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THE EFFECTS OF STORAGE PATTERNS ON THE NEUTRON MULTIPLICATION FACTOR OF SPENT NUCLEAR FUEL CASKS

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Abstract

The type of an array selected for the storage of fissile material such as fuel assemblies or spent fuel casks has a profound effect on the neutron multiplication factor (keff) of the system. Given a particular initial enrichment, the neutron multiplication factor of the system can either increase or decrease depending on either the type of the array used (which may either be an *n* x *n*, or *n* x *m* where *nm*), the distance among various fissile units or the total number of units. This paper will present the effects of changes in distances between spent fuel casks in various storage arrays using fresh fuel assumptions.

Keywords

burnup credit, actinides, effect of separation gap between cask in criticality, spent fuel casks, neutron importance

1. Introduction

Many nuclear reactors that went operational about forty years ago will soon run out of storage space in their spent fuel pool, if no alternative storage has been developed [1, 2]. One of the alternatives that may be a solution to the problem is the spent fuel casks- albeit a short-term solution. This in itself comes with its own of storage capacity problems of Interim or Long Term Storage Facility not being big enough to accommodate all the casks.

In a case where storage space is limited, it is a common practice to reduce the gap between the casks as much as possible with a view of increasing the capacity of the Storage Facility. However, Mayne, Dowson and Abbey have cautioned against this practice warning that the distance between fissile units, such as spent fuel casks and fuel assemblies can only be reduced up to a certain point without running the risk of the system being critical, beyond that, this risk increases quite significantly [3, 4, 5]. This depends on the choice of the Storage Array whether: (a) the chosen Storage Array allows for neutrons from one unit to interact with units of other Arrays [3, 4, 6], (b) the critical mass of the fissile material is concentrated in one small area or is it dispersed over a wider area [3, 4, 6]; (c) the amount of hydrogen atoms available for neutron thermalisation; (d) there is adequate ventilation among different units of the arrays. If the selection has not given consideration to these factors there is a potential risk that the system can be critical depending on the initial enrichment of the fuel and power history the fuel assemblies [5, 6]. The application of burnup credit (BUC) to supplement the selected Storage Array in further increasing the capacity of the Storage Facility has been implemented in a number of countries, but only a small set of nuclides have been accepted by their respective regulators. Even then, different levels of approval have been granted as follows; 1) actinides and fission products: have only been approved for spent fuel pools (SFP) only; 2) actinides only have been approved for spent fuel casks.

Hence this study will only focus on major actinides only for the same reason just stated.

2. Methodology

The overall goal of this project was to determine the effect the type of storage array has in the criticality of the spent fuel storage as well as the effect of changing one of the parameters. The simulation was performed using KENO-VI and

STARBUC two modules of the Standardized Computer Analyses for Licensing Evaluation (SCALE) code [7, 8, 9]. The project was divided into two main categories; Fresh Fuel and Spent Fuel. Each category was further divided into two groups; thirty casks and four casks. For thirty casks this was divided into 2X15 and 3X10 Arrays while the four casks were divided into 1X4 (linear) and 2X2 (square) Array. All the casks under study are kept in a 60 m × 23.5 m × 8.97 m building with a 48.5 cm thick concrete slab. Since the radius of the casks is 144.3 cm, to separate two casks by 100 cm the 100 cm is added to the radii of the two casks in question and the resulting pitch becomes 288.6 cm + 100 cm = 388.6 cm or simply 100 cm surface-to-surface.

2.1 Fresh fuel

2.1.1 Four casks

2.1.1.1 Linear (1X4) array storage

To ensure that the casks were resting on the floor their origins were shifted to z = -180.2 taking into consideration the 48.5 cm thick concrete slab, making $+z_{slab}$ = - 400 cm and $-z_{slab}$ = - 448.5. The initial and subsequent x,y,z co-ordinates of the casks were as summarized in the grid indicated in Table 1, taking into account that gap between adjacent casks were increased by 50 cm in every subsequent simulation. The distance among the adjacent casks were only increased along the x-axis and the y-axis was kept constant

