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RATIONALE FOR THE DEUTERIUM AND BERYLLIUM LIMITATIONS IN THE MODIFIED PARA. 672(A) IN IAEA TRANSPORT REGULATION

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ABSTRACT
Para. 672 in the IAEA Regulations (TS-R-1)[1)] shows the exceptions from the requirements for package containing fissile material. This provision is important and widely applicable for the reasonable transportation of material with small amounts of fissile material. But deuterium and beryllium are restricted to less than 0.1% of the maximum allowable fissile material mass in para. 672(a) in the 1996 version (shown in Appendix-1). It means the maximum allowance is just less than 0.4 grams. This is because the fissile material limitations in the provision are based on minimum critical mass for the hydrogenous material and deuterium or beryllium may be more effective than hydrogen in terms of critical safety[2),3)].
Deuterium exists naturally in hydrogen (about 0.015 atomic%) and the average concentration of beryllium in the earth's crust (Clark Number) is 1.5ppm[4)]. Massive LLW (Low Level Waste) that is stabilized by cement has been transported by exclusive ships in the world. Total deuterium mass naturally contained in the consignment might have exceeded several hundred grams. As a result, the LLW should be classified as fissile material even though there is no possibility of reaching critical.
To rationalize the situation, the revised proposal have been submitted from Japan sequentially based on the detailed quantitative evaluation of deuterium and beryllium on their effects. As a result, exception of deuterium with natural concentration in hydrogen from the restriction has been approved in the Regulations (TS-R-1) and the Advisory Material (TS-G-1.1) of 2003 version. Furthermore, exception of beryllium with less than 0.1% concentration from the restriction has been also approved in the 2007 version.

INTRODUCTION
Many proposals from several nations and organizations have been submitted for the exceptions from the requirements for package containing fissile material shown in para. 672 in TS-R-1 for about ten years. Many discussions have been held among the experts on critical safety in

working groups under the Revision Panel meeting (RPM), TRANSSC or Consultant Meeting (CSM) in IAEA. Although some proposals are under discussion now, proposals for deuterium and beryllium from Japan have been approved and subparagraph 672(a) has been revised. The rationales of these proposals have been discussed in the IAEA meeting, but the basis of the revision of the regulations is not published. This paper shows the rationale for the revision on deuterium and beryllium and clarifies the process of revision of the regulation.

REVISION FOR DEUTERIUM (2003 VERSION)

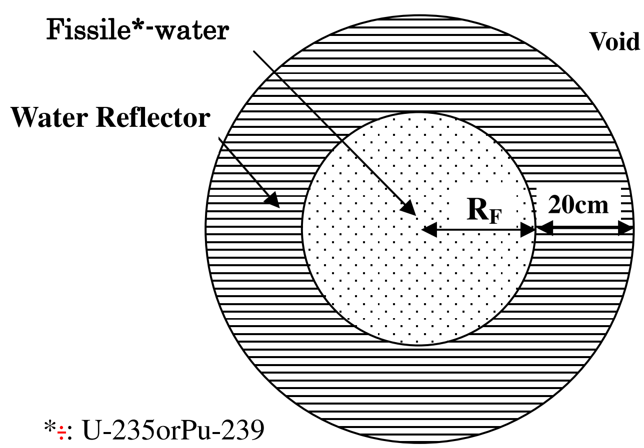
Calculation conditions

The deuterium effects on criticality safety are estimated using a simple model with various fissile and deuterium concentrations. The calculations are performed for the water reflected homogenous fissile-water sphere system shown in Figure 1. Two kinds of fissile materials, U-235 (100%) and Pu-239 (100%), are considered and the fissile material concentration (expressed by hydrogen to fissile atomic ratio H/X, where H means hydrogen and X means fissile material) varies from 10 to 10^6. The one-dimensional discrete ordinate code XSDRN and 238-group library in SCALE code system[5] are used. Also, a temperature of 20 degrees Celsius is used in the calculation.

