Original Research Article

Criticality Study for Large Masses of Low Enriched Uranium Samples in an Active Well Neutron Coincidence Counter

ABSTRACT

Aim: A criticality study has been conducted while verifying large masses of Low Enriched Uranium (LEU) samples in an Active Well Neutron Coincidence Counter (AWCC). Fissile material mass limits were determined for some setup conditions to assure safe operation of the counter.
Place and Duration of Study: Department of Safeguards and Physical Protection, Nuclear and Radioliogical Regulatory Authority (ENRRA), between February 2015 to December 2015.
Methodology: The AWCC device was assumed to be employed in verification activities including measurements of different Nuclear Material (NM) samples in different setup configurations, forms and conditions. The MCNP5 code was used to estimate k_{eff} of relatively large masses of LEU in different forms including uranium oxide powder, compacts and fuel rods. All calculations were performed assuming the operation of the AWCC in active thermal mode at maximum capacity of its cavity (19.3 ℓ). The uranium powder samples were modeled as dry and with different values of water contents. For compacts and fuel rods, the calculations were performed with and without the existence of moderating materials in the cavity of the device.
Results: All studied cases were found to be subcritical except for a few cases of uranium oxide powder containing water. Criticality was reached for samples containing ^{235}U masses ranged between 1.5 to 8 kg with corresponding percent water content from 67 to 25. **Conclusion:** Criticality study was conducted to assure safe operation of the AWCC device while verifying large masses of LEU samples. The estimated mass limits of LEU samples with certain characteristics that could be safely verified in the device are presented.

Keywords: Criticality, Safeguards, AWCC, Uranium, Monte Carlo

1. INTRODUCTION

Inspection on nuclear facilities for safeguards purposes is one of the main functions of a nuclear regulator. Usually, the inspection activities include the performance of some measurements to verify the declared quantities of Nuclear Materials (NM). Sometimes the inspection activities should be performed while facility shutdown, which necessitates minimizing the time of inspection to avoid any delay or interruption to facility operation. The optimum goal of an inspection is to verify all NM in a relatively short time. However, in most cases, this could not be achieved due to either limitations in time or the presence of large number of items, and representative sample has to be selected. The probability of detection of diversion or inconsistency increases as the quantity or the number of items in representative sample increases [1]. The AWCC - member of the neutron coincidence family [2, 3] - can provide essential solutions for these situations. It is designed to measure the NM non-destructively. The components, operation and characteristics of the AWCC were described in many articles [4-11]. A recognized advantage of the AWCC is that it could accommodate relatively large masses or large number of items of NM. Accordingly, it can be efficiently used to achieve inspection goals with relatively higher accuracy and short time. However, the selection of large NM samples may raise the issue of criticality. Therefore, criticality checks have to be carried out to assure safe operation. To our best knowledge, criticality calculations for the AWCC were performed for High Enriched Uranium (HEU) samples [12-14]. The present study aimed to perform criticality calculations for relatively large masses of Low Enriched Uranium (LEU) samples measured in the AWCC using the general Monte Carlo Code MCNP5.

2. CALCULATIONS

Criticality calculations were performed for different NM samples in the AWCC. The samples include uranium oxide powder, NM compacts and nuclear fuel rods. All calculations were performed assuming thermal mode operation of the AWCC. Also, the maximum capacity of the counter was considered. The maximum capacity is achieved via removing the upper and lower polyethylene plugs except for modified polyethylene disks in which the interrogation sources are placed. With this setup the volume of the cavity is about 19.3 ℓ.

