and release calculations require solution of the diffusion equation in the steel, with the interfacial boundary condition obtained from the mass flux for (a) ~ (c). This Solution - Precipitation model for radioactive CP transfer behavior in FBR has been investigated by M. V Polley⁸⁾. In this study, we used the expressions for the concentrations of species in the steel and sodium and mass flux as reported in previous publications^{1,2)}.

Particle model

In the Particle model, particle behavior including the CP transfer behavior in a sodium loop is expressed as three steps (Figure 3):

- (A) Particle generation in steel surface and diffusion to sodium coolant
- (B) Particle transfer along with the circulating sodium coolant
- (C) Particle deposition onto steel surface from sodium coolant

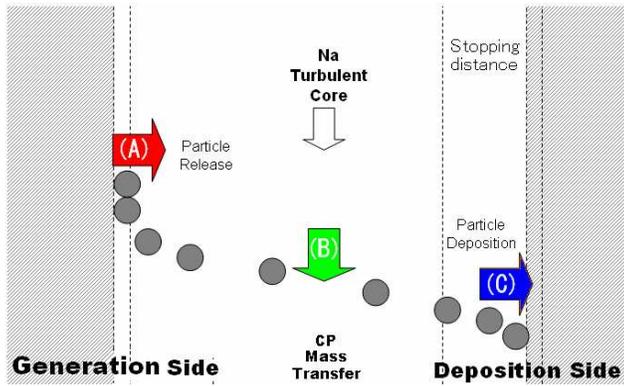


Figure 3 The Particle model.

- (A) Particle generation in steel surface and diffusion to sodium coolant

In the results of observations of the fuel cladding in JOYO, the steel surface showed pits (dissolved areas) and particulates (insoluble areas)⁹⁾. Therefore, we have assumed that particles are produced by corrosion of the steel surface in the core region.

Simple modeling of particle generation is performed as below. The geometry for each calculation region is modeled as a cylinder, in which the sodium and material properties are regarded as constant. Maruyama's equation for the dissolution of austenitic steel, used in PSYCHE code, is used to calculate the number of particles generated from the steel surface¹⁰⁾. The solution rate u_c (cm/s) is expressed as

$$u_c = 3.17 \times 10^{-12} [O]^{0.803} \exp \left(12.63 - \frac{22.0}{RT} + 0.00591 \frac{L}{D} \right) \quad \dots (1)$$

where, O is the oxygen concentration [w_t, ppm], R is the gas constant [J/mol · K], T is the fluid temperature [K], L is the distance along the region of interest [cm] and D is the diameter of pipe [cm]. Then, L/D is the downstream factor. This rate is expressed as a function of temperature, oxygen concentration in sodium and a downstream factor for isothermal regions. We assumed that there is a proportional relationship between the rate of particle generation and the solution rate u_c [cm/s]. We define the number of generated particles in the coolant inside the region under examination per time step interval (N_{gen}) as (The number of particles generated in one time step interval in a region) / (the amount of coolant to pass through the region in one time step interval). If ϕ [-] is the proportional constant, N_{gen} [particle · cm³] is expressed as

$$N_{gen} = \phi \pi DL u_c \Delta t \quad \dots (2)$$

where πDL [cm²] is the surface area of the region under examination, Δt [s] is time step interval. Then, ϕ is an operation period-independent coefficient to compensate for the inadequacy of the current generation-side model for particles.

This model is done as a first approximation; in the future we are planning to conduct precise modeling based on analysis of the surface of the irradiated fuel cladding in JOYO.

- (B) Particle transfer along with the sodium coolant

Generated particles on the steel surface are transferred instantaneously to the main flow of sodium coolant. Then, the concentration of particles in the sodium coolant is postulated to diffuse uniformly throughout a grid in an instant.

- (C) Particle deposition onto steel surface from sodium coolant

In this model, particles transported by turbulence are thought to be deposited onto the steel surface when the particles penetrate the layer of stopping distance. The concept was proposed by S. K. Friedlander¹¹⁾, S. K. Beal¹²⁾ and J. Escobedo¹³⁾ introduced this concept of stopping distance in regard to the behavior of particles in liquids. We applied this concept to the deposition particles onto the steel surface in sodium liquids.