The multiplication factors are calculated for various deuterium concentrations (atomic ratio: D/H) in the fissile-water region. The thickness of the reflector is fixed at 20 cm, whereas the radius (R_F) of the fissile-water sphere is established to achieve near criticality (k_{eff}=0.95) in the reference case where the deuterium concentration (D/H) is equal to zero. Although R_F can be determined in the reference case for high fissile concentrations (H/X=10, 10^2, 10^3), the multiplication factors for low fissile concentrations (H/X=10^4, 10^5, 10^6) are very low in each reference case. So R_F is determined to be nearly critical under the condition that the deuterium concentration is 100% (D/H=1.0). R_F of each case is also shown in Figure1.

The multiplication factors for various deuterium concentrations (D/H) are calculated with the same R_F for each fissile concentration (H/X).

It is thought that deuterium may be more effective than hydrogen as a reflector material. The multiplication factors are also calculated at various deuterium concentrations (D/H) in a water reflector. The calculations are



*: U-235orPu-239

	R_F:Radius of Fissile-water Sphere (cm)	
H/X	X = U-235	X = Pu-239
10	9.65	8.81
10^2	10.56	10.81
10^3	19.88	15.25
10^4	44.89 (41.29)**	37.81(33.27)**
10^5	127.9 (300)**	97.91 (300)**
10^6	918 (infinity)**	431 (infinity)**

**: value in the parentheses used for beryllium case only performed for the typical case where the fissile material is U-235 and H/X=100 in the fissile-water sphere region.

Calculation results

The calculated reactivity effects of deuterium in the moderator are shown in Table 1 and Figures 2 and 3. Table 1 shows the reactivity changes (⊿k) from each reference case (the deuterium concentration is zero) and the deuterium quantities (in grams) as well as the multiplication factor for various concentrations of deuterium and fissile material in U-235 100% case. These results show that the reactivity effects of deuterium are negative when the fissile concentration is high ($H/X<10^3$) and positive when the fissile concentration is low ($H/X>10^3$). When R_F is small (<20 cm) and the fissile concentration is high, the effects of the increase in neutron leakage from the system are larger than the effects from the decrease in neutron absorption due to the increase of deuterium. Conversely, the effects of neutron leakage from the system are relatively small and the decrease in neutron absorption due to the increase of deuterium makes the multiplication factor increase when R_F is large (>30 cm) and the fissile concentration is low.

No significant difference is observed between U-235 and Pu-239 as shown in Figures 2 and 3.

Table 1 Reactivity effects of deuterium in the water moderator (U-235 : 100%)

D/~~D+~~H [%]	H/X=10 k_{eff}~~k_{eff}~~	H/X=10 Δk[%]	H/X=10 Deuterium [g]	H/X=100 k_{eff}~~keff~~	H/X=100 Δk[%]	H/X=100 Deuterium [g]
0	0.94998	0.0	0.0	0.94981	0.0	0.0
0.015	0.94995	-0.003	1.1E-01	0.94978	-0.003	1.6E-01
0.1	0.94981	-0.017	7.4E-01	0.94960	-0.021	1.1E+00
1	0.94827	-0.171	7.4E+00	0.94771	-0.210	1.1E+01
10	0.93234	-1.764	7.4E+01	0.92760	-2.221	1.1E+02
100	0.69853	-25.145	7.4E+02	0.56032	-38.949	1.1E+03

D/~~D+~~H [%]	$H/X=10^3$ k_{eff}~~Keff~~	$H/X=10^3$ Δk[%]	$H/X=10^3$ Deuterium [g]	$H/X=10^4$ k_{eff}~~keff~~	$H/X=10^4$ Δk[%]	$H/X=10^4$ Deuterium [g]
0	0.94992	0.0	0.0	0.30663	0.0	0.0
0.015	0.94994	0.002	1.1E+00	0.30667	0.004	1.3E+01
0.1	0.95007	0.015	7.3E+00	0.30686	0.023	8.4E+01
1	0.95142	0.150	7.3E+01	0.30896	0.233	8.4E+02
10	0.96423	1.432	7.3E+02	0.33159	2.496	8.4E+03
100	0.73537	-21.455	7.3E+03	0.95010	64.347	8.4E+04