2.1 Modeled samples

Homogeneous dry- and moisture contained-powder samples of U_3O_8 compound were modeled. Nine density values were considered for the dry U_3O_8 samples covering a range starts from 1 up to the theoretical density (D_T) of 8.3 g/cm^3. At the maximum capacity of the AWCC cavity (19.3 ℓ) these densities correspond to a range of U_3O_8 masses between 19.29 and 160.11 kg respectively. The uranium enrichment for both powder and compact samples is about 19.77%. As a safety margin an enrichment of 20% was assumed for U_3O_8 powder samples (noted later by $U_3(20)O_8$). For NM-water mixtures eleven samples were considered. The modeled samples contain U_3O_8 masses range between 4.823 to 154.323 kg. Taking into consideration the maximum capacity of the counter (19.3 ℓ), the corresponding range of water will be between 79.51 to 0.45 percent by weight.

Compact samples are mixtures of U_3O_8 and Aluminum compressed in a cuboid form. They are used for manufacturing nuclear fuel of MTR type. Each compact has dimensions of 6.9×6.05×0.85 cm, density equal to 4.84 g/cm^3 and contains 21.3 g of ^{235}U isotope.

The EK-10 fuel rods contain LEU (10% enrichment) with a matrix material. In the present study, these rods were assumed to contain pure uranium with 11.112 g ^{235}U isotope mass content per fuel rod. The dimensions of the rod are 50 cm length and 0.7 cm diameter. The cladding material is Aluminum (0.15 cm thickness).

For compacts and fuel rods cases, the calculations were performed with and without moderation materials fill the spaces between items in the cavity of the counter. The existence of moderating material in the cavity was considered for two reasons. First, is to take into consideration the worst possibility of flooding with water. Second, polyethylene (CH_4) may be used as a moderating material especially for LEU samples that may contain relatively small masses of ^{235}U. The presence of CH_4 increases the fission rates and improves the counting statistics via increasing the fraction of thermal neutrons.

2.2 Modeling

Calculation of k_{eff} for all configurations was performed using the general Monte Carlo Code MCNP5. “KCODE” card was used to run criticality problems with “KSRC” card to locate the initial spatial distribution of fission points. Initial fission source points were located in every cell containing fissionable material. A nominal number of source histories was selected as 5000 per cycle. The initial guess of k_{eff} was determined according to each problem. Fifty source cycles were skipped before k_{eff} accumulation, while 250 active cycled were considered. The estimated relative standard deviations were always below 0.25% for all calculations.

The “LIKE n BUT” feature was used to create repeated structure of ^{3}He tubes in the counter and that for compacts, while “U” (universe), “FILL” and “LAT=2” (lattice, hexagonal prism) cards were used to create the repeated structure of fuel rods.

Fig.1 illustrates the longitudinal (i) and cross (ii) section calculational model geometries for the problem of $U_3(20)O_8$ powder as drawn by MCNP5 visual editor. The cavity of the counter is completely filled with NM.