Concentrations of nuclides in CP particles are determined by the results of PSYCHE calculations for nuclide concentration in structural materials. This study does not take into account detachment of particles that have been deposited. This is because of the extreme difficulty of factoring in the decay of radioactive nuclides among randomly detached particles.

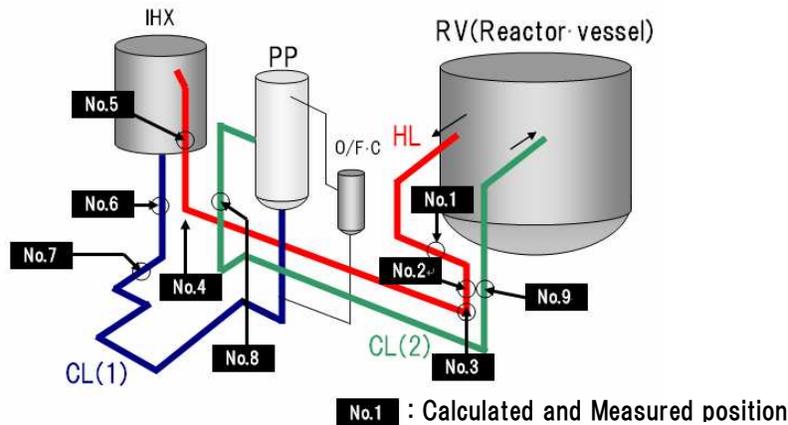


Figure4 The calculations systems for primary cooling system of JOYO.

Consideration of sodium flow

In this study, we introduced the CP transfer behavior analysis code in consideration of the sodium coolant flow. This program system calculated the coolant temperature and flow rate using the NETFLOW++ (Numerical Experimental Tool for Liquid-metal or Water flow system, and Plant Ultimate System), and calculated the CP transfer and solution or deposition with the PSYCHE code using those values. The input data of the coolant temperature and flow rate were calculated automatically using the NETFLOW++ code, and used for PSYCHE code calculations.

As for the NETFLOW++ code developed by H. Mochizuki, it has been confirmed that highly precise calculation results are provided for the heat flow motion calculations for FBR. Thermal-hydraulics and neutronics of the coupled system of the core, heat transport systems and the turbine system can be calculated by the NETFLOW++ code. In this study, a steady state of the JOYO (Mk-II) was calculated using a full model of the primary cooling systems; pumps, throttling of valves, vanes. Models relating the heat transport systems from the primary to the secondary systems were already explained by H. Mochizuki¹⁴.

Analysis system

The nominal power of JOYO (Mk-II) is 100 MWt. JOYO has a primary cooling system consisting of two main cooling system and one overflow / purification system^{4,15}. In the main cooling system, sodium coolant heated in the core flows into the hot-leg pipe (HL), intermediate heat exchanger (IHX 1-5), primary pump (PP) that has overflow column (O/F-C), and the cold-leg pipes extending from IHX to PP (CL(1)) and back to the core inlet (CL(2)).

Calculation regions for the JOYO system consist of 38 core regions and 57 cooling system regions. The system data were made based on a public report⁴. We distinguished clearly

between straight piping and elbow piping in order to compare the calculated values with the measured values^{16,17}. Figure 4 shows the measurement positions and the calculation positions. This paper presents and discusses the results for points at which there are measured values. In the calculation using the Particle model, calculations are carried out with the core designated as the particle generation region and the core and primary cooling system as the particle deposition regions. Figure 5 shows the input Power ratio - Running days. Input data for the reactor operation is formulated in accordance with the actual operation history^{4,15}. Consideration was also given to the radioactive decay of the CP species. Furthermore, timestep interval for the calculations of CP deposition for the precipitation and particle deposition Δt is 0.01 day.

The calculations covered the 7th and 8th periodical inspections of JOYO. This is because in these inspections, detailed measured values for the cooling system were reported^{16,17}.