D/~~D+~~H [%]	$H/X=10^5$ k_{eff}~~keff~~	$H/X=10^5$ Δk[%]	$H/X=10^5$ Deuterium [g]	$H/X=10^6$ k_{eff}~~Keff~~	$H/X=10^6$ Δk[%]	$H/X=10^6$ Deuterium [g]
0	0.03995	0.0	0.0	0.00414	0.0	0.0
0.015	0.03995	0.001	2.9E+02	0.00414	0.0001	1.1E+05
0.1	0.03999	0.004	1.9E+03	0.00415	0.0004	7.2E+05
1	0.04034	0.039	1.9E+04	0.00419	0.004	7.2E+06
10	0.04423	0.428	1.9E+05	0.00460	0.046	7.2E+07
100	0.95019	91.025	1.9E+06	0.94997	94.582	7.2E+08

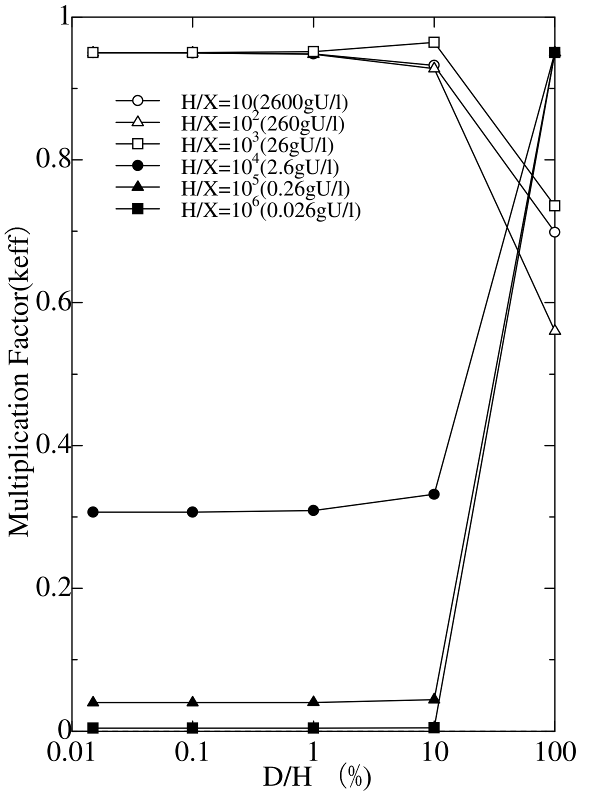


Figure 2 Reactivity effects of deuterium in the moderator of ^{235}U -H_2O system

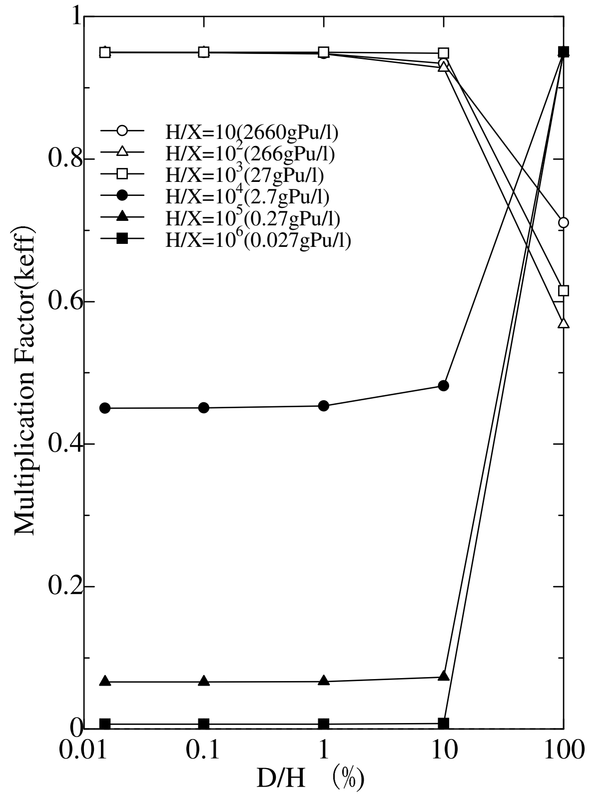


Figure 3 Reactivity effects of deuterium in the moderator of ^{239}Pu -H_2O system

The calculated reactivity effects of deuterium in the reflector are shown in Table 2. These results indicate that the reactivity effects of deuterium are positive, but the reactivity effects in the reflector are smaller than in the moderator.

