Spherical Droplets Preparation by Using a Simulated PVA Solution

K-C Jeong, Y-K Kim, S-C Oh, M-S Cho, Y-M Kim, W-K Kim, S-H Na and Y-W Lee *HTGR Fuel Development Division, Korea Atomic Energy Research Institute*

150 Dukjin-dong, Yusong-Gu, Taejon, Korea, 305-353

kcjeong@kaeri.re.k

1. Introduction

Key issues of the next decades will be a growing energy demand, global warming from carbon dioxide gas, and a fossil fuel, oil and coal, depletion, and so on. With these circumstances in mind, intensive studies and discussions on the R&D of a hydrogen gas and a large electricity production by using a HTGR, a typical clean energy, are making steady progress worldwide. The R&D study for a clean hydrogen production with HTGR in Korea started in the 1990's, with a strong desire to study the production of a clean hydrogen energy.

The HTGR fuel element developed in the USA and Japan is a large, hexagonal prism(Prismatic) and Germany and Russia use a spherical fuel element (Pebble). Despite these very different shapes, all the fuel elements have the same basic design. The fuel they contain is in the form of micro-spheres, each the size of the head of a pin, coated with several layers of a ceramic material. These coated particles are embedded in a graphite matrix. The graphite serves simultaneously as a structural and a moderator material.

In general, the Pebble or Prismatic type fuel, which is prepared with a spherical ceramic UO_2 kernel particle, is inserted into the HTGR. The UO_2 kernels obtained from a sol-gel method change to a TRISO (TRISOtropic) shape[1]. The TRISO-shaped fuel particle: UO_2 kernel spheres coated with layers of porous pyrolytic carbon(PyC), inner dense PyC, silicon or zirconium carbide(SiC, ZrC), and an outer dense PyC. Sol-gel technology is a promising way to prepare a spherical UO_2 kernel, because of its merits of a high density of the UO_2 powder and its easily controlled components[2].

The production of spherical fuel kernels can be carried out by wet chemical processes, a sol-gel process, based on a solidification of droplets. Sol-gel process has advantages in a high purity and a low processing temperature. Despite these advantages, there are only a few reports on the preparation of a spherical UO_2 kernel by a sol-gel. In the production of UO_2 kernels by a gel supported precipitation, a uranyl nitrate solution is the starting material. This solution is pre-neutralized to a certain degree with aqueous or gaseous ammonia before the addition of organic additives, PVA(polyvinyl alcohol), and THFA(tetra-hydrofurfuryl alcohol).

To obtain a proper UO_2 kernel, first the spherical droplets should be formed by a vibro-dropping of an initial feed solution. The droplets are subjected to a lateral stream of air and gaseous ammonia as they fall. This results in a surface solidification of the droplets based on the fact that ammonia containing ammonium

di-uranate is formed, which covers the droplet like an elastic skin. The droplets are now so stable that they no longer loss their spherical shape when they contact the surface of the ammonia solution in the gelation column. The reaction of uranyl nitrate with an ammonia solution is brought to an end in the gelation column. Small translucent, amorphous kernels, ADU, are obtained.

In this study, first we carried out a same size spherical droplets preparation with a vibrating nozzle system by using simulated PVA solutions.

2. Experimental

The experimental apparatus for the spherical droplets preparation mainly consists of a simulated PVA solution storage tank, a droplet nozzle, a vibrating system, and a gas pressing line, as shown in Figure 1.



Figure 1. Experimental apparatus.

The simulated PVA solutions are prepared with a reagent, such as Mowiol, Poval, and Aldrich type A and B products. And the simulated solution was made by a dissolution of a reagent above PVA with a pure water to 8~10 wt %. Thermal decomposition characteristics of these PVA reagents are analyzed with TG/DTA, and a spherical droplets shape was observed by digital carmera. Viscosity of the simulated PVA solutions is measured with a spindle type viscometer.

3. Results and Discussion

3.1 Viscosity of simulated PVA solution

The viscosity of the broth solution should be adjusted before a droplet preparation. In general, the final products, a UO_2 sphere particle, sphericity is determined by the droplet's sphericity prepared at this

first step, and here the viscosity role is the most important factor in a sphere droplets preparation.

The viscosity values and characteristics data obtained from the dissolution of the PVAs, such as the Mowiol, Poval, Aldrich A and B type, with pure water are shown in Table 1.

	M.W.	Conc.	Viscosity	Remarks
Mowiol	195000	10 %	2050 c.P.	26 °C
		8 %	661	"
Poval	?	10 %	2010	"
Aldrich A	160000	10 %	3485	"
		8 %	918	"
Aldrich B	130000	10 %	2180	"

Table 1. Viscosity Values for Several Reagent PVAs.

The viscosities of the simulated PVA solutions were measured about 2000 c.P. in the case of Mowiol, Poval, and Aldrich B type, and about 3500 c.P. in the case of Aldrich A type. Viscosity was measured at a room temperature of 26°C. If the temperature is lowered, the measured viscosity values will be increased considerably because it is function of the temperature. To obtain proper value's of the viscosity, these simulated PVA solutions should be used after a dilution with pure water.

3.2 Spherical droplet preparation

The spherical droplets are formed by dropping the simulated PVA solutions from a vibrating nozzle into a gelation column. Mono dispersed spherical droplets form from the effect of a surface tension and viscosity of a feeding solution, and the frequency and amplitude of the vibrating nozzle system. Figure 2 shows a simple schematic flow diagram of the spherical droplets preparation.



Figure 2. Dropping phenomena in nozzle system.

Table 2 and Figure 3 show the operational conditions and spherical droplets photographs obtained from the same size droplets preparation experiments in our vibrating nozzle system. The droplets size in Table 2 was calculated with the flow rate, the used nozzle diameter, and the natural laminar jet length observed to about $5\sim10$ mm, in Figure 3. If the relation between the

flow rate, frequency and the amplitude of the vibrating system is not discordant, small satellite drops are formed as shown in Figure 3.

Table 2. Drop Size Obtained in Dropping Experiments.

	Viscosity	Flow	Freq.	Drop size
		rate	(Hz)	
Poval	72 c.P.	49.2	180	2057 µm
Mowiol	70	49.3	180	2046
Aldrich A	71	50.2	180	2057
Aldrich B	82	57.0	180	2145



Figure 3. Dropping phenomena in our experiments.

The formation of a different size of the droplets is because the flow rate of the simulated PVA solution is so high that the flow phenomena of a steady state are broken. As a result, the harmony between the flow rate of the feed solution and the frequency and the amplitude of the vibrating system are important factors for mono size spherical droplets and the protection of a satellite droplet formation[3].

4. Conclusion

In this study we present a spherical droplets formation by using a simulated PVA solution in a vibrating nozzle system. The most important fact is a harmony of the flow rate of the simulated solution and the frequency and amplitude of the vibrating system.

ACKNOWLEDGEMENT

This study was performed under the Atomic Research Development Program sponsored by the Ministry of Science and Technology (MOST) of Korea.

REFERENCES

[1] J.L.Collins, M.H.Lloyd, and R.L.Fellows, "The basic chemistry involved in the internal gelation method of precipitating uranium as determined by pH measurements", Radiochimca Acta, vol. 42, pp.121~134, 1987.

[2] A.W.Mehner et al., "Spherical fuel elements for advanced HTR manufacture and qualification by irradiation testing", J. Nuclear Materials, vol.171, 9~18, 1990.

[3] J.H. Hilbing and S.D.Heister, "Droplet size control in liquid jet breakup", Phys. Fluids, vol. 8(6), 1574~1581, 1996.