



S. KATO, S. YOSHIMUTA, T. HASUMI, K. SATO  
Nuclear Fuel Industries, Ltd,  
Tokai-mura

K. SAWA, S. SUZUKI, H. MOGI, S. SHIOZAWA, T. TANAKA  
Japan Atomic Energy Research Institute,  
Oarai-machi

Ibaraki-ken, Japan

## Abstract

This paper summarizes the fabrication of the first loading fuel for HTTR, High Temperature engineering Test Reactor constructed by JAERI, Japan Atomic Energy Research Institute. The fuel fabrication started at the HTR fuel facility of NFI, Nuclear Fuel Industries, Ltd., June 1995. 4,770 fuel rods were fabricated through the fuel kernel, coated fuel particle and fuel compaction process, then 150 fuel elements were assembled in the reactor building December 1997.

Fabrication technology for the fuel was established through a lot of R&D activities and fabrication experience of irradiation examination samples spread over about 30 years. Most of all, very high quality and production efficiency of fuel were achieved by the development of the fuel kernel process using the vibration dropping technology, the continuous 4-layer coating process and the automatic compaction process. As for the inspection technology, the development of the automatic measurement equipment for coated layer thickness of a coated fuel particle and uranium content of a fuel compact contributed to the higher reliability and rationalization of the inspection process. The data processing system for the fabrication and quality control, which was originally developed by NFI, made possible not only quick feedback of statistical quality data to the fabrication processes, but also automatic document preparation, such as inspection certificates and accountability control reports.

The quality of the first loading fuel fully satisfied the design specifications for the fuel. In particular, average bare uranium fraction and SiC defective fraction of fuel compacts were  $2 \times 10^{-6}$  and  $8 \times 10^{-5}$  respectively. According to the preceding irradiation examinations being performed at JMTR, Japan Materials Testing Reactor of JAERI, the specimen sampled from the first loading fuel shows good irradiation performance.

## 1. Introduction

Table 1 and Figure 1 show the primary specification and structure of the first loading fuel for the HTTR. The fuel assembly represents a hexahedral prism of 360mm across flats and 580mm in length, and consists of a graphite block, fuel rods to be inserted and retained in a graphite block and so on. 31 or 33 fuel rods are inserted in a fuel assembly. The fuel rod accommodates 14 fuel compacts in a graphite sleeve of 34mm in outer diameter. The fuel compact has a shape of annular cylinder which consists of graphite matrix and Tri-isotropic (TRISO)-coated fuel particles of 0.92 mm in diameter, dispersed within the matrix body. A coated fuel particle consists of a  $\text{UO}_2$  kernel, 600  $\mu\text{m}$  in diameter, and 4 layers of pyrolytic carbon(PyC) and silicon carbide(SiC) [1].

## 2. Fabrication Schedule

As is shown in Figure 2, the fuel fabrication started June 1995. 4,770 fuel rods with 12 kinds of enrichment in total, which correspond to 66780 fuel compacts, were fabricated

TABLE 1 MAJOR SPECIFICATION OF HTTR FUEL

Item	Diameter or Thickness ( $\mu$ m)	Density (g/cm <sup>3</sup> )
UO <sub>2</sub> kernel	600	10.63
Low density PyC (1st)	60	1.10
High density PyC (2nd)	30	1.85
SiC (3rd)	25	3.20
High density PyC (4th)	45	1.85
Coated fuel particle	920	(4.45)