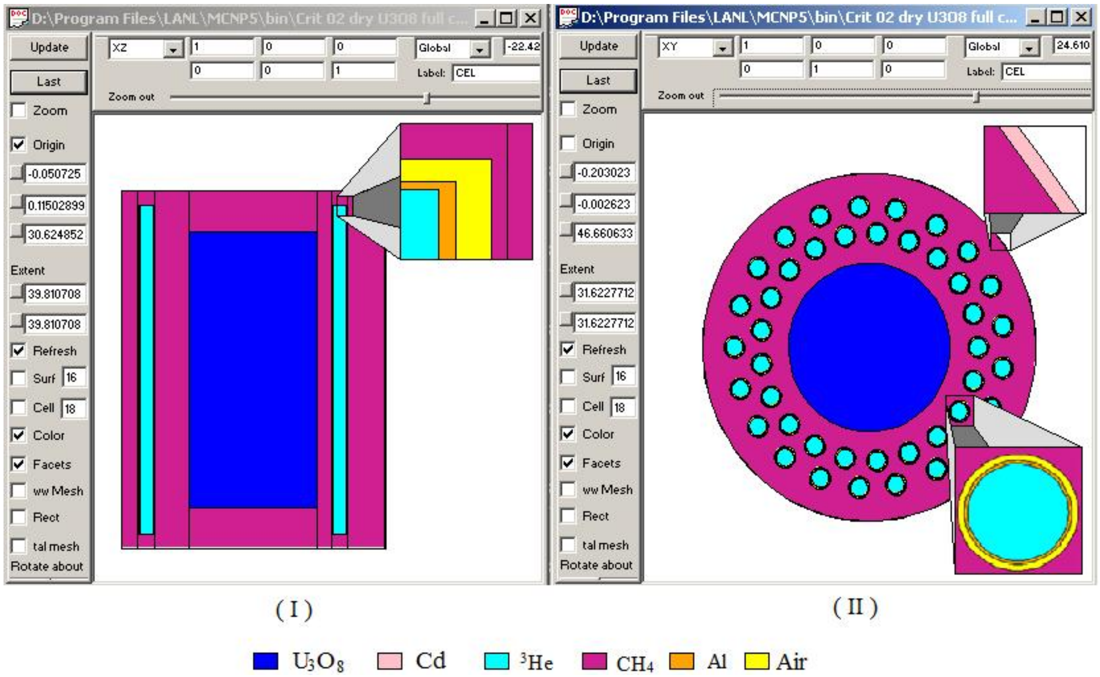


Fig. 1. MCNP longitudinal (I) and cross (II) section Calculational model geometries for U_3O_8 powder samples.

Two configurations for the compacts in the counter were modeled. In the first one the compacts were staked to approximately fill the cavity of the counter with a total uranium mass of 31.2 kg. In the second configuration 125 compacts with a total uranium mass of 13.515 kg were regularly distributed in the cavity with spacing in between as shown in Fig. 2. In both cases the rest volume of the cavity was assumed to be filled with a moderating material.

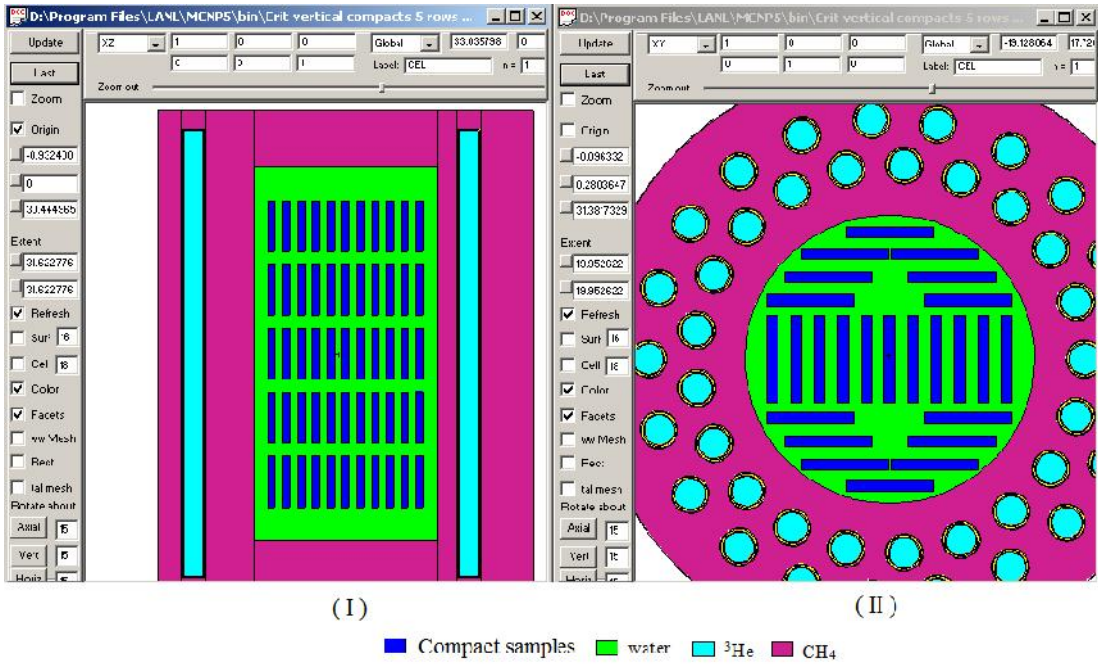


Fig.2. MCNP longitudinal (I) and cross (II) section Calculational model geometries for regularly distributed compact materials.

