Criticality of Mixtures of Plutonium and High Enriched Uranium

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This paper presents a criticality evaluation of moderated homogeneous plutonium-uranium mixtures. The fissile media studied are homogeneous mixtures of plutonium and high enriched uranium in two chemical forms: aqueous mixtures of metal and mixtures of nitrate solutions. The enrichment of uranium considered are 93.2wt.% ^{235}U and 100wt.% ^{235}U. The ^{240}Pu content in plutonium varies from 0wt.% ^{240}Pu to 12wt.% ^{240}Pu.

The critical parameters (radii and masses of a 20 cm water reflected sphere) are calculated with the French criticality safety package CRISTAL V0. The comparison of the calculated critical parameters as a function of the moderator-to-fuel atomic ratio shows significant ranges in which high enriched uranium systems, as well as plutonium-uranium mixtures, are more reactive than plutonium systems.

KEYWORDS: critical dimension, sphere, plutonium, HEU, homogeneous metal water mixture, nitrate solution, full water reflected

1. Introduction

The available experimental criticality data of homogeneous mixtures of plutonium-uranium systems are limited to mixtures of plutonium with natural or depleted uranium typical for fast breeder reactor or MOX fuel fabrication process applications.

In some research reactor and other applications, mixtures of plutonium with high enriched uranium (HEU) can be found but adequate criticality data for mixtures of plutonium with high enriched uranium are rare. Some experiments are presented in the OECD Handbook of Critical Experiments [1], but not sufficient to derive and justify a critical parameter set for plutonium-HEU mixtures.

In order to determine the most reactive mixture of plutonium with high enriched uranium as a function of the moderator-to-fuel atomic ratio H/X (X=U+Pu, U, Pu) a parameter study was undertaken. The aim of this study is to determine the range of H/X in which either the plutonium system or the HEU systems are a bounding medium for plutonium-HEU mixtures. The study includes homogeneous metal water mixtures as well as aqueous nitrate solutions.

At first pure ^{239}Pu and pure ^{235}U systems are studied and their infinite multiplication factors k_{inf} are compared over the full range of moderation.

In a following step more complex fissile media are compared. Although the plutonium may be composed of various plutonium isotopes the study assumes only the isotopes ^{239}Pu and ^{240}Pu. The HEU in the plutonium-uranium mixture considered here is 93.2wt.% ^{235}U and 6.8wt.% ^{238}U. Consequently the following three media are studied:

i. Homogeneous water moderated systems of ^{239}Pu and ^{235}U (as metal and nitrate solution).
ii. Homogeneous mixtures of plutonium metal and HEU-metal with water. The fissile media are plutonium composed of ^{239}Pu with 0wt.% ^{240}Pu up to 12wt.% ^{240}Pu and HEU.
iii. Homogeneous mixtures of Pu^{III}-nitrate solutions with HEU nitrate solutions. The fissile media are $Pu(NO_3)_3 \cdot 5H_2O$ with the same isotopic vector as described in (ii.) and $UO_2(NO_3)_2 \cdot 6H_2O$ with an uranium enrichment of 93.2wt.% ^{235}U. To be conservative, Pu^{III}-nitrate solution is used in the study instead of Pu^{IV}-nitrate, since it is more reactive.

The number densities of the nitrate solutions are calculated as a function of H/X with the code CIGALES V2.0 which uses an 'isopiestic' density law for Pu^{III} and Pu^{IV}-nitrate solutions[2]. The minimum possible H/Pu ratio for Pu^{III}-nitrate solutions given by the code CIGALES V2.0 is limited on its lower side to about H/Pu = 24.4[2]. The number densities of the plutonium metal and HEU metal water mixtures are calculated with CIGALES V2.0 on the basis of a simple additional and individual volume and mass (AIVM) law.

The calculations are performed with the criticality safety package CRISTAL V0[3]. The cross sections are generated in the code APOLLO 2 in a 20 energy group structure based on the CEA93-V4 neutron cross section library[4]. The critical dimensions are determined by radius iteration with the APOLLO 2-SN module.

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2. Results

2.1 Homogeneous systems of ^{239}Pu and ^{235}U

Fig.1 compares the infinite multiplication factor k_{inf} of two metal water media and two nitrate solutions as a function of the H/X ratio. It is obvious that the ^{239}Pu systems are not a criticality bounding media over the full range of moderation. A simple comparison of the microscopic cross sections of ^{235}U and ^{239}Pu substantiates this fact.