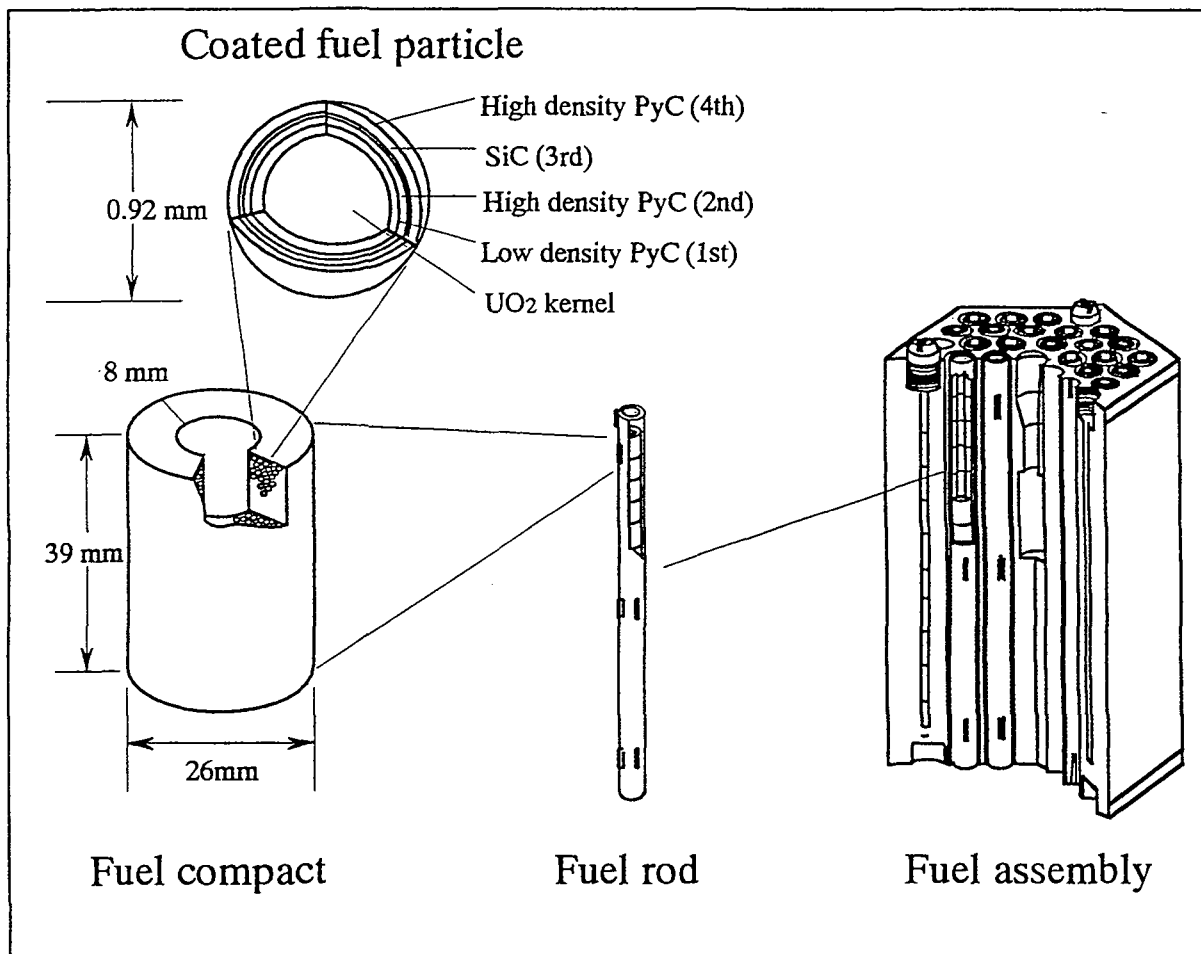


FIG. 1 STRUCTURE OF HTTR FUEL ASSEMBLY

through the fuel kernel, coated fuel particle and fuel compact process. The fuel rods were transported to the reactor building of the HTTR for three times. Then 150 fuel elements were assembled there December 1997. All fuel assemblies, sorting by a column of 5 fuel assemblies, were stored in the new storage cells under helium atmosphere. At present, fuel loading is under way.

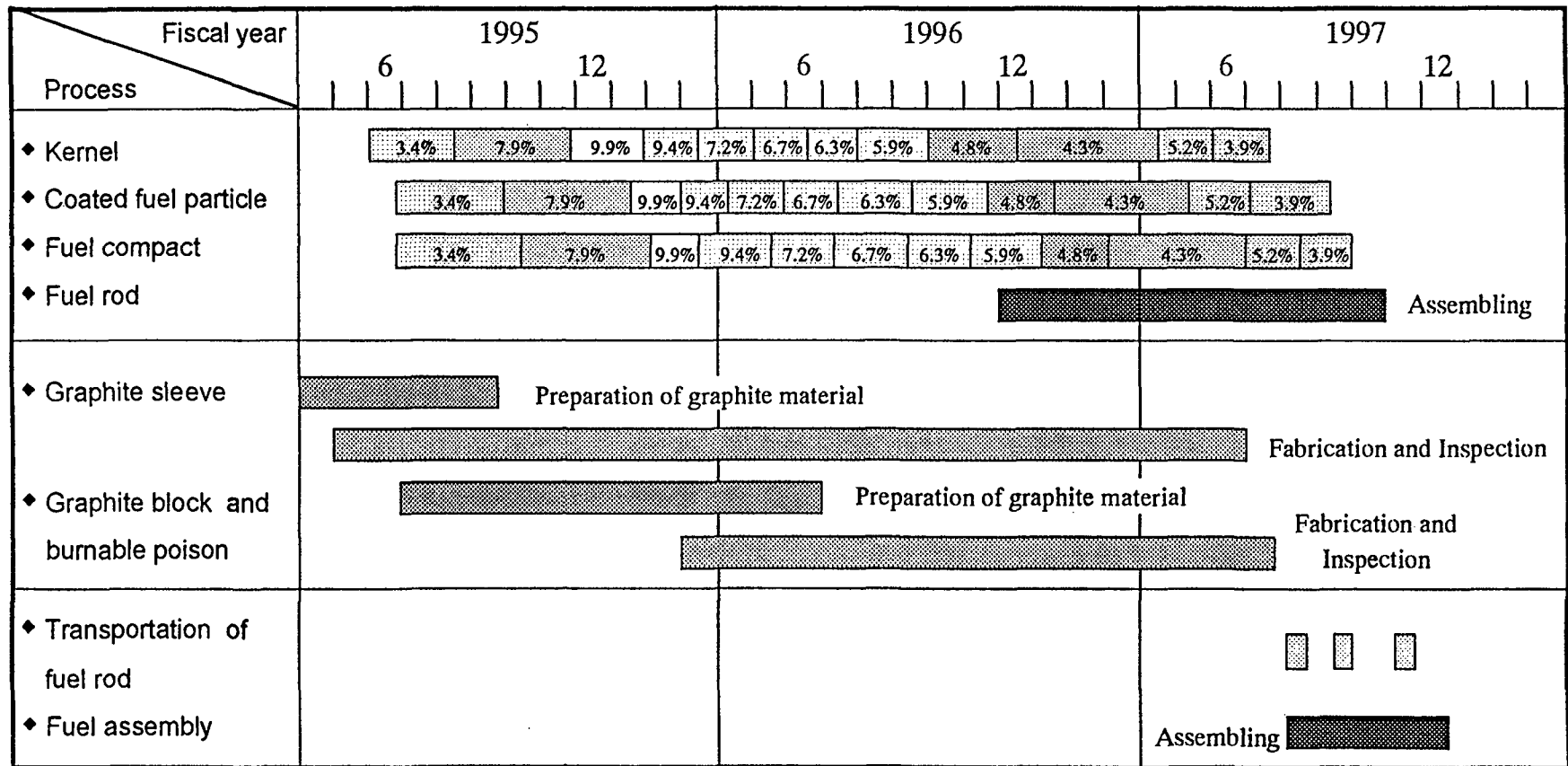


FIG. 2 SCHEDULE OF HTRR FIRST LOADING FUEL FABRICATION

### 3. Fabrication Technology and the Quality

In late 1980s, NFI set up semi-mass-production equipment with licensed capacity of 200 kgU/year and fabricated 260 kgU of fuels for Very High Temperature Reactor Critical Assembly in JAERI. After that, NFI fabricated the fuels for the irradiation tests in Oarai Gas Loop-1 of JMTR etc., continuing not only to study the fabrication technology to achieve better sphericity of coated fuel particle and less defective fraction of coated layer, but also develop the rational fabrication technology and equipment. In 1992, NFI launched the HTR fuel plant with the licensed capacity of 400 kgU/year, which incorporated all the results obtained until then. Finally, the fabrication of 900 kgU of the HTTR first loading fuel successfully completed last December. Fabrication technology and the quality of the HTTR first loading fuel are described below.