According to these calculation results for deuterium in the moderator and the reflector, the reactivity effects of deuterium are less than 0.3 %⊿k when the deuterium concentration (D/H) is less than 1 atomic %[6)].

Table 2 Reactivity effects of deuterium in the reflector

U235:100%

D/~~(D+H)~~	H/X=100	
[%]	k_{eff}	Δk [%]
0	0.94981	0.0
0.015	0.94982	0.001
0.1	0.94990	0.009
1	0.95074	0.093
10	0.95040	0.068

<u>Revision of TS-R-1</u>

The maximum deuterium limit in subparagraph 672(a) in the 1996 version is 0.4 g, which corresponds to a fissile material limit of 400 g. For example, 0.4 g of deuterium is approximately equal to the case of 0.05% of deuterium concentration for H/X=10 and 0.0005% of deuterium concentration for $H/X=10^4$ as shown in Table 1.

According to the discussion on the basis of these calculation results at TRANSSC VII in 2002, the last part of subparagraph 672(a) in TS-R-1 (underlined sentence in Appendix-1) and 672.4 in TS-G-1.1 have been revised as follows;
(TS-R-1 1996 Edition As Amended 2003)
Neither beryllium nor deuterium in hydrogenous material enriched in deuterium shall be present in quantities exceeding 1% of the applicable consignment mass limits provided in Table XII.

(TS-G-1.1/2003, 2005 Edition)
672.4 *In assessing the mass of deuterium in a consignment, deuterium contained in hydrogenous materials in natural concentrations should not be considered.*

Furthermore, TS-R-1 was revised again to clarify the meaning for exempted concentration of deuterium in 2005 edition as follows;

(TS-R-1 2005 Edition)
Neither beryllium nor deuterium shall be present in quantities exceeding 1% of the applicable consignment mass limits provided in Table 12, except for deuterium in natural concentration in hydrogen.

REVISION FOR BERYLLIUM (2007 VERSION)

Calculation conditions

Beryllium effects on criticality are also estimated in the same manner and with the same model (Figure 1) are used for the deuterium calculations. The multiplication factors are calculated with various beryllium concentrations (Be/(Be+H2O)) in the homogeneous fissile-water region. The multiplication factors for the low fissile concentration cases (H/X=10^5 and 10^6) are low even for the case of an infinite system with a 100% beryllium moderator. So 300 cm is used for R_F for the case of H/X=10^5, while an infinite radius system is used for the case of H/X=10^6.
The multiplication factors are also calculated for various beryllium volume concentrations (Be/(Be+H_2O)) in the water reflector. The calculations are only performed for the typical case where the fissile material is Pu-239 and the fissile material concentrations are (H/Pu)= 100 and 10^4 in the fissile-water sphere region. The thickness of the reflector as shown in Figure 1 is 50 cm for this case only.
Furthermore, genuine beryllium layer around fissile material may be more effective for the multiplication than the homogeneous reflector. The multiplication factors are calculated with the genuine beryllium reflector, as shown in Figure 4, thickness of beryllium layer between fuel region and water reflector. The calculations are also performed for the above typical case.