For fuel rods many configurations were modeled including different number of fuel rods (^{235}U-mass), regular and irregular distributions. Twenty seven cases were modeled in the present work as presented in Table 1. Each case is identified by two characters; the first is a letter indicating the moderating material while the second is a numeric indicating the number of fuel rods arranged at a certain configuration as illustrated in Fig 3. The figure illustrates only the nine cases without moderating materials.

NR* → CM+ ↓	73	91	100	113	127	150	188	200	361
None	N1	N2	N3	N4	N5	N6	N7	N8	N9
Water	W1	W2	W3	W4	W5	W6	W7	W8	W9
CH_4	P1	P2	P3	P4	P5	P6	P7	P8	P9
^{235}U mass (g)	811	1011.2	1111.2	1255.7	1411.2	1666.8	2089	2222.4	4011.4

Table 1. Codes for different cases of criticality calculations of fuel rods in the AWCC

**NR: Number of Fuel Rods distributed as illustrated in Fig. 3*
+CM: Moderating material in the cavity

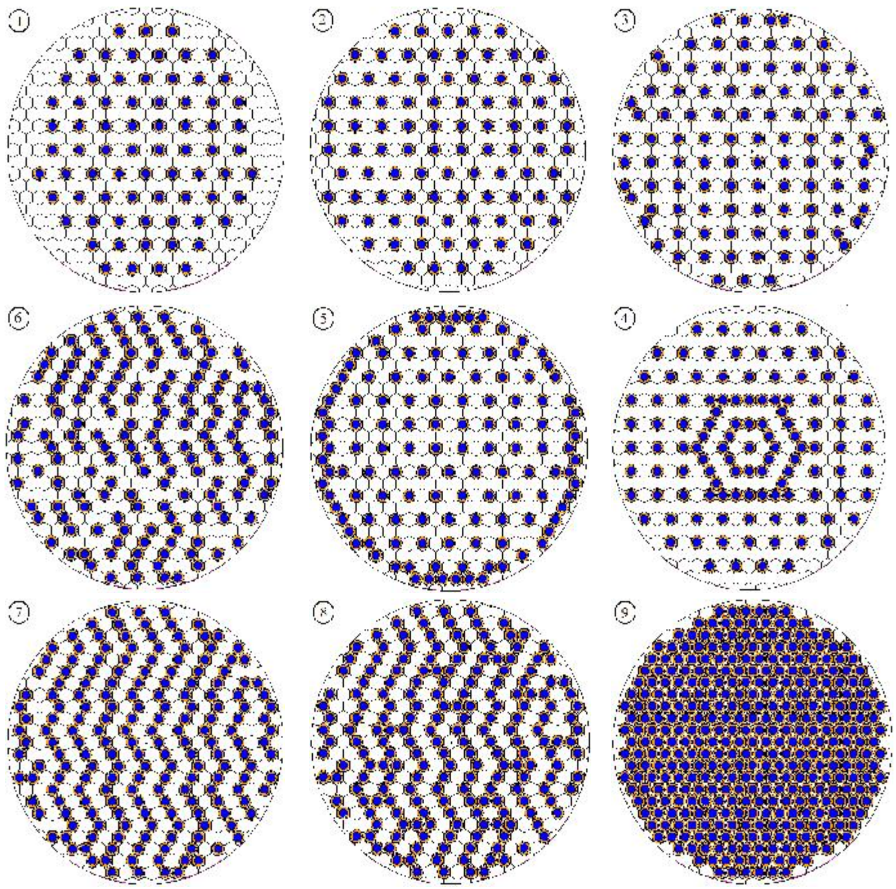


Fig. 3. Nine configurations for fuel rods distributions.