#### 3.1 Fuel Kernel

Fuel kernels are fabricated by, so called, "Gel Precipitation Process". The process is shown in Figure 3. Metal solution is prepared as the mixture of the starting material of uranyl nitrate solution and additives to control the viscosity of the solution. Droplets of the metal solution are generated at the vibrating nozzles and fall into ammonia water to be aged to ammonium diuranate(ADU) particles. The reaction products of ammonium nitrate etc. are washed off, then the particles are dried and calcinated to  $UO_3$  particles at 500 °C in air. The  $UO_3$  particles are reduced and sintered to  $UO_2$  particles with about 97 % T.D. at 1600°C under hydrogen atmosphere.

Mainly, coated layers of the coated fuel particles have a function to enclose fission products generated in fuel kernel. The mechanical strength of the coated layers depends on their thickness and sphericity. These characteristics strongly depend on the diameter and sphericity of the kernel. Therefore it is essential to establish the fabrication technology to obtain the kernels with more uniform diameter and excellent sphericity. The vibrating nozzles from which droplets are emitted with high speed were developed for such kernels fabrication. The vibrating nozzles can emit droplets with uniform diameter continuously since the diameter of a droplet is determined by the combination of the flow rate of metal solution and the frequency of the nozzles as indicated in the equation (1). The diameter of a droplet is controlled to have the same uranium content as that of a fuel kernel.

$$Q = \frac{\pi \cdot D^3}{6} \cdot f \quad (1)$$

where Q : flow rate of metal solution  
D : diameter of droplet  
f : frequency of vibrating nozzle

Most of the degradation of kernel sphericity is caused by the deformation at the stage of droplet formation and wet-ADU particle. At the stage of droplet formation, a process was applied to prevent the deformation of droplets when landing on the ammonia water. In the process, droplets are solidified while falling in ammonia gas blown against the droplets. At the stage of wet-ADU particle, a process was applied in which aging, washing and drying are

carried out in the same conical dryer. It is possible to mitigate the impact against the particles by retaining very soft wet-ADU particles in the same conical dryer during the operations, and came to prevent the deformation.