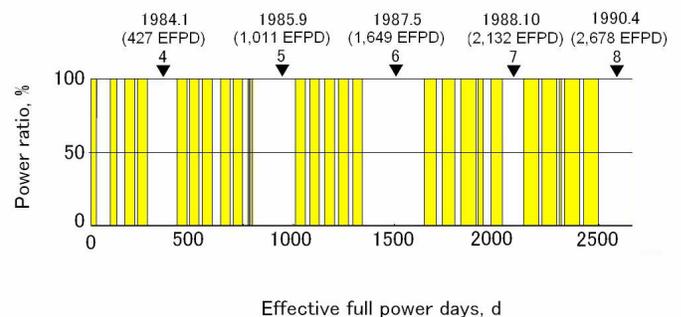


Figure5 Operation history of JOYO (Mk-II).

Model parameters

In PSYCHE calculations, diffusion, fitting factor, chemical partition parameter are necessary parameters. Data used in this study was obtained primarily from the AMTL

(Activated Material Test Loop) experiments at PNC and elsewhere^{18, 19}. Those model parameters were evaluated by fitting the model to experimentally determined solution rates from activated cladding, and the distribution of radioactive precipitation.

Calculations Case

In this paper, we performed calculations for the three following cases. Case A: using the conventional PSYCHE code (Solution-Precipitation model with fitting factor). Case B: using the conventional PSYCHE code (Solution-Precipitation model without fitting factor). Case C: using the improved PSYCHE code (Solution-Precipitation model without fitting factor + Particle model, combined with the NETFLOW++ code).

Fitting factor

In the calculation using conventional PSYCHE code, U_d was used to express the precipitation rate of the boundary. The precipitation rate U_d is expressed in the form of an Arrhenius equation, as in equation (3)^{4, 20}.

$$U_d = f \cdot A \exp(-B/RT) \quad \dots(3)$$

Here, f is fitting factor for precipitation rate of CP [-], A is frequency factor [cm/s], B is activation energy [K/mol], R is gas constant [KJ/deg. mol], T is absolute temperature [K]. A , B , R and T are constants. In conventional PSYCHE calculations, fitting factor are necessary parameters. The fitting factor was obtained primarily from the experiments at JOYO⁴. Table 1 shows the fitting factor for precipitation rate of CP.

This parameter accords with the previous experience that CP is easily distributed, mainly over the reactor core region and the cooling system downstream regions. The fact that the value of Mn exceeds that of Co may be explained by the greater quantity of Mn deposit in comparison to that of Co .

Table 1 The fitting factor for precipitation rate of CP.

Region	⁵⁴ Mn	⁶⁰ Co
Core (deposition region)	6.01	3.24
HL, IHX1	-	-
IHX2	1.67	0.90
IHX3-5, CL1	3.34	1.80
CL2, PUMP	6.01	3.24

RESULT AND DISCUSSION

The C/E (Calculated / Experimentally observed^{16, 17}) values in the primary cooling region in JOYO, using the improved PSYCHE code (Solution-Precipitation model without fitting factor + Particle model + considered of the sodium coolant flow) and conventional PSYCHE code (Solution-Precipitation model with fitting factor, or Solution-Precipitation model without fitting factor), are shown in Table 2. The C/E values were 0.3 to 3 for the ⁵⁴Mn or ⁶⁰Co deposits using improved PSYCHE code. When it is thought that the fitting factor is not used, this agreement between observation and calculation is satisfactory. Table 2 shows, C/E values were in the 0.3-2.5 range when the conventional PSYCHE code with fitting factor was used. An improved PSYCHE code seems to have worse calculation precision than the conventional PSYCHE code. However, the purpose of this study is the calculation of CP transfer behavior in the primary cooling system of FBR plant without using the fitting factor. For exact radiation exposure evaluation, we insist that a calculation code without fitting factor should be developed.

C/E values were improved in Case C in comparison with Case B. These results indicate that calculation precision was raised by adopting more accurate sodium flow and the Particle model. This could be obtained by using accurate sodium flow results from the NETFLOW++ code. It was suggested that the transduction of the particle model for the code was effective in downstream regions with high flow rate, such as CL(2).

