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6.6 Effect of Mixing State on Criticality Safety Evaluation in MOX Powder and Additive

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Criticality safety analyses are discussed in which MOX powder and additive (e.g. zinc-stearate) are mixed in a powder treatment process of MOX fuel fabrication. The multiplication factor k_{eff} is largely affected by how they are mixed, i.e., how the density and volume change with the mixing. In general, k_{eff} increases when MOX powder is mixed with zinc-stearate. However, plutonium content and density of MOX powder make a difference in the k_{eff}'s changes. Especially, MOX powder with a higher plutonium content and a higher density is not always unsafe in terms of criticality if it is mixed with zinc-stearate.

KEYWORDS: MOX, criticality safety, k_{eff}, additive

1. Introduction

To assure criticality safety in processes handling MOX powder, the content of water or additive within MOX powder have to be carefully controlled. Excess addition of additive into a large homogenizer in the process of MOX powder handling could be one of hypothetical criticality accident scenarios since additive such as zinc-stearate contributes to neutron moderation. Criticality safety in a powder system depends on the density, volume, and equivalent water content of MOX powder. The mixing condition of the powder is important to make reasonable evaluations of criticality of MOX powder or to determine appropriate criticality accident conditions. However, it is not an easy task to identify mixture conditions because it requires some aspects of the powder technology. The authors discuss the effect of mixture conditions on criticality of MOX powder in this paper.

2. Two Mixing Assumptions

Here, as an abnormal event scenario, it is assumed that pure MOX powder is mixed with excess amount of additive (zinc-stearate, $[CH_3(CH_2)_{16}COO]_2Zn$) in a large homogenizer and a completely homogeneous mixture of MOX powder and zinc-stearate is formed by the mixing. Criticality calculations are performed for the homogeneous mixture to obtain the k_{eff}, and it is determined whether the criticality safety can be assured. To perform the calculations, one has to know the density, or volume of the mixture. When determining the density or volume, we assume two extreme mixing conditions as shown in Fig.1. The one assumption, mixing assumption (a), is that additive enters the void space of MOX powder. In this assumption, the initial density and volume of MOX powder remain unchanged. For example, when zinc stearate is added to MOX powder of 5.5 g/cm^3 and they are mixed homogeneously, the MOX density is assumed not to change from 5.5 g/cm^3. Since the theoretical density of MOX fuel is approximately 11 g/cm^3, the maximum volume fraction of the void space in the powder is 50% (=1-5.5/11). Another assumption, mixing assumption (b), is that the total volume of the homogenized mixture is the sum of the volume of MOX powder and zinc-stearate. In this assumption, the density of MOX powder decreases and the volume increases. After the void space of MOX powder is occupied by zinc-stearate, which is added on the mixing assumption (a), the mixing condition switches to the mixing assumption (b).

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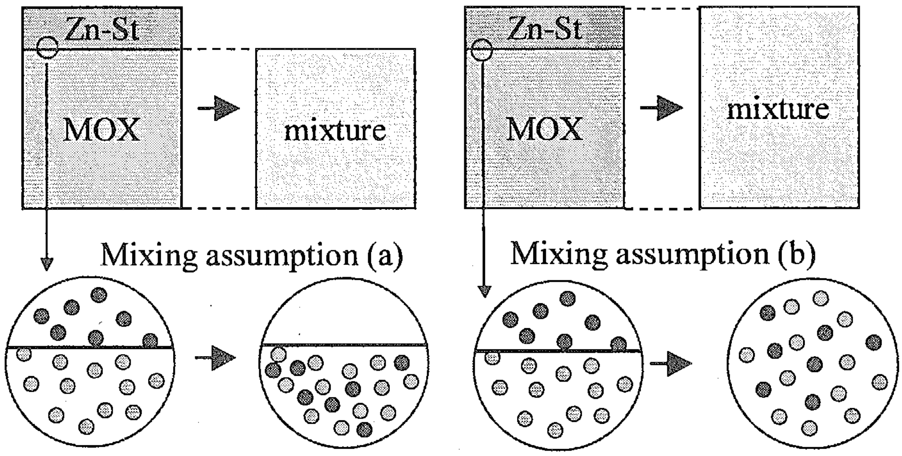

Fig.1 Two mixing assumptions

The effects of adding zinc-stearate to MOX powder on criticality in both mixing assumptions are compared. In the mixing assumption (a);

- Neutron moderation increases,
- Neutron leakage decreases.

In the mixing assumption (b);

- Neutron moderation increases,
- Geometrical buckling B^2 decreases,
- Density decreases.

The neutron leakage is proportional to DB^2 where D is one group diffusion coefficient. The decrease of the density due to addition of zinc-stearate means the decrease of D. Therefore, whether neutron leakage increases or decreases due to addition of zinc stearate depends on which effect is dominant, D or B^2.