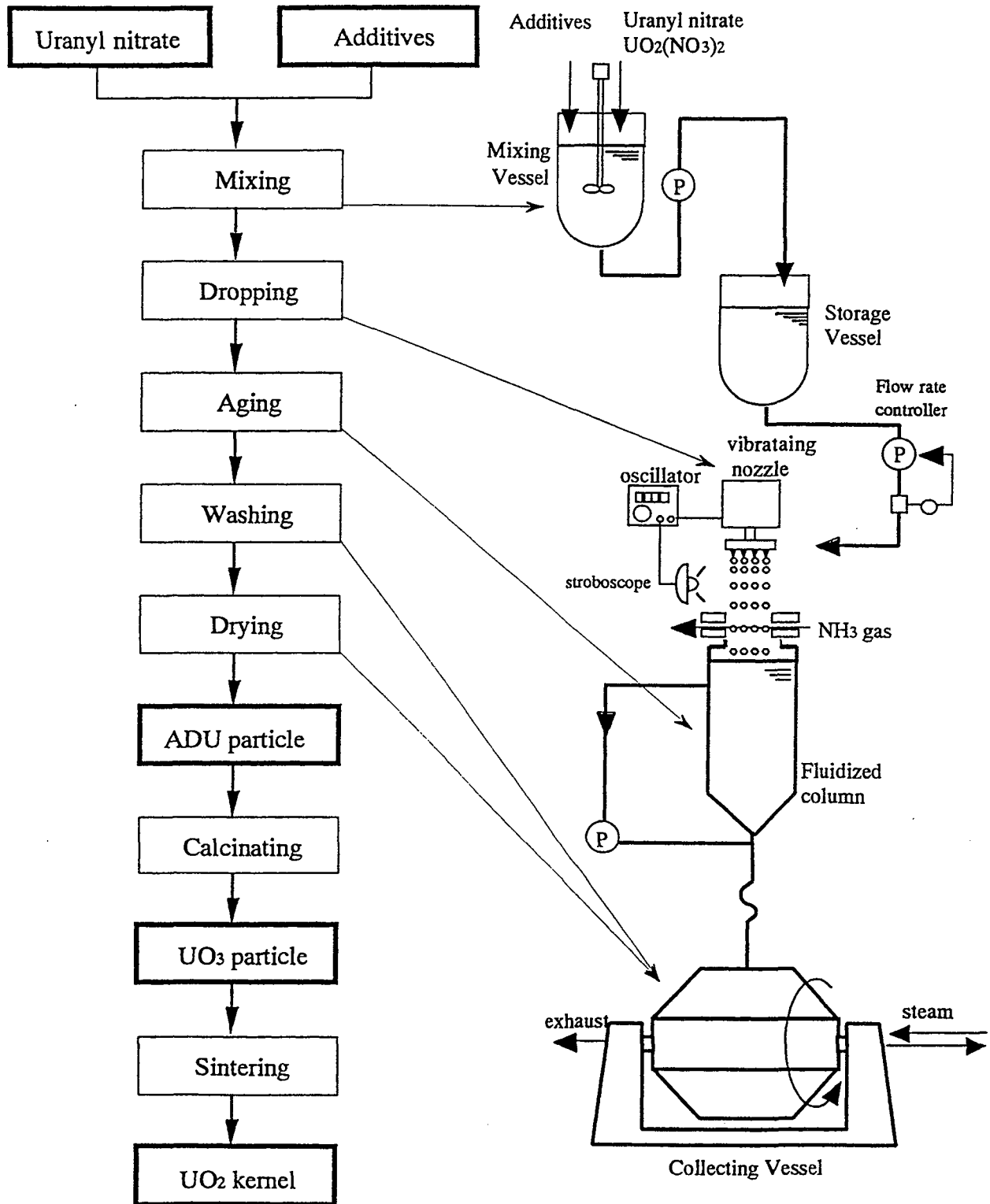


FIG. 3 FABRICATION PROCESS OF  $UO_2$  KERNELS

Figure 4 shows the inspection result of kernel diameter for enrichment lots. Almost all standard deviations were less than  $10 \mu\text{m}$  and uniform diameter of kernels were obtained. Figure 5 shows the inspection result of sphericity of kernel for enrichment lots. The average of each lot was about 1.05 which indicated the sphericity was very much excellent.

### 3.2 Coated Fuel Particle

Figure 6 shows the fabrication flow diagram of coated fuel particles. A coated fuel particle consists of a kernel and four coating layers formed around the kernel by vapor-deposition technology using a fluidized bed type of coater. Mixing gases of acetylene( $\text{C}_2\text{H}_2$ ) and argon are used for the deposition of porous and low density pyrolytic carbon(PyC) for the first layer ; propylene( $\text{C}_3\text{H}_6$ ) and argon for the deposition of dense pyrolytic carbon for the second and fourth layer; methyl-trichloro-silane(MTS) and hydrogen for the deposition of silicone carbide( $\text{SiC}$ ) for the third layer. After first and second layer coating, test specimen for density measurement etc. are sampled.

At the fabrication stage of coated fuel particles, failure fraction of coating layers other than the thickness and density is most important to ensure the function to retain fission products inside the particles. Causes of coating layer failure might be broadly classified into two groups. One is to be the mechanical impact against the particles caused by the intermediate loading and unloading of the particles for the previous coating process. The other is to be the random strong collisions between particles themselves during coating. With respect to the former one, developing the continuous coating process to eliminate intermediate loading and unloading, the failure fraction was reduced. A sampling method