Fig. 4 shows the MCNP model for case N5 as a selected configuration for irregular distribution of 127 fuel rods without moderating material. A regularly distributed fuel rods configuration model (case W1), in which the moderating material is water, is shown in Fig. 5.

Another selected configuration (case P9) is shown in Fig. 6. It indicates that the number of fuel rods is 361 (the maximum capacity of the counter) with polyethylene moderating material.

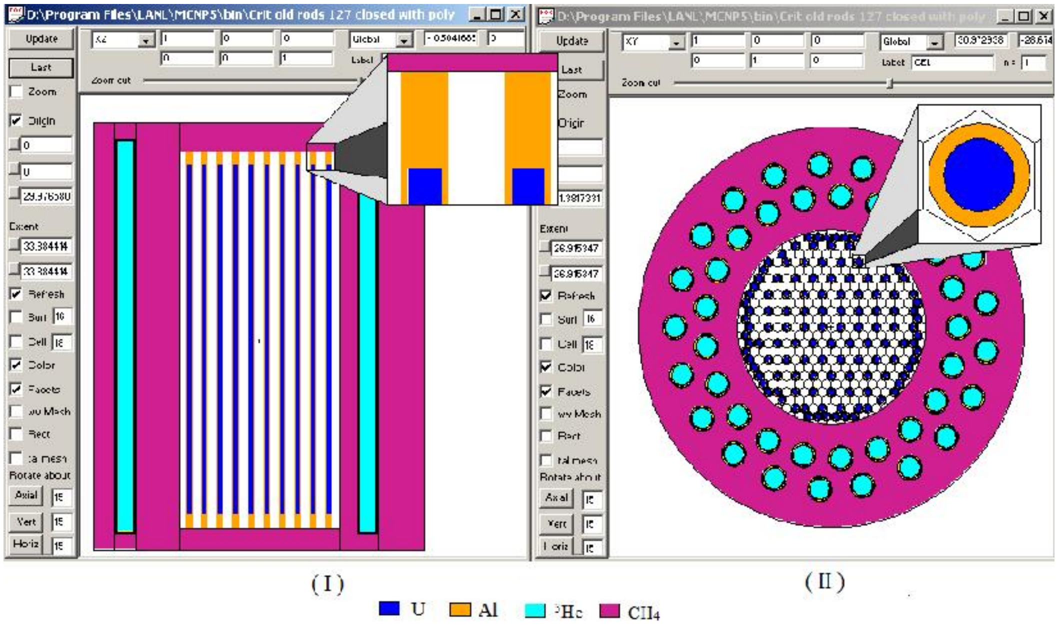


Fig. 4. MCNP longitudinal (I) and cross (II) section Calculational model geometries for irregular distribution of fuel rods without moderating material (case N5).

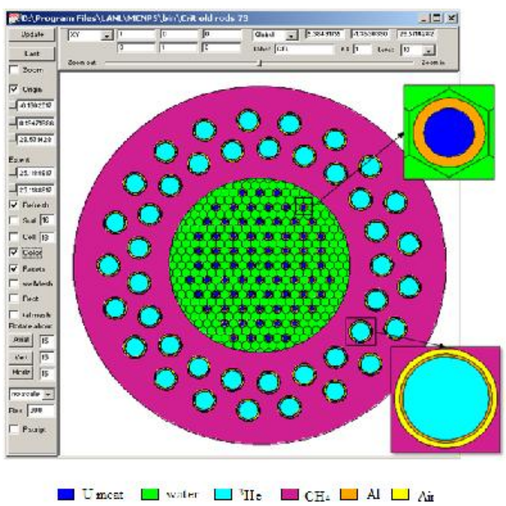


Fig. 5. MCNP model for regularly distributed fuel rods with water as moderating material (case W1).