Compared to the results calculated using the conventional PSYCHE code with the fitting factor, the amount of CP deposition increased in the HL region when estimated using the conventional PSYCHE code without the fitting factor was increased (see Case A vs. Case B). This is thought to be caused by the definition of the fitting factor. When a correction factor of 1 is assumed (i.e. Case B), the amount of deposition decreases in the coolant system region (from CL(1) to CL(2)) for which the fitting factor had been applied, resulting in a higher concentration of CP nuclides dissolved in sodium coolant. Furthermore, the fitting factor is not applied to the HL region. It is thought that the quantity of CP deposition in the HL region is influenced by an increased CP nuclide density in the sodium coolant.

The C/E values at position No. 2 are higher in all three cases for ⁵⁴Mn. We judged that the measured values at position No. 2 are considerably lower than the value predicted. In past reports^{16, 17}, the measurement point was excluded from the evaluation subject. It is possible that at this measurement position, the problem lies in the measuring method.

Table 3 shows the percentage of total CP deposition accounted for by CP deposition in particle form. From the results of Table 2, it may be judged that the contribution of the particles is not large. In that case, it will be judged that the values of Table 2 are large. Table 3 shows the CP deposition

Table 2 The C/E (Calculated/Experimentally observed) values in the primary cooling region in JOYO, using the improved PSYCHE (Solution-Precipitation Model without fitting factor + Particle Model + considered of the sodium coolant flow) and conventional PSYCHE (Solution-Precipitation Model with fitting factor, or Solution-Precipitation Model without fitting factor).

		Conventional PSYCHE (With fitting factor)				Conventional PSYCHE (Without fitting factor)				Improved PSYCHE (Without fitting factor)			
		⁵⁴ Mn		⁶⁰ Co		⁵⁴ Mn		⁶⁰ Co		⁵⁴ Mn		⁶⁰ Co	
		6th	7th	6th	7th	6th	7th	6th	7th	6th	7th	6th	7th
HL	No.1	0.93	1.36	0.50	0.31	2.33	3.48	0.66	0.42	1.82	2.64	0.69	0.43
	No.2	2.17	2.55	0.63	0.32	5.41	6.49	0.84	0.44	4.20	5.01	0.86	0.44
	No.3	0.75	1.02	0.65	0.30	1.87	2.60	0.88	0.41	1.48	2.04	0.93	0.44
	No.4	0.90	1.05	0.32	0.24	2.25	2.69	0.44	0.33	1.78	2.11	0.45	0.34
	No.5	1.01	1.04	1.10	0.92	2.51	3.55	1.50	1.27	1.93	2.76	1.47	1.21
CL(1)	No.6	2.83	2.73	0.74	0.84	1.47	1.42	0.63	0.72	1.84	1.14	0.71	0.79
	No.7	2.48	2.36	1.14	2.44	1.29	1.23	0.97	2.09	1.04	1.00	1.14	2.36
CL(2)	No.8	2.47	2.77	0.90	1.71	0.73	0.82	0.55	1.06	0.59	0.57	0.66	1.23
	No.9	1.32	2.13	0.86	0.62	0.39	0.63	0.54	0.39	0.32	0.52	0.64	0.45

evaluated as precipitation until now, divided into precipitation and particle adhesion. Table 3 divided CP deposition evaluated as separation into separation and particle adhesion until now and expressed it. This calculation results indicated that the ⁶⁰Co is included in a relatively high ratio in particle formed CP nuclide. Cobalt has less solubility in sodium than Fe and Mn ²¹⁾. Co is strongly corrosion resistant, so there is little corrosion ²²⁾. Meanwhile, there is evidence that highly-concentrated ⁶⁰Co is included in particles on the surface of steel in sodium. The examination of the sodium loop test also revealed the following: Co forms deposited particles mainly composed of an Fe-Co alloy at the steel material's surface ²³⁾. N. Yokoya ²⁴⁾ insists that high-density Co is included in this sodium-chromite particle. The composition of the particles deposited in a low-temperature region of the experimental specimen was 61% Fe, 36% Co, and 3% Cr. This calculation results of this region show good agreement with those previously finding results.