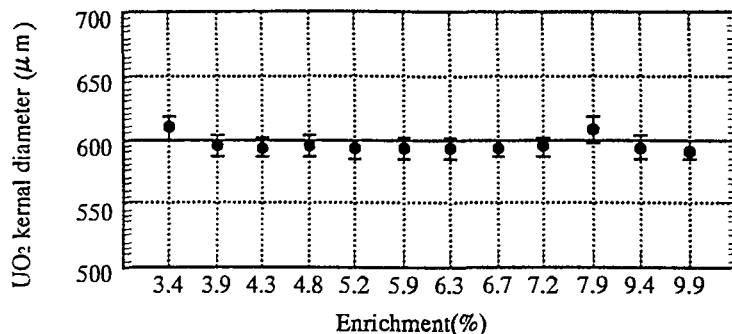


FIG. 4 DIAMETER OF UO<sub>2</sub> KERNEL

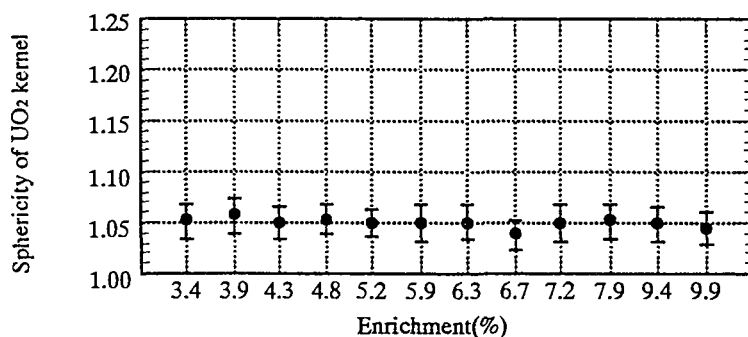


FIG. 5 SPHERICITY OF UO<sub>2</sub> KERNEL

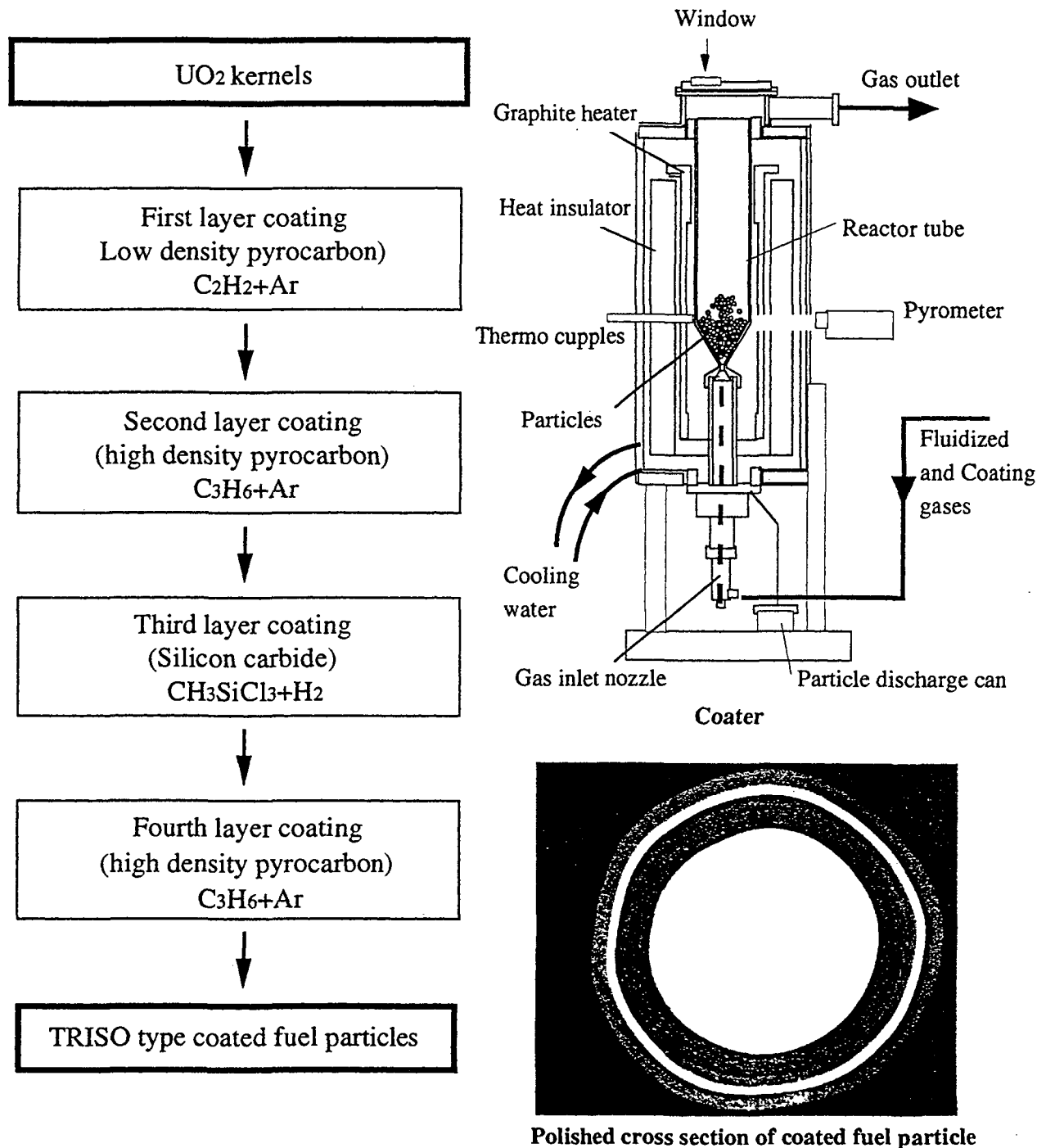


FIG. 6 FABRICATION PROCESS OF COATED FUEL PARTICLE

was also developed to take out a little amount of particles by stopping the gases supply in a moment. With respect to the latter one, gases flow rate, mixing ratio of gases and coating temperatures were optimized empirically to mitigate the random collisions and realize mild flowing condition of particles. As the result, SiC defective fraction came down to the level of  $10^{-6}$ . Figure 7, 8 and 9 show the inspection results of diameter, layer thickness and density of the coated particles respectively, which are more than satisfactory compared with the specifications.

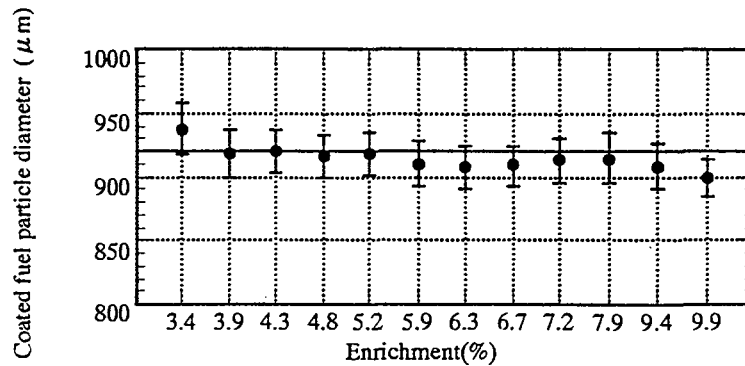


FIG. 7 DIAMETER OF COATED FUEL PARTICLE

In addition to the development of the fabrication technology, NFI developed the automatic inspection system to measure the coating layer thickness to enhance the reliability as well as productivity of the coated fuel particles. The system is composed of a video-microscope and an image processor, which can provide coated layer thickness of 100 particles on X-ray film in a short period of time. The image processor scans the film in X- and Y-direction to collect data of density (darkness) and detect the borders of coating layers of a coated particle.

### 3.3 Fuel Compact

The fabrication flow diagram is shown in Fig. 10. First, natural graphite powder, electro-graphite powder and a binder are mixed, then the mixture makes graphite matrix after fine grinding process. Coated fuel particles are over-coated with the graphite matrix and warm-pressed to make annular cylinder of green compacts. Green compacts are preliminarily heat treated for carbonization at 800°C under nitrogen atmosphere, then sintered at 1800°C under vacuum to make fuel compacts.

Among those processes, the green compact pressing is most complicated, which involves weighing, pre-heating, loading and unloading of over-coated particles and printing a identification number on a green compact and so on. As shown in Fig. 10, the full-automatic pressing system was introduced to rationalize the process. Certain amount of over-coated particles for a green compact are automatically sampled and weighed by the automatic weighing instruments. Weighing accuracy is better than  $\pm 0.05\%$  of the set weight. Weighed over-coated particles are heated in a furnace to soften the graphite matrix preliminary, then poured into dies of the warm-pressing machine which is a rotary hydraulic pressing machine with 8 sets of die and punch. After warm-pressing for about 15 minutes, green compacts are taken out to the printing station where a mark of  $^{235}\text{U}$  enrichment and a serial number are printed on a green compact using a ink jet printer. The ink was selected so that the mark and number should be legible even after the following sintering process at 1800°C under vacuum and produce very little nuclear effect on the reactor operation. The system can make 230 green compacts in about 8 hours.