Calculation Results

The calculated reactivity effects of beryllium in the moderator are shown in Figure 5 and 6. These results show that the reactivity effects of beryllium are negative when the fissile concentration is high (H/X<10^3) and the effects are positive when the fissile concentration is low (H/X>10^3). These tendencies and orders of magnitude are more or less the same as the deuterium case mentioned previously.
The multiplication factor is the highest for the case where the beryllium concentration (Be/(Be+H_2O)) is

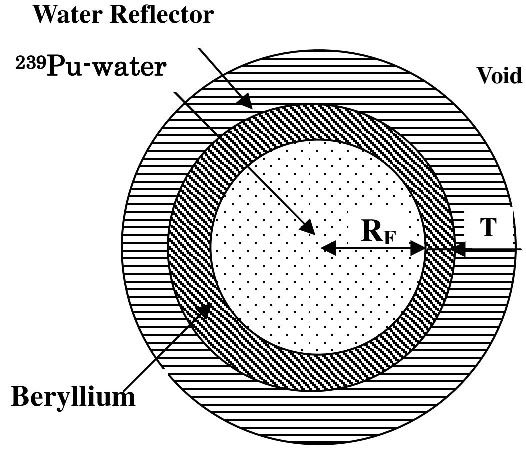


Figure 4 Calculation Model for beryllium reflected 239Pu-water system

50 % and the fissile concentration is $H/X=10^3$. It is thought that this condition is optimum for neutron leakage, moderation and absorption.

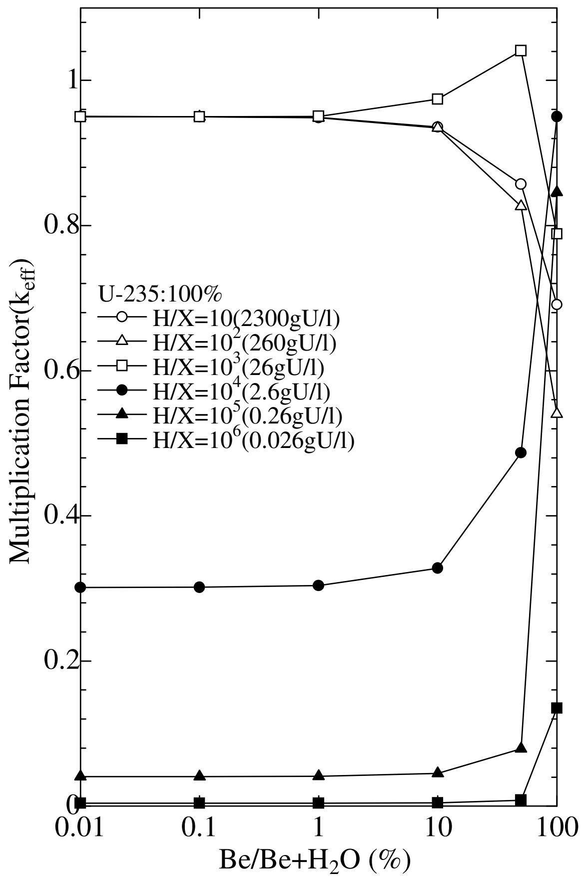


Figure 4 Reactivity effects of Beryllium in the moderator of ^{235}U -H_2O system