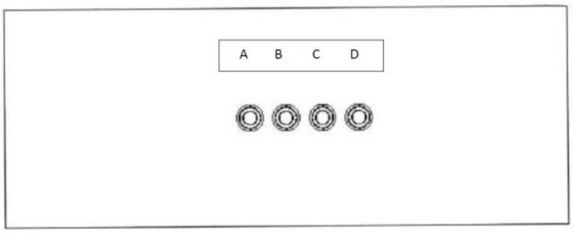

Figure 1: Top view of cask storage room with casks positioned vertically

The pictorial top view of the four casks in 1X4 Array in the storage building is as shown in Figure 1. These simulations were run in preparation for the comparison of the effect of k_{eff} from 1X4 array versus that of 2X2. The simulations will be discussed in detail in the next section.

2.1.1.2 2X2 storage array

In this part of the project, the four casks were arranged in a square matrix and in every subsequent run, the casks were moved apart by 50 cm, and the results of the k_{eff} compared to those of the linear array. The top view of the four casks in the storage building is shown in Figure 2.

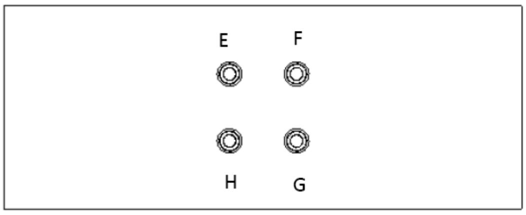

Figure 2: Top view of four casks in a 2X2 Matrix

The general trend of the graph in Figure 3 suggests that there is a slight increase in the k_{eff} of the 2X2 as the distance between adjacent casks increases, whereas there is a more rapid decrease in k_{eff} for 1X4 over the same distance.

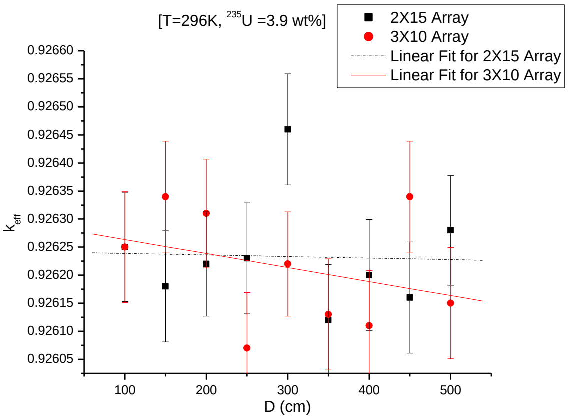

Figure 3: Comparison of reactivity between a 1X4 Array and 2X2 Array.

The reason for the difference in criticality of the two arrays are largely due to the amount of shielding seen by the neutrons in each array, coupling among the casks, back-scattering of neutrons from air, floor surface and from the surfaces of the casks, energy of the neutrons, neutron importance and end-effect due to uneven distribution of neutron flux in the core. These factors will be discussed individually in the next section as to how they contribute to the behavior of graph.

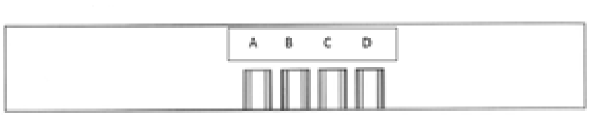

Figure 4: Front view of vertically positioned casks

Table 1: Co-ordinates of the 4 casks in their storage building

D (cm)	100		150		200		250		300		350		400		450		500	
x-y co-ordinates	x	y	x	y	x	y	x	y	x	y	x	y	x	y	x	y	x	y
	-388	0	-438	0	-488	0	-538	0	-588	0	-638	0	-688	0	-738	0	-788	0
	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	388	0	438	0	488	0	538	0	588	0	638	0	688	0	738	0	788	0
	776	0	876	0	976	0	1076	0	1176	0	1276	0	1376	0	1476	0	1576	0

Table 2: Measurement of neutron spectrum around Castor cask [16]

Distance of BSS from cask (m)	Height above the floor surface	Angular position (degrees)	φ_E E ($cm^{-2}min^{-1}$)	Energy range (MeV) of the peak (3 Group Theory)	
1	1.95	2.70	360	1.6E-2	fast
10	0.86	180	31.5	1.8E-8	thermal
1	3.92	270	370	1.0E-1	fast
top centre of cask	0.47 above the cask lid	not specified	285	1.0E-1	fast