In order to improve the failure fraction of coating layers at the stage of a fuel compact, it is necessary to disperse coated fuel particles in a green compact as uniformly as possible.



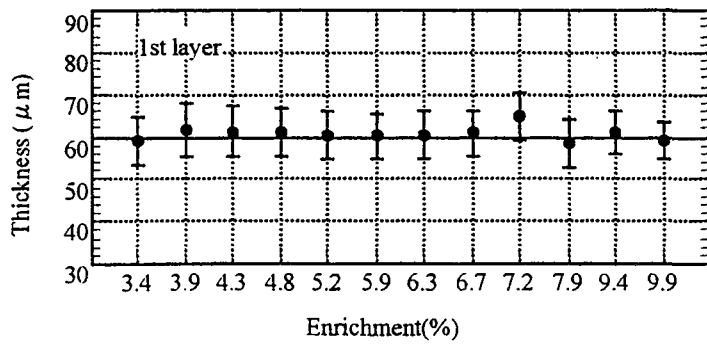
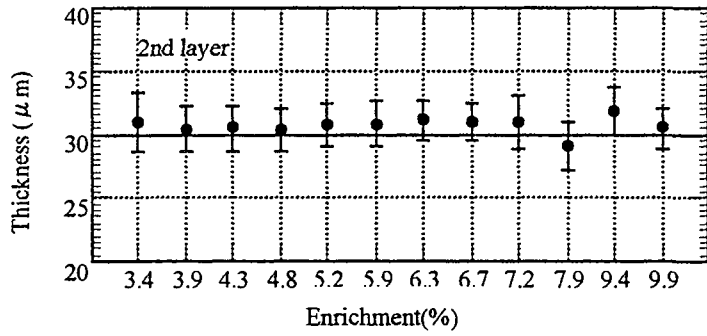
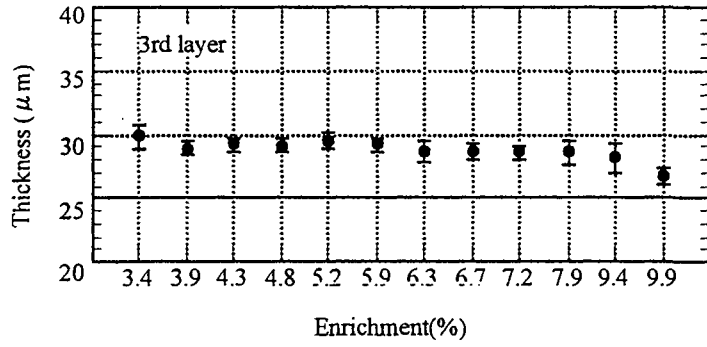
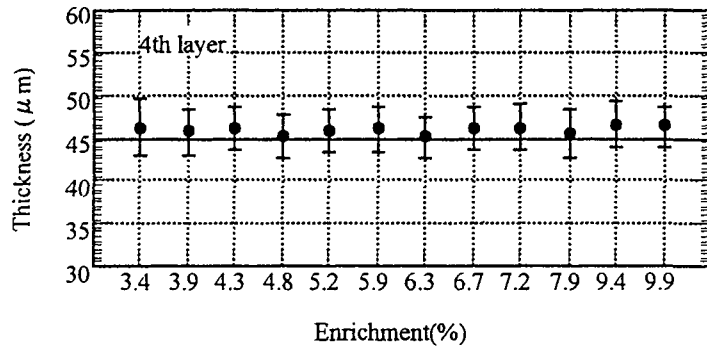


