Improvement of the Homogeneity of Atomized Particles Dispersed in High Uranium Density Research Reactor Fuels

Chang-Kyu Kim, Ki-Hwan Kim, Jong-Man Park, Yoon-Sang Lee, Don-Bae Lee
Woong-Hee Sohn* and Soon-Hyung Hong*

Korea Atomic Energy Research Institute

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

*Dept. of Materials Science and Engineering, Korea Advanced Institute of Science and Technology

373-1, Kusong-dong, Yusong-gu, Taejon 305-701, Korea

Presented at the

21st International Meeting on Reduced Enrichment for

Research Reactors (RERTR)

18-23 October 1998

Sao Paulo, Brazil

Improvement of the Homogeneity of Atomized Particles Dispersed in High Uranium Density Research Reactor Fuels

Chang-Kyu Kim, Ki-Hwan Kim, Jong-Man Park, Yoon-Sang Lee, Don-Bae Lee
Woong-Hee Sohn and Soon-Hyung Hong**

Korea Atomic Energy Research Institute

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

**Dept. of Materials Science and Engineering, Korea Advanced Institute of Science and Technology*

373-1, Kusong-dong, Yusong-gu, Taejon 305-701, Korea

Abstract

A study on improving the homogeneous dispersion of atomized spherical particles in fuel meats has been performed in connection with the development of high uranium density fuel. In comparing various mixing methods, the better homogeneity of the mixture could be obtained as in order of Spex mill, V-shape tumbler mixer, and off-axis rotating drum mixer. The Spex mill mixer required some laborious work because of its small capacity per batch. Through optimizing the rotating speed parameter for the V-shape tumbler mixer, almost the same homogeneity as with the Spex mill could be obtained. The homogeneity of the extruded fuel meats appeared to improve through extrusion. All extruded fuel meats with U_3Si powder of 50-volume % had fairly smooth surfaces. The homogeneity of fuel meats by V-shaped tumbler mixer revealed to be fairly good on micrographs.

Introduction

Low enriched uranium (LEU) fuel has been developed to replace high enriched uranium (HEU) fuel in research and test reactors since 1978 [1-2]. Low enriched U_3Si_2-Al dispersion fuel has been converted in many research reactors [3]. However, the application of the fuel is limited to approximately 90% of all research reactors due to the uranium density of 4.8g/cm^3. Several high performance reactors still require a higher uranium density fuel up to 8de9 g-U/cm^3 [4].

Uranium density in dispersion fuel can increase basically by choosing a fuel material with higher uranium density or increasing the fuel powder fraction in fuel meats. In the latter method, the fuel powder should be dispersed homogeneously in fuel meat. In general, the homogeneously mixing of fuel and aluminum powders is difficult due to a greater difference of densities. In case that the fuel particles are segregated locally, the segregated area with fuel powder is easy to induce voids during fabrication as well as has an adverse effect on in-reactor performance. As the homogeneity of mixed powders is improved, the fuel powder loading would increase possibly. At KAERI, the atomization process has been applied to the fabrication of dispersion fuels since about 10 years ago [5]. The comminuted fuel particles have angular shapes, while the atomized fuel particles have spherical shapes with smooth surfaces. The homogeneity of mixed powders might be varied with the particle shape. The results of BWXT showed that the atomized U_3Si_2 spherical particles were more segregated in fuel meat than the comminuted particles [6].

In this study, the homogeneity was evaluated by standard deviation of the measured apparent densities for the mixed powders and fuel meats. The influences of particle shape and density on the homogeneity were investigated. Three kinds of mixing methods, which are the off-axis rotating drum mixing, Spex mill mixing, and V-shape tumbler mixing, were compared by the homogeneity of the fuel powder mixtures with aluminum powder and the fuel meats. An effort was made to choose the mixing method suitable for the atomized spherical fuel powder and to optimize the mixing parameters. Using the optimized mixing parameters, the dispersion fuels with high uranium density were fabricated and examined.