We pay attention to contribution of the particle deposition with the driving force. The calculation results show the few years after the start of operation, the proportion of particle contribution is not high. However, as operation time increases, the CP particles accumulate on the structural material surfaces. In fact, it has been shown that the density of particles deposited on the cladding surface in JOYO, which is consistent with the calculation results ⁵⁾. For the CP particle deposition, in the latter phase of operation, calculation results are expected to trend higher than actual measured values. This is because a model for particle detachment has not been incorporated. The

adoption of a particle detachment model is required in order to make predictions based on physical background.

Table 3 Estimated percentage of the particles among CP deposition amounts.

		Region	6th	7th
⁵⁴ Mn	HL	No.1	2.25%	3.02%
		No.2	1.51%	3.01%
		No.3	3.79%	5.08%
		No.4	3.79%	5.08%
		No.5	0.78%	1.04%
	CL(1)	No.6	1.07%	3.42%
		No.7	5.08%	6.42%
	CL(2)	No.8	5.62%	7.10%
		No.9	5.17%	6.52%

⁶⁰ Co	HL	No.1	4.59%	6.17%
		No.2	3.11%	4.18%
		No.3	7.81%	10.5%
		No.4	7.94%	10.6%
		No.5	1.64%	2.21%
	CL(1)	No.6	5.01%	6.42%
		No.7	9.46%	12.11%
	CL(2)	No.8	7.98%	10.1%
		No.9	7.44%	9.43%

High particle deposition density in the down-stream regions (such as CL(1) and CL(2)) compared to the upper-stream region (such as HL) was also shown. This is because the flow rate of coolant in the down-stream regions is high compared to that of the up-stream regions (see Figure 6). As the flow rate was higher, the inertial force becomes stronger and the number of particles passing through increases, resulting in an increase in the number of particles deposited. Actually, the deposition of the CP particles is mainly confirmed in the JOYO circuits in the down-stream regions²⁵. Then there is a qualitative agreement between the experimental observations and the calculations. The estimation of CP particle deposition could also be used for the definition of decontamination techniques.

With the current analysis code, it should be noted that the analysis system is expressed in an extremely simple model. All the account system is modeled as geometry of the straight pipe arrangement. Then, the calculation precision may be improved by considering the particle deposition in the elbow part. The fluid dynamics acting in pipe elbows are different from those in straight piping. Notably, centrifugal force and inertial force differ²⁶. In JOYO, the dose rate in the elbow-piping region increased compared to the straight piping region. The Particle model is of particular importance in performing separate evaluations of the elbow-piping and other areas.

When a decontamination technique is chosen, a code which can evaluate a particle coating build-up for the quantity of CP total deposit may be effective. In the case of decontamination of the cooling system components and piping, physical decontamination and chemical decontamination are used. Attachments in particle form can be eliminated physically. Deposits, on the other hand, need chemical cleaning. It is thought that this code is effective for selection of this decontamination.

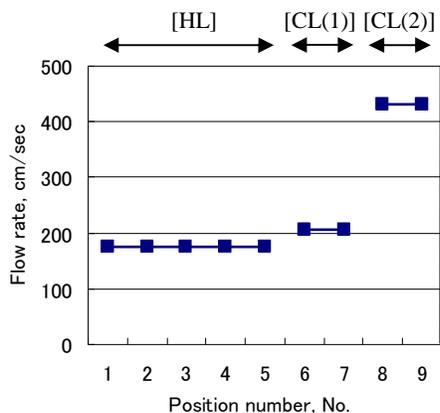


Figure 6 The flow rate of the JOYO primary cooling systems.

CONCLUSION

In this study, in addition to existing Solution-Precipitation model in PSYCHE code, a transfer-model of CP species in particle form was applied to calculations of CP behavior in the primary cooling system of JOYO, and the following findings are obtained:

- The calculation results using improved PSYCHE code are consistent with the measured results for actual components in JOYO. The C/E (Calculated/Experimentally observed) values were improved by introduction of the Particle model and considered of the sodium coolant flow.
- The C/E values were in the 0.3-3.0 range when the improved PSYCHE code without fitting factor for precipitation rate of CP was used.
- It was supposed that a particle was easy to deposit in cooling system in the down stream region of primary coolant systems in comparison with an upper-stream region. It is thought that this computed method is also made use of for decontamination technique.

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