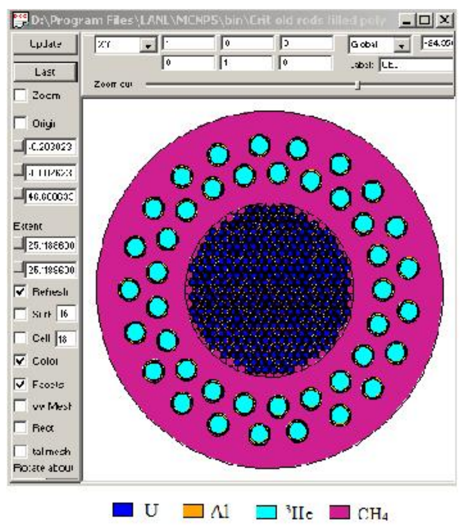


Fig. 6. MCNP model for case P9, full capacity of cavity with polyethylene moderating material.

4. RESULTS AND DISCUSSION

Table (2) presents the calculated k_{eff} values for LEU dry powder samples. All samples are far subcritical even for the maximum weight at the U_3O_8 theoretical density. As uranium mass (density) increases more fissions are expected to take place due to increase in interaction cross section [3]. A direct proportionality is noticed between U_3O_8 mass and k_{eff}, however it is not linear. This is due to the reason that the produced fission neutrons will directly interact with uranium nuclide without having enough chance for being thermalized. This is clear from the last three columns in the Table (2) which gives the percent of fission neutrons caused by thermal (*Th, <0.625 eV*), intermediate (*Int, 0.625 eV-100 keV*) and fast (*F, >100 keV*) neutrons as calculated and given by the MCNP5 Code. Consequently the rate of increase of calculated k_{eff} values will decrease as uranium mass increases. The maximum calculated k_{eff} value (0.65) is limited by the maximum U_3O_8 density (D_T) and the size of the AWCC cavity.

Table 2. Values of k_{eff} for dry U_3O_8(20) powder with varying mass

U_3O_8 (kg)	^{235}U (g)	Sample density (g/cm^3)	k_{eff}	Percent of fissions caused by neutrons		
				Th	*Int*	*F*
19.290	3272	1	0.36	76	15	9
38.581	6543	2	0.42	65	21	14
57.871	9815	3	0.46	57	24	19
77.162	13087	4	0.50	52	25	23
96.452	16358	5	0.54	47	26	27
115.742	19630	6	0.57	44	26	30
135.032	22902	7	0.61	41	26	33
154.323	26173	8	0.64	38	27	35
160.110	27154	8.3	0.65	37	27	36

The obtained values at half (D_h) and full (D_f) theoretical density are comparable with those obtained by Miller and Yearwood [14], although some differences exist in calculational conditions. The differences include material compositions, values of D_h and D_f and ^{235}U enrichment. Table 3 presents some selected k_{eff} values obtained by Miller and Yearwood in comparison with those obtained in this study with specific differences in calculational conditions.

Table 3. Comparison of some keff selected values obtained in this work by those obtained by Miller and Yearwood with specific differences in calculational conditions.

	NM	^{235}U Enr	D_T (g/cm^3)	k_{eff} at D_h	k_{eff} at D_f	k_{eff}	
						5 kg, ^{235}U	*10 kg, ^{235}U*
Miller & Yearwood	UO_2	93%	11	Ranges between **0.44 - 0.55** for ^{235}U masses between 5 and 10 kg	**0.50 - 0.63**	**0.44**	**o.63**
This work	U_3O_8	20%	8.3	**0.52** ~ 14.5 kg ^{235}U mass	**0.64** ~ 27.2 kg ^{235}U mass	**0.39** ρ~1.5 g/cm^3	**0.46** ρ~3 g/cm^3