3. Calculations

3.1 Comparison of mixing assumptions

Criticality calculations were performed for powder systems in which MOX powder and zinc stearate are mixed in a hypothetical homogenizer with the maximum volume of 1,000 liter[1)] as shown in Fig.2. A water reflector with the thickness of 2.5 cm is attached on the side surface of the homogenizer to take account of neutron reflection of surrounding structures. A continuous energy Monte Carlo code MCNP 4C and a pointwise cross section library based on JENDL-3.2 were used for the calculations. In the criticality calculations, zinc-stearate was replaced by water with the same mass, which has very little effect on criticality[1)]. This assumption is underpinned by the fact that mass ratios of hydrogen in zinc-stearate and water are 0.1116 and 0.1119, respectively and both ratios are almost the same.

First of all, criticality calculations were performed for a MOX powder system in which 640 kg-MOX powder with 50% plutonium content and 3.5g/cm^3 density is mixed with zinc- stearate. The density of zinc stearate is 0.3 g/cm^3, which is a usual apparent density of zinc-stearate[1)]. "Equivalent water content" is regarded as a synonym with the weight fraction of zinc-stearate. Note that the mass of 640 kg-MOX powder and the equivalent water content larger than 5% are not practically anticipated. In addition, the plutonium content is normally less than 18% in a final blending, and MOX powder with 50% plutonium content is very unlikely to be poured into a large homogenizer for final blending because of rigid control of plutonium content. Thus, this situations postulates that three criticality controls (mass control, moderation control, and plutonium control) fail at the same time. In the mixing assumption (a), k_{eff} increases with the equivalent water content until the void space of MOX powder is filled with zinc-stearate at the equivalent water content of 6%. This is due to the

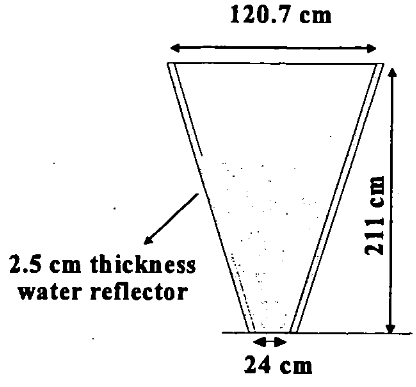

Fig.2 Hypothetical homogenizer

increase of neutron moderation and the decrease of neutron leakage. Beyond this equivalent water content, the mixing condition changes to the assumption "(b)", and k_{eff} decreases with the equivalent water content, which is caused mainly by the decrease of the density of MOX powder. In the mixing assumption (b), k_{eff} also increases with the equivalent water content because of the increase of neutron moderation. However, k_{eff} in the mixing assumption (b) is much smaller than that in the mixing assumption (a) because MOX powder is diluted with the increase of the equivalent water content.

An experiment was performed to study an actual mixing condition of MOX powder and zinc-stearate[2)]. In the experiment, tungsten powder was used as a substitute material for MOX powder. This experiment indicated that a true mixing condition is close to (b) rather than (a). Therefore, if one considers zinc-stearate is gradually added to pure MOX powder, k_{eff} changes according to the mixing assumption (b) and the mixing assumption (a) gives much higher k_{eff} than an actual situation. That is, criticality safety evaluations based on the mixing assumption (a) give conservative results.

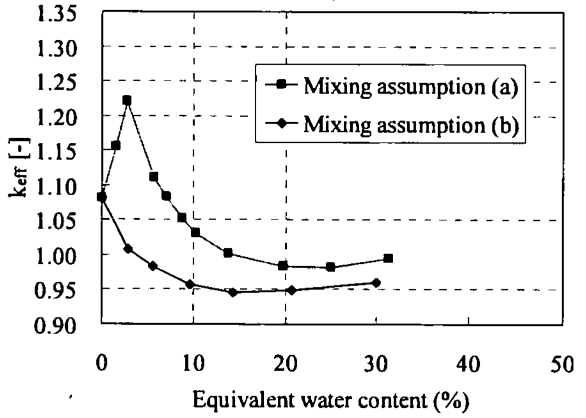

Fig.3 k_{eff} vs. equivalent water content for 640kg MOX(50%Pu content, 5.5g/cm^3), zinc stearate (0.3g/cm^3).