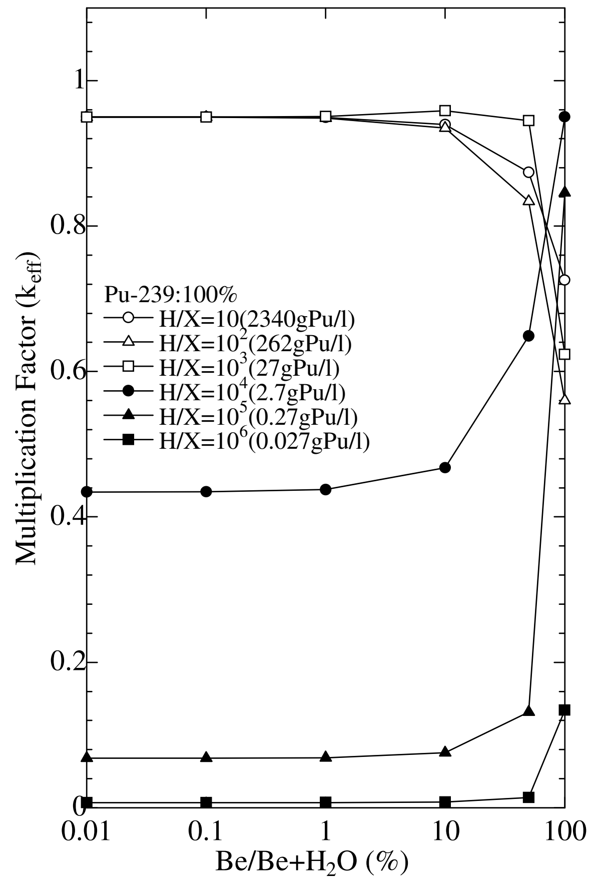


Figure 5 Reactivity effects of Beryllium in the moderator of ^{239}Pu -H_2O system

Table 3 Reactivity effects of beryllium in the water reflector (Pu-239 : 100%)

Be/Be+H_2O	H/X=100			H/X=10^4		
[%]	keff	Δk[%]	Be [g]	k_{eff}keff	Δk[%]	Be [g]
0	0.94974	0.0	0.0	0.43424	0.0	0.0
0.01	0.94975	0.001	1.7E+02	0.43427	0.003	4.2E+02
0.1	0.94994	0.020	1.7E+03	0.43428	0.004	4.2E+03
1	0.95186	0.213	1.7E+04	0.43438	0.014	4.2E+04
10	0.97152	2.178	1.7E+05	0.43542	0.118	4.2E+05
50	1.07217	12.243	8.7E+05	0.44145	0.721	2.1E+06

Table 4 Reactivity effects of genuine beryllium reflector (Pu-239 : 100%)

Be thickness	H/X=100			H/X=10^4		
[cm]	keff	Δk[%]	Be [g]	k_{eff}keff	Δk[%]	Be [g]
0	0.94974	0.0	0.0	0.43424	0.0	0.0
0.1	0.95564	0.591	2.7E+02	0.43464	0.040	2.6E+03
1	1.00188	5.215	3.0E+03	0.43796	0.372	2.7E+04
5	1.12463	17.490	2.1E+04	0.44801	1.377	1.5E+05
10	1.20207	25.234	6.0E+04	0.45479	2.055	3.4E+05
20	1.26522	31.549	2.2E+05	0.46039	2.615	8.9E+05

The calculated reactivity effects of beryllium in the reflector are shown in Table 3 and 4. These results show that the reactivity effects of beryllium are positive and that the values for high fissile concentration ($H/X=10^2$) are much larger than the values for low fissile concentration ($H/X=10^4$). The genuine beryllium reflector is more effective than the homogeneous reflector with water and beryllium if same amount of beryllium is used for the reflector.
According to these calculations for beryllium in the moderator or the reflector, the reactivity effects of beryllium are less than 0.4%Δk when the beryllium concentration ($Be/(Be+ H_2O)$) is less than 1 volume %.

Revision of TS-R-1

0.4 g of beryllium is approximately equal to the case of 0.007% of beryllium concentration for H/X=10 and 0.00007% of beryllium concentration for $H/X=10^4$.
As the discussion on the basis of the calculation results and the concerns about the uniformity of beryllium has been held at RPM in 2004, the revisions of subparagraph 672(a) in TS-R-1 and 672.4 in TS-G-1.1 have been approved at TRANSSC XII in 2006 as follows;

(TS-R-1/2007 Edition, unpublished)
Beryllium shall not be present in quantities exceeding 1% of the applicable consignment mass limits provided in Table 13 except where the concentration of beryllium in the material does not exceed 1 gram beryllium in any 1000 grams
Deuterium shall also not be present in quantities exceeding 1% of the applicable consignment mass limits provided in Table 13 except where deuterium occurs up to natural concentrations in hydrogen.