In Table (4), the values of calculated k_{eff} are given on a range of homogeneous mixtures of water and U_3O_8 powder. As the fraction of water content increases (increase in Hydrogen content "H" as indicated in the last column of the Table), more thermal neutrons becomes available to induce more fissions. Samples remain subcritical till water content reaches about 25% by weight in the sample. Then, the sample becomes and remains critical as the water content increases till about 65% by sample weight. Then as the water content is increased more, the effect due to decrease in the mass of fissile material predominates and the system becomes again subcritical. The range at which the samples becomes critical, as given in Table (4), is between about 1.5 and 8 kg of ^{235}U mass which corresponds to about 65 and 25% water contents, respectively. The trend of increasing and then decreasing of k_{eff} as water content in the sample increase is in consistent with that obtained in other literature [14].

Table 4. Values of k_{eff} for U_3O_8(20) powder with varying mass and water content.

U_3O_8 (kg)	^{235}U (g)	Weight percent Water in sample	Sample density (g/cm^3)	k_{eff}	Percent of fissions caused by neutrons			H-content (%)
					Th	*Int*	*F*	
154.323	26173	0.45	8.036	0.67	37	31	32	0.05
135.032	22902	2.19	7.157	0.72	37	39	24	0.24
115.742	19630	4.41	6.277	0.77	41	42	17	0.49
96.452	16358	7.37	5.398	0.83	48	39	13	0.82
77.162	13087	11.47	4.518	0.88	57	34	9	1.27
57.871	9815	17.55	3.639	0.94	67	27	6	1.95
48.226	8179	21.85	3.199	0.97	73	22	5	2.43
38.581	6543	27.51	2.759	1.00	78	18	4	3.06
19.290	3272	46.79	1.880	1.03	88	10	2	5.20
9.645	1636	65.27	1.440	0.99	94	5	1	7.25
4.823	818	79.51	1.220	0.87	96.5	3	0.5	8.83

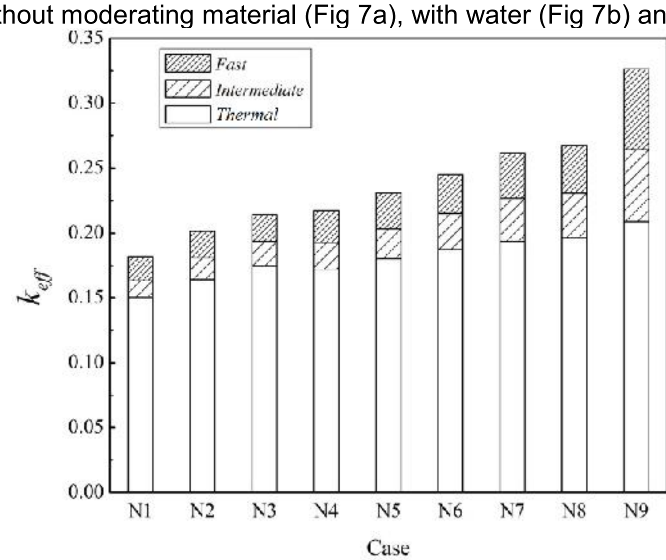
As reflected in Table 5, calculations for compact NM indicate fairly safe values for staked compact samples even for full capacity of counter and moderation materials. However, caution should be considered for distributed samples with polyethylene moderating material (last raw in the Table 5) which correspond to a number of samples of 125 (about 2.7 kg of ^{235}U).

Table 5. Values of k_{eff} for U-compacts in different configurations and cavity moderators.

Case	^{235}U mass (g)	Cavity moderator	k_{eff}	Percent of fissions caused by neutrons		
				Th	*Int*	*F*
		None	0.39	61	19	20
1	6147	Water	0.51	63	21	16
		Poly	0.53	64	21	15
		None	0.35	76	14	10
2	2662.5	Water	0.85	85	12	3
		Poly	0.94	87	10	3

Fig 7(a, b and c) shows the results of calculations for fuel rod cases. The values of keff are drawn for device cavity without moderating material (Fig 7a), with water (Fig 7b) and with polyethylene (Fig 7c).