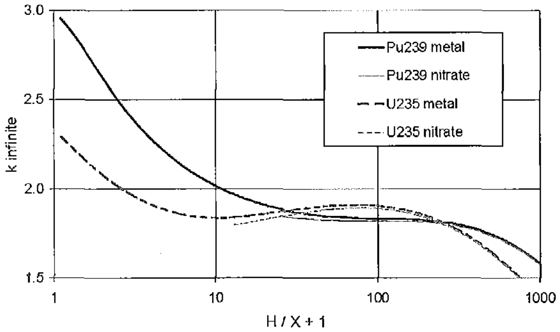

Fig.1 Infinite multiplication factor k_{inf} versus H/X of four media containing ^{235}U and ^{239}Pu

Fig.2 shows the ratio of the production-to-absorption cross sections of ^{235}U and ^{239}Pu. The energy depending cross sections $\nu\Sigma^{fis}$ (E) and Σ^{abs}(E) of the isotopes ^{235}U and ^{239}Pu are taken from the JEF2.2 nuclear data bank[4].

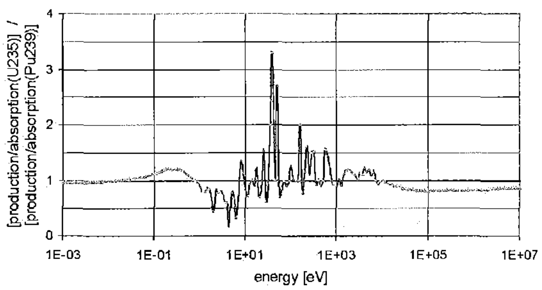

Fig.2 Ratio of ^{235}U to ^{239}Pu production-to-absorption cross section versus energy

It is apparent that in the energy range between 0.1eV and 10keV this ratio is mainly greater than 1 that indicates that ^{235}U has in this energy range a higher neutron multiplication rate than ^{239}Pu. Therefore the k_{inf} of ^{235}U-water mixture is in the H/X range between ~25 and ~250 higher than the k_{inf} of the ^{239}Pu-water mixture.

In case of nitrate solutions the difference in the k_{inf} of ^{235}U-nitrate solution and ^{239}Pu-nitrate solution increases due to the different crystalline composition of the nitrate compounds of U-nitrate and Pu-nitrate.

2.2 Homogeneous systems of Pu and ^{235}U

2.2.1 Comparison of Pu-metal and ^{235}U-metal systems

Let us consider now the influence of ^{240}Pu in the Pu metal system. The presence of ^{240}Pu in the plutonium lowers the values of k_{inf}, therefore the differences between the k_{inf} of ^{235}U-metal and the k_{inf} of Pu-metal increases with increasing ^{240}Pu content as indicated in Fig.3. Moreover this figure shows that the range where ^{235}U systems are more reactive than plutonium systems enlarges with increasing ^{240}Pu content in the plutonium.

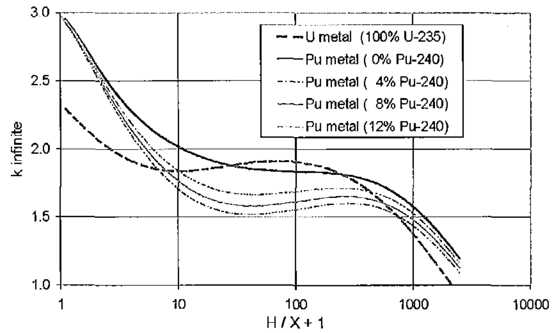

Fig.3 Infinite multiplication factor k_{inf} versus H/X for ^{235}U metal water and various plutonium metal water systems

2.2.2 Comparison of ^{235}U-nitrate and Pu-nitrate systems

In Fig.4 the infinite multiplication factor k_{inf} is compared for uranium nitrate and plutonium nitrate solutions versus the H/X ratio (X=U, Pu). The uranium is 100wt.% ^{235}U. Different ^{240}Pu contents in plutonium are considered. The plutonium nitrate is supposed to be Pu^{III}-nitrate.