Experimental Procedures

The homogeneity of the powder mixture was evaluated by the standard deviations of composition calculated from the apparent densities measured for a number of samples taken from the powder mixture. The apparent densities of samples were measured using the Hall Flow Meter consisting of a precision volume cup and a funnel. For the fuel meats, the densities were measured for the samples taken by cutting fuel rods. In order to investigate the effect of a different particle shape on the homogeneity, two kinds of powders, which are Cu-Sn alloy powder spherical and irregular Cu powder, were mixed with Al powder using the Spex mill mixer and off-axis rotating drum mixer, and then compared for homogeneity. Using atomized and comminuted U_3Si powders the comparison of homogeneity was also performed. The influence of the density difference of powders on homogeneity of mixture was investigated by mixing spherical Cu-Sn and U_3Si powders in an off-axis rotating drum mixer. A mixing test for the irregular Cu powders with different size distributions and Al powders was carried out using the off-axis rotating drum mixing method. The homogeneities of mixtures were compared for various size distributions of Cu powders.

Atomized U_3Si powders were mixed with Al powders in three kinds of mixing methods, which are off-axis rotating drum mixer, Spex mixer and V-shape tumbler mixer. The angle between the cylinder and the centerline in the off-axis rotating drum mixer is 40°. The volume fraction of the powders charged in mixer is between 25 and 40 %. The operating conditions of mixers are as shown in Table 1.

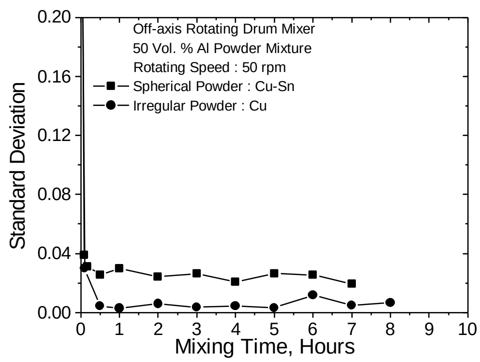
For the selected mixing method suitable for atomized powders, the operating parameters such as rotating speed and time were optimized. Using the optimized mixing conditions, the fuel meats with high fuel powder fraction were prepared and investigated for the homogeneity in comparison with the previously fabricated ones.

Table 1. Operation conditions of three kinds of mixers

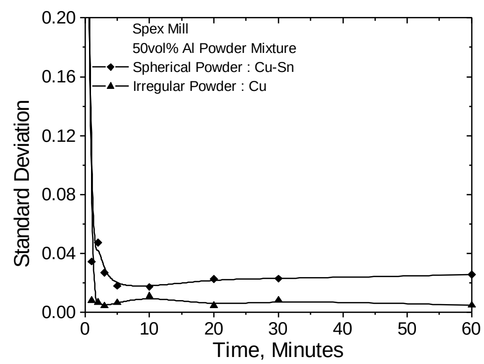
	Off-Axis Rotating Drum Mixer	Spex Mill	V-shape Tumbler Mixer
Rotating Speed (RPM)	50	1,700	25,50,75
Amount of Mixture (cm^3)	50	10	50

Results and Discussion

The spherical Cu-Sn powder showed to have less homogeneous distribution in Al powder than the irregular Cu powder in off-axis drum mixing and Spex mill mixing methods as shown in Figure 1. The apparent densities of Cu-Sn and Cu powders are 5.07 g/cm^3 and 2.92 g/ cm^3. If considering that the true densities of those powders are 8.87 g/cm^3 and 8.9 g/cm^3, respectively, the irregular Cu powder has a greater amount of voids than the spherical powder. It is assumed that the voids allow small aluminum particles to be seated. The irregular shape powder with a rough surface is considered to have more friction to the flow of aluminum powder. The comminuted U_3Si powder with an irregular shape was shown to have a little smaller standard deviation in mixture than the atomized powder with a spherical shape as shown in Figure 2.