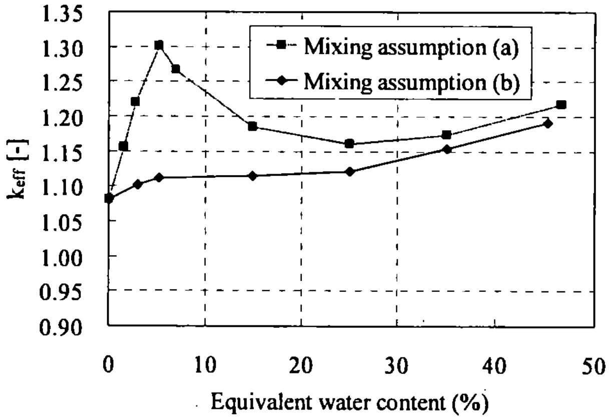

Fig.4 k_{eff} vs. equivalent water content for 640kg MOX(50%Pu content, 5.5g/cm^3), zinc stearate (0.6g/cm^3).

3.2 Effect of powder density

Next, similar criticality calculations were performed for 640 kg-MOX powder with 50% plutonium content and 5.5g/cm^3 density, and it is mixed with zinc-stearate with the density of 0.3g/cm^3. The results are shown in Fig.3[2)]. The tendency of k_{eff} in the mixing assumption (a) is essentially the same as in the case of 3.5g/cm^3 MOX. The result shows k_{eff} is over 1.0 without zinc-stearate in this case. However, this case is more unlikely than in the case of 3.5g/cm^3 MOX because 5.5g/cm^3 MOX powder density is much higher than a usual MOX powder density treated in a homogenizer. In the mixing assumption (b), mixing zinc-stearate with a higher density MOX powder causes the decrease of k_{eff} due to the decrease of MOX density unlike the case of 3.5g/cm^3 MOX. Therefore, an addition of zinc-stearate is not always unsafe from the viewpoint of criticality.

The density of zinc-stearate in powder state can be maximized up to approximately 0.6g/cm^3 (the tap density of zinc-stearate powder). Figure 4 shows calculated k_{eff} when 640 kg-MOX powder with 50% plutonium content and 5.5g/cm^3 density is mixed with zinc-stearate with the density of 0.6g/cm^3. While k_{eff} decreases in the mixing assumption "(b)" by adding zinc-stearate with the density of 0.3g/cm^3, the addition of zinc-stearate with the density of 0.6g/cm^3 increases k_{eff} even in the assumption "(b)". This is because the effect of larger neutron moderation with the higher density of zinc-stearate exceeds the effect of the density decrease of MOX powder. Therefore, the effect of zinc-stearate on criticality depends on the density of zinc-stearate as well as the mixing condition.

3.3 Effect of plutonium content

The addition of 50% plutonium content into a final blending hardly occurs from a technical standpoint. The normal plutonium content in a final blending is no greater than 18%. Here, similar criticality calculations were performed for 640 kg-MOX powder with 18% plutonium content and 5.5g/cm^3 density as shown in Fig.5[2]. The density of zinc-stearate is 0.3g/cm^3. The k_{eff} increases with the equivalent water content in the mixing assumption (b), which shows the opposite tendency of 50% plutonium content as compared with Fig.3. Plutonium content in MOX powder affects the neutron spectrum. MOX powder with lower plutonium content has larger neutron moderation effect of water, or zinc-stearate.

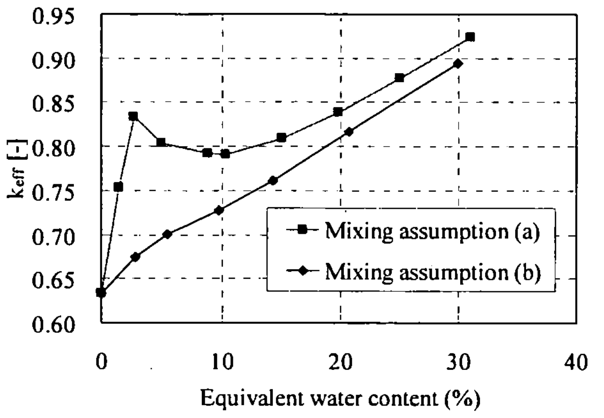

Fig.5 k_{eff} vs. equivalent water content for 640kg MOX(18%Pu content, 5.5g/cm^3), zinc stearate (0.3g/cm^3).

4. Conclusion

If one considers criticality of powder fuel mixed with additive such as zinc-stearate, it is important to know how the density or volume changes with mixing. A realistic mixing state may be represented by the mixing assumption (b) in which the density of MOX powder decreases with mixing with additive. In this mixing assumption (b), k_{eff} mostly increases with the equivalent water content but the increase in k_{eff} is much smaller than in the mixing assumption (a) in which the density or volume of MOX powder does not change with mixing. If MOX powder with a higher density and higher plutonium content is mixed with zinc-stearate with its apparent density (0.3g/cm^3), k_{eff} unusually decreases with the equivalent water content in the mixing assumption (b). Thus, an addition of zinc-stearate is not always unsafe in terms of criticality. However, k_{eff} of MOX powder with normal density and plutonium content, which is treated in a final blending, usually increases with addition of zinc-stearate.

References

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7. Keynote Speech-2

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