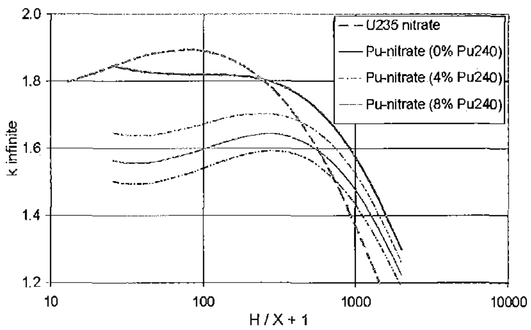

Fig.4 Infinite multiplication factor k_{inf} versus H/X for ^{235}U-nitrate and various plutonium nitrate solutions

As for the metal systems the increasing ^{240}Pu content enlarges the differences between the ^{235}U-nitrate solution and plutonium nitrate solutions in the H/X range around H/X=100.

2.3 Homogeneous mixtures of plutonium and high enriched uranium

2.3.1 Critical parameter of plutonium metal and uranium metal systems

At first, the critical radius of a 20cm water reflected sphere is compared for HEU-systems and plutonium systems. The ^{240}Pu content in the plutonium is varied from 0wt.% ^{240}Pu up to 12wt.% ^{240}Pu. The enrichment of uranium is 93.2wt.% ^{235}U. Fig.5 shows the critical radii of full water reflected spheres as a function of the H/X atomic ratio.

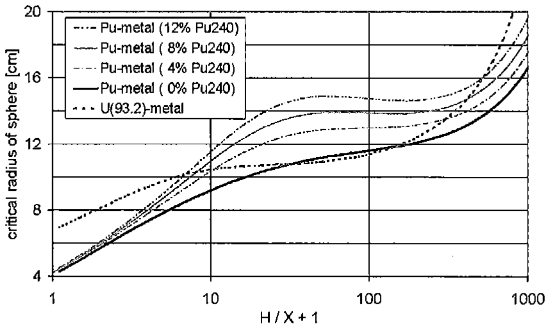

Fig.5 Critical radius of a 20cm water reflected sphere versus H/X for HEU metal and various plutonium metal water mixtures

As already indicated in Fig.3, the H/X range in which the HEU system is more reactive than plutonium systems extends with increasing ^{240}Pu content in the plutonium.

2.3.2 Critical parameters of uranium-plutonium metal mixtures

In the two following figures critical parameters of mixtures of plutonium-uranium metal water systems are illustrated. The isotopic composition of the plutonium is now conserved and defined as follows:

$^{239}Pu/Pu_{tot}$ = 92 wt.%, $^{240}Pu/Pu_{tot}$ = 8 wt.%, where

$Pu_{tot} = {}^{239}Pu + {}^{240}Pu$. (1)

The critical radii and the corresponding critical masses of a sphere are compared in Fig.6 and Fig.7 for various mixtures of plutonium (8wt.% ^{240}Pu) and uranium (93.2wt.% ^{235}U).

The uranium content c_U in the mixture is defined as:

c_U [wt.%] = mU/(mU+mPu) • 100 (2)

with $U = {}^{235}U + {}^{238}U$ and $Pu = {}^{239}Pu + {}^{240}Pu$.

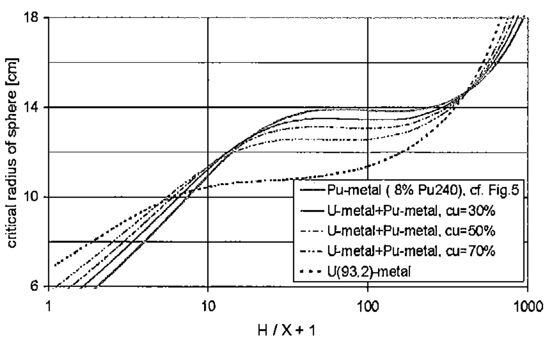

Fig.6 Critical radius of a 20cm water reflected sphere of a plutonium-uranium metal water mixture versus H/X, plutonium with 8wt.% ^{240}Pu

The critical radius curves show that there is a range of moderation between H/X ≃ 7 and H/X ≃ 400 where homogeneous plutonium-uranium mixtures are more reactive than plutonium systems. This means that any ‘pollution’ of a plutonium water system with HEU leads to an increase of the system reactivity if the system is in the moderation range between H/X ≃ 7 and H/X ≃ 400. It is also significant