(a)



(b)

Figure 1. The variation of the standard deviation of the measured apparent densities is shown with time. (a) off-axis rotating drum mixer and (b) Spex mill for Cu-Sn or Cu and Al mixture

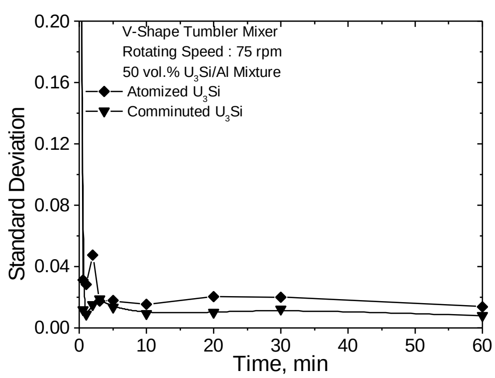


Figure 2. The variation of the standard deviation of the measured apparent densities is shown with time for U_3Si and Al mixture.

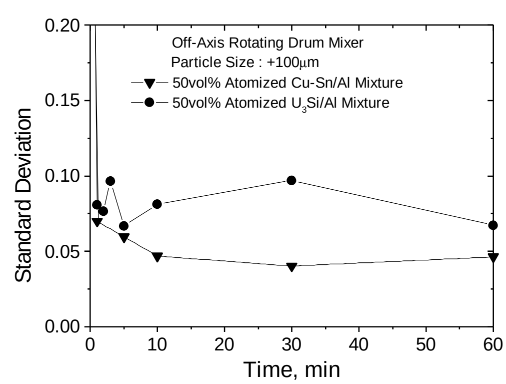
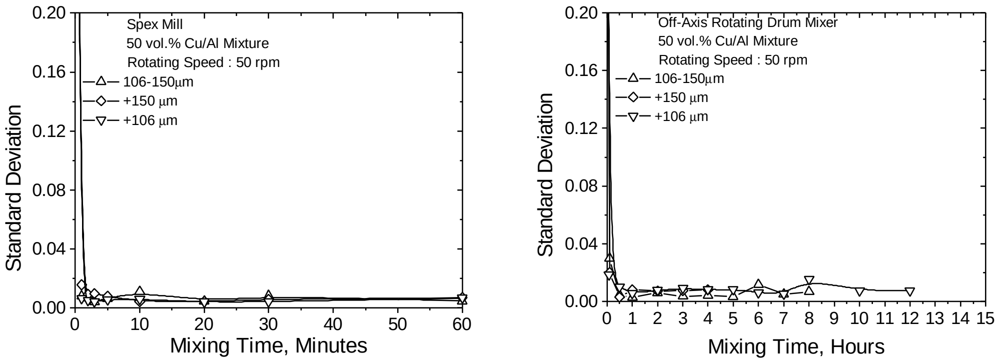


Figure 3. The homogeneities of U_3Si/Al and Cu-Sn/Al mixture with two kinds

of density differences are compared
with time.

Figure 3 shows the results obtained from comparing the homogeneity for two mixtures of spherical U_3Si and Cu-Sn powders with aluminum powder. The densities of U_3Si and Cu-Sn are 15.3 and 8.77 g/cc, respectively, which are much higher than the aluminum density of 2.7 g/cc. The mixture of Cu-Sn and Al powders with smaller density difference appeared to have better homogeneity than the mixture of U_3Si and Al mixture. As the density difference is smaller, the possibility to be segregated is assumed to decrease.

Cu powders with three kinds of particle size distributions are mixed with aluminum powder using off-axis rotating drum mixing and Spex mill mixing. Then the standard deviations of apparent densities with time were obtained as shown in Figure 4. As the range of particle size distribution got broader, the homogeneity of the mixture did not improve remarkably in both mixing methods. If the powder with larger sizes in the mixture has a relatively narrower particle size distribution, the voids among the Cu particles would become a little larger. So, smaller Al particles in the mixture are considered to be more easily seated in the Cu particles in the mixture with the narrower particle distribution. The homogeneity of the mixtures seems not to have changed remarkably with various particles size distributions of the fuel powder.