FIG. 8 LAYER THICKNESS OF COATED FUEL PARTICLE

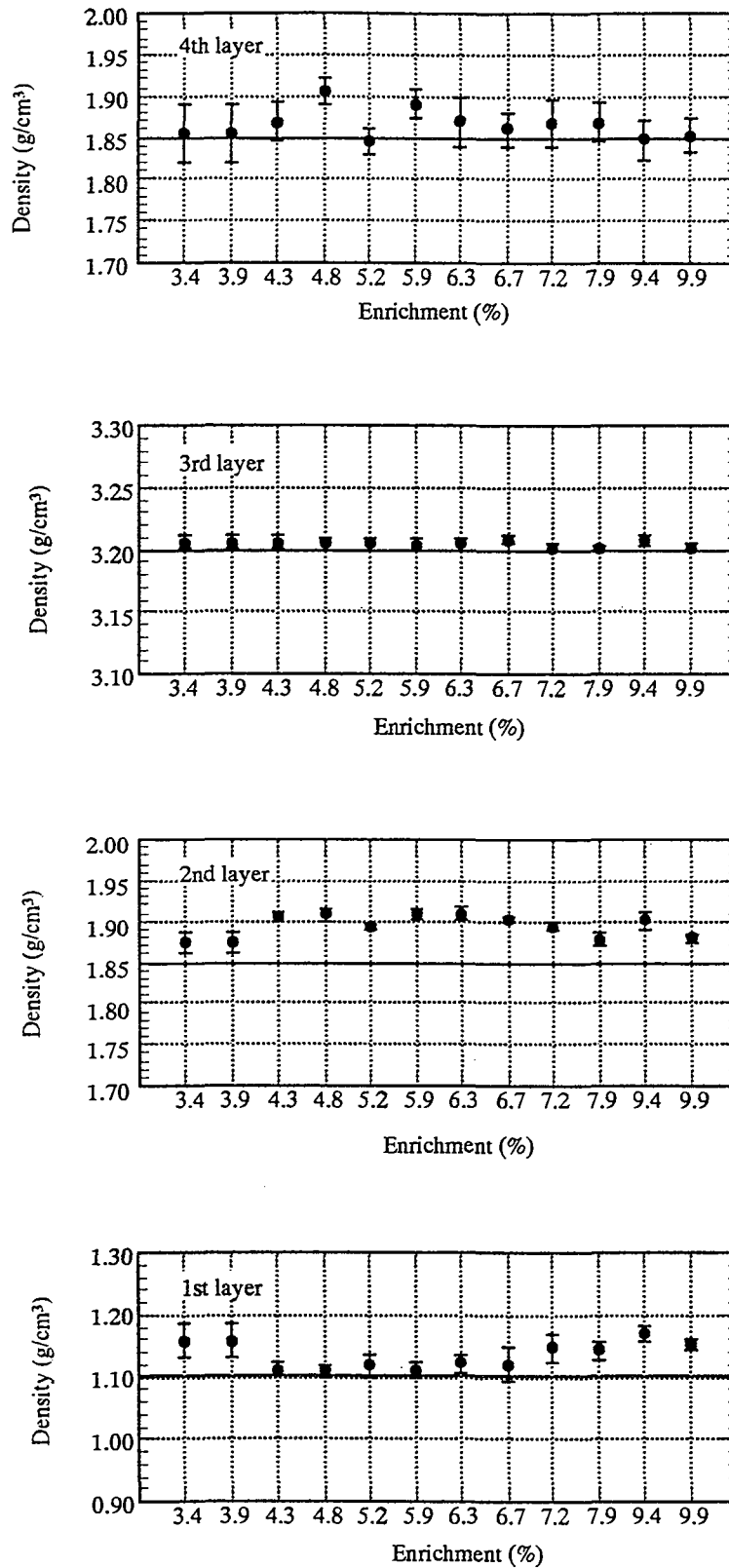


FIG. 9 LAYER DENSITY OF COATED FUEL PARTICLE

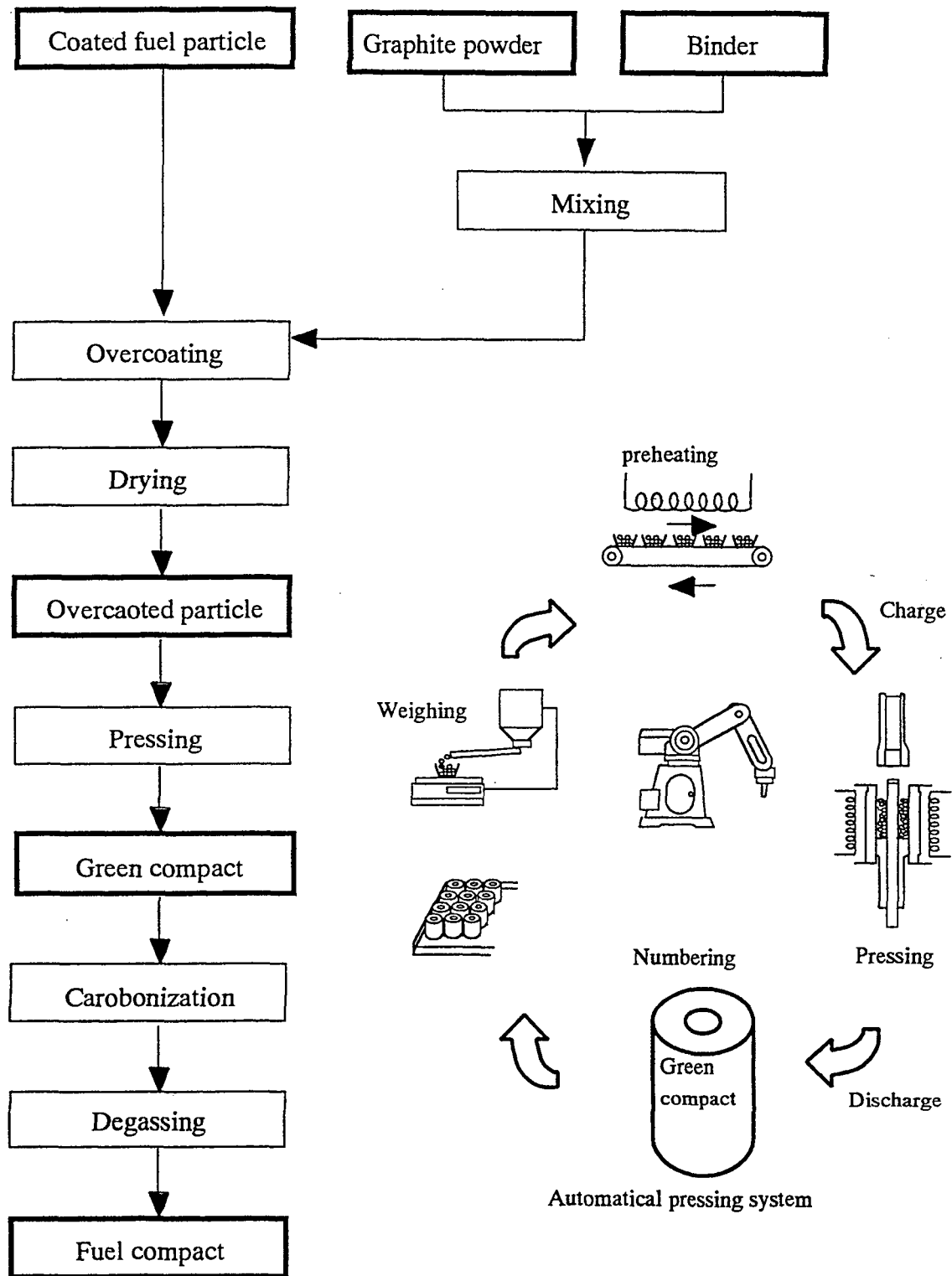


FIG. 10 FABRICATION PROCESS OF FUEL COMPACT

From this point of view, the punching speed and die temperature during the warm-pressing were optimized to fix the best timing of the softening by heating and the plastic flow of matrix graphite by pressing[2]. As the result, it was realized that average bare uranium fraction and SiC defective fraction for all fuel compact lots were  $2 \times 10^{-6}$  and  $8 \times 10^{-5}$  respectively[3,4]. Fig. 11 shows the final result of SiC defective fraction for all lots.