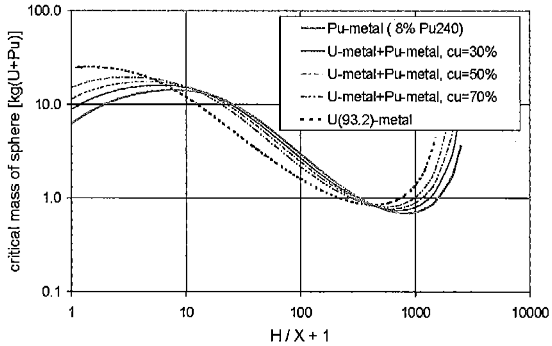

Fig.7 Critical mass of a 20cm water reflected sphere of a plutonium-uranium mixture versus H/X for various uranium concentrations c_U, plutonium with 8wt.% ^{240}Pu

that in this intermediate moderation range plutonium-HEU mixtures have lower critical masses as pure plutonium systems.

In the range of H/X ≲ 7 and in the range of H/X ≳ 400 plutonium systems with 8wt.% ^{240}Pu are more reactive than plutonium-HEU mixtures or HEU-systems. Consequently the minimum critical radius of sphere occurs for Pu-metal. The minimum critical mass of Pu-metal that occurs in the range of H/X ≳ 400 is bounding for all plutonium-HEU mixtures.

2.4 Aqueous plutonium nitrate and uranium nitrate solutions mixtures

2.4.1 Critical parameter of plutonium nitrate and uranium nitrate solutions.

At first critical radii and critical masses of a 20cm water reflected sphere are compared in Fig.8 for HEU-nitrate solution and Pu^{III}-nitrate solutions with various ^{240}Pu contents. It is obvious that in the optimum moderated H/X range HEU uranium nitrate is more reactive than plutonium nitrate.

The comparison of the critical radii shows that below H/X = 120 HEU-nitrate is bounding for plutonium nitrate. For higher H/X ratios ^{239}Pu-nitrate is bounding for HEU-nitrate. The crossing point is shifting to higher H/X values if the ^{240}Pu content in the plutonium increases.

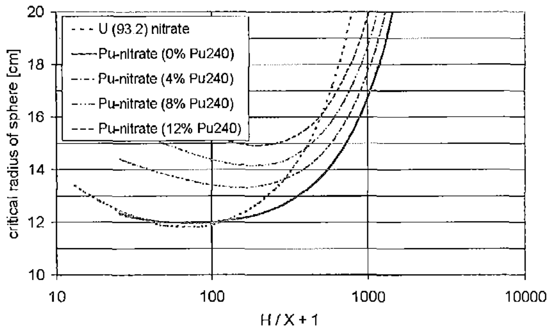

Fig.8 Critical radius of a 20cm water reflected sphere of HEU-nitrate and plutonium nitrate solutions with various ^{240}Pu contents

2.4.2 Critical parameters of plutonium nitrate-uranium nitrate solution mixtures

Critical radii and masses of mixtures of plutonium nitrate and uranium nitrate solutions are shown in Fig.9 and Fig.10. The isotopic composition of the plutonium is the same as described in (1). The HEU-content c_U in the plutonium-uranium nitrate solution mixture is defined as in (2).

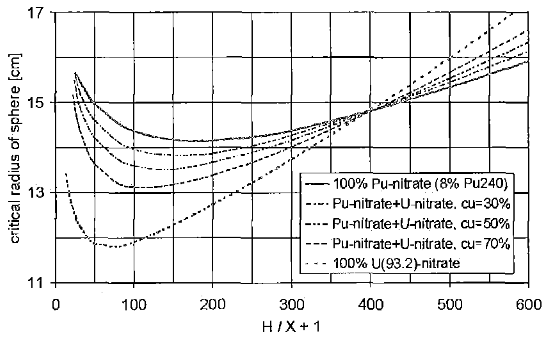

Fig.9 Critical radius of a 20cm water reflected sphere of plutonium nitrate-uranium nitrate solution mixtures for various HEU-nitrate contents, plutonium with 8wt.% ^{240}Pu