(a) (b)

Figure 4. The variation of standard deviation of irregular Cu having different size distributions and Al mixtures are shown with time. (a) Spex mill mixer and (b) off-axis rotating drum mixer.

The homogeneity comparison of the mixtures for three kinds of mixing methods, which are Spex mill, V-shape tumbler, and off-axis rotating drum, was performed using U_3Si powder and aluminum powder as shown in Figure 5. The mixtures by Spex mill mixer appeared to have the best homogeneity on the whole and a large fluctuation of the standard deviation did not occur with time. The V-shape tumbler mixer showed to be relatively better than the off-axis drum mixer in the homogeneity of the mixture. The standard deviation of apparent densities decreased up to the minimum value in about 10 minutes and then increased slightly. The standard deviation after 30 minutes did not show any large fluctuation and kept almost the same value. However, an off-axis rotating drum mixer seems to have a little more fluctuation in standard deviation. In order to improve the homogeneity by using the off-axis drum mixer, the U_3Si and Al powders were mixed by using the 40 vials with 1 cm^3 volume in the off-axis drum mixer. After U_3Si and Al powders are mixed for about 30 minutes, the homogeneity using the off-axis rotating vial-in-drum mixer seems to be improved from the results that the standard deviation of the mixtures by using vials in the off-axis rotating drum mixer were changed from 0.032 to 0.023 in comparison with the off-axis rotating drum mixer.

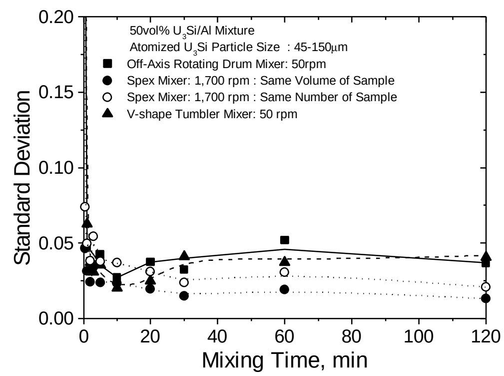


Figure 5. The variation of the standard deviations of the measured apparent densities is shown with time.

The standard deviations for Spex mill mixer were obtained by measuring the same number of samples from one batch mixture as for other mixers. In the case of sampling with the same volume, homogeneity improves due to the relationship between the standard deviation and the sample size as follows. Figure 6 shows the relationship between the standard deviation and the sample size.

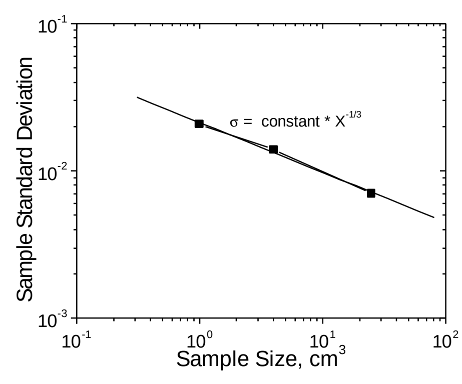
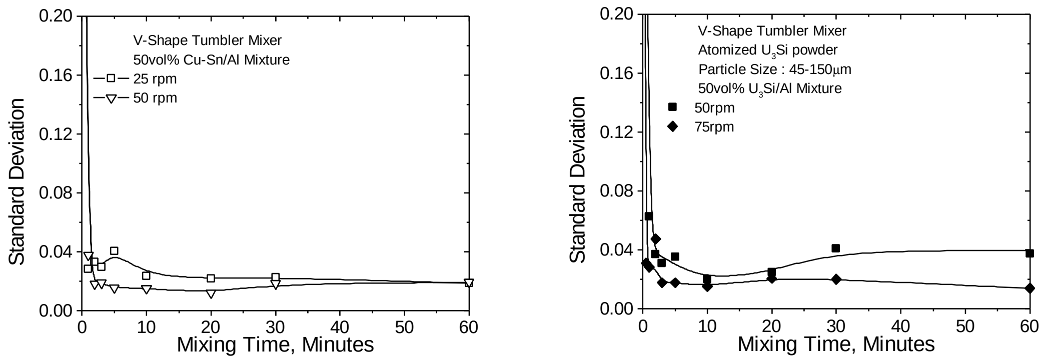