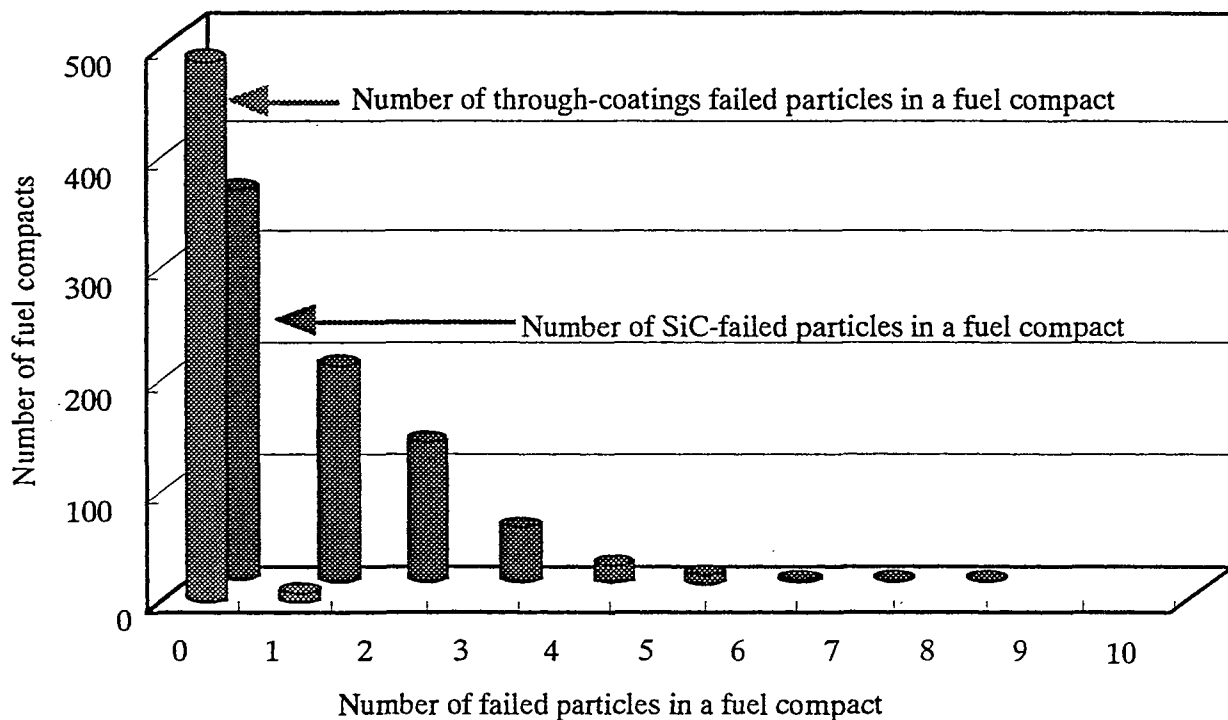


FIG. 11 NUMBER OF FAILED PARTICLES IN A FUEL COMPACT

#### 4. Commercial Production of HTR Fuel

Fuel fabrication cost depends largely on the rate of plant operation and the production. According to the information based on the fabrication experience with pebble type of fuels in former West Germany, they said that the production scale over one million of pebbles per year (about 10 tU/y) would have the effect of mass production.

The HTR fuel plant of NFI were designed and constructed so that the plant could fabricate the first loading fuel and reload fuels just for the HTTR in accordance with the schedule following the construction and operation plan for the HTTR. The fabrication equipment including incidental facilities installed were developed based on the HTR fuel fabrication technology accumulated over 30 years in the past and NFI's plant design technology for the fabrication of light water reactor (LWR) fuels. The equipment could be easily introduced to the above-mentioned scale of the production. For example, the main equipment of two lines of kernel and coated fuel particle fabrication could produce 5 tU/y of coated fuel particles as the case may be. Then, as a module, they could be installed as they are to a 10 tU/y scale of plant. On the other hand, if even a 5 tU/y scale of HTR fuel plant like NFI's plant with such equipment came to continuously produce 5 tU/y of fuels by taking measures listed below, it could be also said that HTR fuel fabrication substantially got into the commercial stage.

- reinforcement of a part of the facility for such as fuel storage, intermediate storage for semi-finished products, and so on
- automatic handling for fuels and semi-finished products

- reductions in cost for raw materials, consumables and tools
- reductions in the consumption of raw materials and consumables
- rationalization of the fuel specification
- reductions of inspection items and frequency
- rationalization of the inspection to be made by the client or competent authority

## 5. Conclusion

After establishing the highly developed fabrication technology for HTR fuel, NFI successfully completed the fabrication of about 900 kgU of the HTTR first loading fuel last December. The quality of the fuel was entirely satisfactory. In particular, average bare uranium fraction and SiC defective fraction for all fuel compact lots were  $2 \times 10^{-6}$  and  $8 \times 10^{-5}$  respectively.

## REFERENCES

- [1] JAERI, "Present Status of Testing and Research for High Temperature Engineering", P21 (1997).
- [2] K. Minato, et al., "Improvements in Quality of As-Manufactured Fuels for High-Temperature Gas-Cooled Reactors", Journal of NUCLEAR SCIENCE and TECHNOLOGY, Vol. 34, No. 3, p. 325-333 (1997).
- [3] K. Sawa, et al., "Evaluation Method and Prediction Result of Fuel Behavior during the High Temperature Engineering Test Reactor Operation", JAERI-Research 98-016 (1998).
- [4] T. Oda, et al., "Fabrication Characteristics of First-loading Fuel Compact of the HTTR", I-43, 1998 Fall Meeting of the Atomic Energy Society of Japan (1998).

## SESSION 3