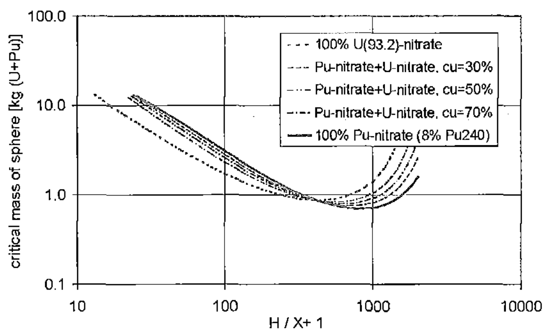

Fig.10 Critical mass of a 20cm water reflected sphere of plutonium nitrate-uranium nitrate solution mixtures for various HEU-nitrate contents, plutonium with 8wt.% ^{240}Pu

The comparison of the critical parameters shows that in the range of optimum of moderation plutonium nitrate-uranium nitrate solution mixtures have smaller critical dimensions than pure plutonium nitrate solutions.

Contrary to the metal-water systems, the minimum critical dimensions of nitrate systems occur for HEU-nitrate, that is bounding for all mixtures of HEU-nitrate and plutonium nitrate. But the minimum critical mass again occurs for plutonium nitrate in moderation range of $H/X \gtrsim 400$ as for the metal-water systems.

2.5 Summary of results

2.5.1 Metal systems

The critical parameter curves of mixtures of plutonium metal with 8wt.% ^{240}Pu and HEU metal show three significant ranges:

In the range $0 < H/X \lesssim 7$ plutonium systems have smaller critical dimensions than plutonium-uranium mixtures and than pure uranium systems. The minimum critical radius occurs in this range for plutonium metal of maximum density.

In the range $7 \lesssim H/X \lesssim 400$ plutonium-uranium mixtures have smaller critical dimensions than pure plutonium systems. Pure HEU-metal systems are in this range bounding for all plutonium-HEU mixtures.

In the range $H/X \gtrsim 400$ plutonium systems have smaller critical dimensions than plutonium-HEU mixtures and than pure uranium systems. The minimum critical mass occurs in this range for plutonium and is bounding for all critical masses of mixtures of plutonium and HEU.

2.5.2 Nitrate solutions

Contrary to the metal water systems, the critical parameter curves for plutonium nitrate-HEU nitrate solution mixtures show only two H/X ranges:

In the range $24 \lesssim H/X \lesssim 400$ plutonium nitrate-HEU-nitrate solution mixtures have lower critical dimensions than pure plutonium nitrate solutions. HEU-nitrate solution is bounding for all plutonium nitrate-HEU-nitrate solution mixtures. The minimum critical dimension of a sphere occurs for HEU-nitrate.

In the range $H/X \gtrsim 400$ plutonium nitrate solutions have lower critical dimensions than plutonium nitrate-HEU-nitrate solutions mixtures. The minimum critical mass of plutonium nitrate therefore in this range is smaller than the minimum critical mass of HEU-nitrate.

3. Conclusion

The comparison of the critical parameters of HEU-systems and plutonium systems shows that there is a moderation range of H/X where HEU-systems are systematically more reactive than plutonium systems. Consequently mixtures of plutonium-uranium systems are found in this range of H/X also more reactive than plutonium systems. The width of this H/X range and the differences in the critical parameters increases with increasing of the ^{240}Pu content in plutonium. The HEU content c_U in the mixture does not significantly change the H/X range, but influences the strength of this "shifting effect" on the critical parameters.

For metal systems the moderation range where HEU-systems are found to be more reactive than plutonium systems with 8wt.% ^{240}Pu is between H/X $\simeq$ 7 and H/X $\simeq$ 400. This moderation range corresponds to plutonium-uranium concentrations ranging from ~3.1g/cm^3 to ~0.06g/cm^3 that are in the range of relevant process applications.

For nitrate solutions the upper boundary of the moderation range up to which the HEU-systems are more reactive than plutonium systems with 8wt.% ^{240}Pu is H/X $\simeq$ 400 corresponding to a plutonium-uranium concentration of ~0.065g/cm^3.

For oxide systems similar trends as for metal systems are found.

In conclusion the reactivity of a moderated plutonium system (metal, oxide or nitrate) in the H/X ranges described above will be influenced by the presence of additional HEU in the mixture. Any establishment of criticality safe limits for plutonium systems in this H/X range must be based on the critical parameters of HEU or of plutonium-HEU mixtures if the concentration of HEU in the system can be guaranteed to a certain limit.

References

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