Figure 6. The relationship between sample size and standard deviation.

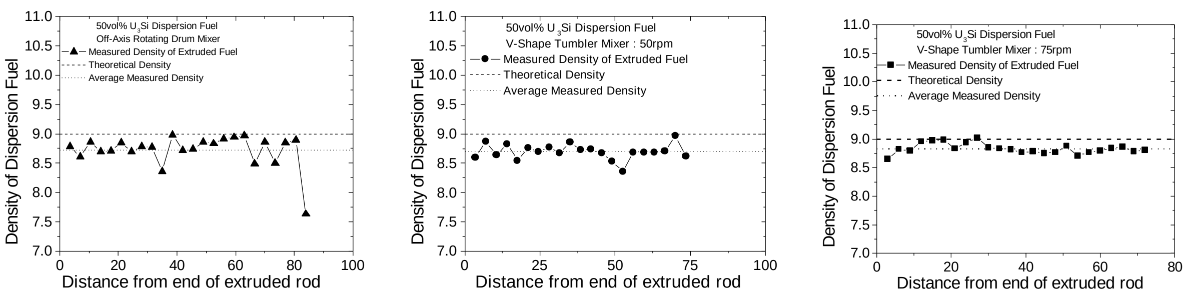
The V-shape tumbler mixer is considered to be suitable for fuel fabrication process in viewpoints of the operation capacity. In order to optimize the mixing parameters, mixing tests were done with variations in the rotating speeds of 25 rpm and 50 rpm using spherical Cu-Sn powder and Al powders. The results showed that the higher rotating speed of 50 rpm had better homogeneity as in Figure 7. In addition, the results that were obtained from mixing tests of U_3Si and Al powders with 50 rpm and 75 rpm revealed that the higher rotating speed of 75 rpm had better homogeneity in the mixture. The values of standard deviations in using V-shape tumbler mixer with 75 rpm rotating speed were almost as low as the values in the Spex mill mixer.



(a) (b)

Figure 7. The variation of the standard deviation is compared for two kinds of rotating speeds time. (a) 25 and 50 rpm for Cu-Sn/Al mixture and (b) 50 and 75 rpm for U_3Si/Al mixture.

Using the selected mixing methods with the optimized parameters, fuel meats were fabricated with 50-volume % of U_3Si powder in the mixture. All extruded fuel meats appeared to have fairly smooth surfaces. In order to examine the macro-homogeneity of atomized U_3Si powder in fuel meats, density measurements of fuel meats were performed and the results were obtained as shown in Figure 8. Densities of fuel meats mixed by V-shape tumbler mixer showed a lower standard deviation than those mixed by the off-axis rotating drum mixer. The distribution of atomized U_3Si fuel particles seems to get more homogeneous through the extrusion process as shown in Table 2.



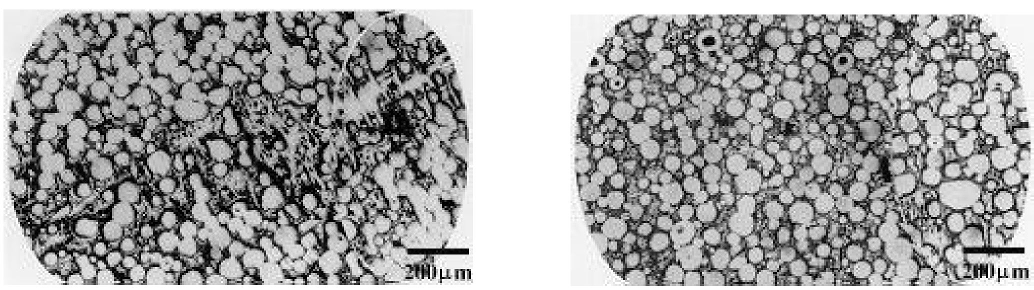
(a) (b) (c)

Figure 8. The variation of the density of samples sliced along the extruded fuel rod.