(TS-G-1.1/2007 Edition, unpublished)
672.4 The quantity of beryllium and deuterium are limited in a consignment that uses the fissile exceptions of para 672(a). The limits are 1% of the mass of the consignment limit for fissile nuclides as provided in Table XIII.
However, only material with an enriched concentration of deuterium are of concern, thus deuterium occurring in natural concentrations in hydrogenous materials is excluded from consideration in this limit. Calculational results provided by Japan demonstrated that beryllium will have no impact on increasing the risk for criticality when the beryllium mass is less than 0.1% of the material mass that makes up the consignment.

CONCLUSIONS

The numerical calculations are performed with a wide range of deuterium, beryllium and fissile material concentrations. These results show that the reactivity characteristics of beryllium and deuterium are similar and that their concentrations are very important. Furthermore, the reactivity effects caused by 0.4 g of deuterium or beryllium, which is the restriction for an exempted fissile package by TS-R-1 in the 1996 version, are very limited. It is thought that the restriction based on deuterium or beryllium mass alone is not appropriate. Therefore, deuterium or beryllium with very low concentration in the material can be neglected when assessing the mass of deuterium and beryllium in a consignment for an exempted fissile package.
The subparagraph 672(a) about deuterium and beryllium in TS-R-1 has been revised to the reasonable provision in the basis of these numerical evaluations. The revision makes massive LLW classify as exempted from fissile material, though LLW may contain hundred grams of deuterium or beryllium. It is confirmed that the massive LLW transport by ship is applicable for para. 672 of IAEA regulations (TS-R-1).

ACKNOWLEDGMENTS

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REFERENCES

1) Regulations for the Safe Transport of Radioactive Material, 1996 Edition, Safety Standards Series ST-1, International Atomic Energy Agency, Vienna (1996).
2) Advisory Material for the Regulations for the Safe Transport of Radioactive Material (1996 Edition), Safety Standards Series No. ST-2, International Atomic Energy Agency, Vienna (1999).
3) H. Okuno, H. Akiyama, H. Mochizuki, "Deuterium Effect of the Subcritical limit for Fissile-to-Hydrogen Ratio," J. Nucl. Sci. Technol., 40[1],p.57-60 (2003).
4) National Astronomical Observatory, " Chronological Scientific," p.626 (2003)
5) SCALE4.3: Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation for Workstation and Personal Computers, ORNLRSIC, CCC-545 (1995)
6) D.Ito, et.al., "Investigation of Criticality Effects of Deuterium and Beryllium in Packages Containing Fissile Material," J. Nucl. Sci. Technol., 44[6],p.1-6 (2007).

Appendix-1 Para. 672(a) of IAEA Transport Regulation 1996 version
(Regulations for the Safe Transport of Radioactive Material/ ST-1)

Exceptions from the requirements for packages containing fissile material

672. *Fissile material* meeting one of the provisions (a)–(d) of this paragraph is excepted from the requirement to be transported in *packages* that comply with paras 673–682 as well as the other requirements of these Regulations that apply to *fissile material*. Only one type of exception is allowed per *consignment*.

(a) A mass limit per *consignment* such that:

$$\frac{\text{mass of uranium - 235 (g)}}{\text{X}} + \frac{\text{mass of other fissile material (g)}}{\text{Y}} < 1$$

where X and Y are the mass limits defined in Table XII, provided that either:

TABLE XII. CONSIGNMENT MASS LIMITS FOR EXCEPTIONS FROM THE REQUIREMENTS FOR PACKAGES CONTAINING FISSILE MATERIAL

Fissile material	Fissile material mass (g) mixed with substances having an average hydrogen density less than or equal to water	Fissile material mass (g) mixed with substances having an average hydrogen density greater than water
Uranium-235 (X)	400	290
Other fissile material (Y)	250	180

(i) each individual *package* contains not more than 15 g of *fissile material*; for unpackaged material, this quantity limitation shall apply to the *consignment* being carried in or on the *conveyance*, or
(ii) the *fissile material* is a homogeneous hydrogenous solution or mixture where the ratio of fissile nuclides to hydrogen is less than 5% by mass, or
(iii) there is not more than 5 g of *fissile material* in any 10 litre volume of material.

Neither beryllium nor deuterium shall be present in quantities exceeding 0.1% of the fissile material mass.