(a)



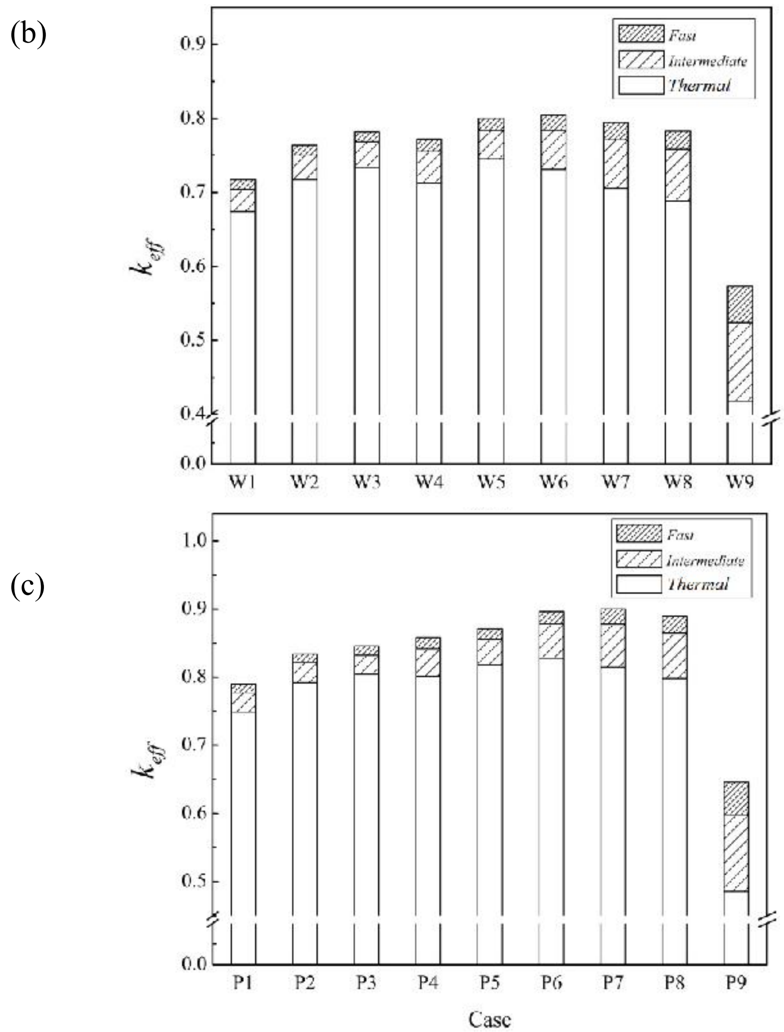


Fig. 7. ke_{ff} values for different fuel rods cases, (a) without-, with (b) water and (c) polyethylene moderating materials.

The Figure shows also the contribution of each range of neutron energy. All fuel rods studied cases were found to be subcritical. The maximum k_{eff} values were obtained for the cases P6, P7 and P8 at which k_{eff} approaches 0.9. This corresponds to ^{235}U masses between 1.7 and 2.2 kg. However all cases are still subcritical. The trend of keff increase and then decrease as the mass of fissile material increases (in cases of moderating material exist) is described before. Maximum k_{eff} values were obtained for the case of using polyethylene as moderating material as long as, it contains more hydrogen atoms in a given volume than any other substance [17].

5. CONCLUSION

A criticality study has been conducted for the measurement of LEU samples using the AWCC. Fifty three cases were studied including dry and water contained powder samples, compact samples and fuel rods. All samples under this study could be safely measured in the AWCC with and without moderating materials in the cavity of the counter. The only exception was found for some NM-water mixtures contained U_3O_8 powder. For these samples the system becomes critical for masses ranges between 1.5 and 8 kg of ^{235}U with weight percent water in samples between 67 and 25.

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