Table 2. The comparison of homogeneities for the powder mixture and fuel rods of U_3Si and Al dispersion fuel.

Standard Deviation	Off-Axis Rotating Drum Mixer	Off-Axis Rotating Drum Mixer Using Vial	V-shape Tumbler Mixer
Powder Mixture	0.036	0.023	0.018
Dispersion Fuel Meats	0.015	0.010	0.008

Typical micrographs of fuel meat cross-sections were shown as Figure 9. Metallo-graphic examinations of dispersion fuel meats revealed that although U_3Si particles are distributed homogeneously without directionality, aluminum rich regions are frequently found to exist along the longitudinal direction in the fuel rods by off-axis rotating drum mixer as shown in Figure 9 (a). However, the homogeneity of fuel meats by V-shaped tumbler mixer is assumed to be fairly good even though some inhomogeneous regions exist such as in Figure 9(b).



(a) (b)

Figure 9. Microstructures of extruded fuel meats containing 50 volume % atomized U_3Si powder mixed by (a) off-axis rotating drum mixer showing inhomogeneous distribution of U_3Si and (b) V-shape tumbling mixer showing homogeneous distribution of U_3Si particles.

Conclusions

Spherical powders showed a tendency to have less homogeneity in dispersion fuel meats. The mixture with a smaller density difference appeared to have better homogeneity than the one with larger density difference. The homogeneity of the mixture seems not to be affected by the broadness of the fuel particle size distribution. In comparing mixing methods, the better homogeneity of the mixture could be obtained as in order of Spex Mixer, V-shape tumbler mixer, and off-axis rotating drum mixer. But by optimizing the rotating speed parameter, the V-shape tumbler mixer with 75 rpm could have almost the same homogeneity as the Spex mill mixer. The homogeneity of the extruded fuel meats seems have improved through extrusion. All extruded fuel meats with U_3Si powder of 50-volume % appeared to have fairly smooth surfaces. Densities of fuel meats mixed by V-shape tumbler mixer showed a lower standard deviation than those mixed by the off-axis rotating drum mixer. The homogeneity of fuel meats by V-shaped tumbler mixer is assumed to be fairly good on micrographs.

Acknowlegement

This project has been performed under the Nuclear R&D by MOST.

References

1. G. L. Hofman, J. Nucl. Mater., **140**, 256 (1986).
2. J. L. Snelgrove, G.L. Hofman, C.L. Trybus and T.C. Wiencek, Pro. of 19th International Meeting on Reduced Enrichment for Research and Test Reactors, p.46, Seoul, Korea (1996).
3. J.P. Durand and Y. Fanjas, Pro. of 16th International Meeting on Reduced Enrichment for research and Test Reactors, p.71, Oarai, Japan, October 4-7 (1993).
4. A. Travelli, Pro. of 19th International Meeting on Reduced Enrichment for Research and Test Reactors, p. 3, Seoul, Korea, October 6-10 (1996).
5. C.K. Kim, K.H. Kim, C.T. Lee and I.H. Kuk, Proceedings of 14th International Meeting on Reduced Enrichment for Research and Test Reactors, p.109, Jakarta, Indonesia, Nov. 4-7 (1991).
6. L. E. Todd and B.W. Pace, Pro. of 19th International Meeting on Reduced Enrichment for Research and Test Reactors, p. 65, Seoul, Korea, October 6-10 (1996).