Table 3: Range of energy spectrum

Designation of Energy Group	3 Group Theory	4 Group Theory
Thermal Energy	0.02 eV to 1 eV	0.02 eV to 0.625 eV
Epithermal	1 eV to 10 keV	0.625 eV to 5.5 keV
		5.5keV - 0.86 MeV
Fast Neutrons	10 keV - 10 MeV	0.86 MeV - 10 MeV

a) Self-shielding

Ignoring all back-scattered neutrons, a neutron in Figure 4 leaving cask A for cask D (and vice versa) in a 1X4 array must first pass through B and C without being absorbed before it can reach D and cause fission. Thus the two middle casks form a shield between A and D by virtue of:

- Their spatial position;
- Thickness of the material, especially concrete and steel;
- Distance between adjacent casks for which the intensity of neutron flux from either cask should follow the inverse square law;
- The magnitude of the attenuation coefficients of shielding materials (notably concrete and Fe);
- Material density of the shielding material.

These factors play a very significant role in preventing any neutron from A to reach D and thus causing fission [10, 11]. In a 2X2 array (Figure 2) on the other hand, except for the increase in distance none of the casks is shielded by any of them as was the case in Figure 4. Not only does neutron originating from E have an equal chance of reaching F, G or H without the risk of being absorbed, but also because the some of the cask materials consists of light elements, such as hydrogen which have a very high lethargy gain ξ, which corresponds to average logarithmic energy loss of a neutron in a collision of nucleus of a given mass. Thus a neutron undergoes a significant amount of moderation as a result of inelastic scattering by hydrogen atoms in 2X2 array compared to the 1X4 array which will subsequently lead to an increase in k_{eff}. Thus everything being the same, a neutron in Figure 2 has greater chance of causing fission than the same neutron in Figure 4, which is why the k_{eff} of 1X4 Array in Figure 3 is generally lower than that of a 2X2 Array.

In addition to spatial shielding, one has also has to take into consideration Energy Self-shielding which is as important as spatial shielding. According to available literature, if a nuclide has a large narrow resonance, the macroscopic cross section at the energy where resonance occurs will increase sharply over the range of resonance causing the neutron flux to decrease drastically, resulting in a flux depression over the region of the resonance compared to where there is no resonance. The flux will subsequently return to its original level at energies just below the resonance [12]. This implies that only those neutrons that are scattered down into the energy range of the resonance will be absorbed, but those which are scattered either above or below the resonance energies will escape resonance capture. To illustrate this consider ^{238}U with the resonance energy between 1×10^{-5} MeV and 1 MeV. Any neutron that falls between these two points will be absorbed and result in a flux depression in that region [13].

b) End-effect

It is a well-known fact that the burnup in a reactor core is not uniform whether one looks at it in radial or axial distribution. Radial distribution does not have as big an influence in criticality as axial distribution does. Looking at the fuel assembly along the axial direction, it is observed that the central region, (between about 50 cm and 250 cm) has a higher burnup than those at the extreme ends, of the two extreme ends, the top-end has an even lower burnup compared to the bottom-end as indicated in Figure 5 [10, 12, 14]. This results in a phenomenon known as end-effect where the underburned regions have a higher concentration of fissile material than in the central region, which can potential be a safety risk if not accounted for in spent fuel storage [15].

When looking at Figure 3, it is noted that there is a peak at 150 cm for 2X2 Array and which is not as prominent in 1X4. This is due to the end-effect which resulted in concentration of under reacted fissile material in the ends of the fuel assembly, which become apparent in the increase in fission when casks are stacked closer to one another leading to an increase in k_{eff} noted as in Figure 5. The peak is much higher in 2X2 because they do not shield one another as they do in a 1X4 array and also because of the an increase in logarithmic energy decrement in 2X2 due to an increase in the hydrogen atoms seen by the neutron compared to those of 1X4 array.

c) Back-scattering

According to neutron measurement performed by Buchillier around Castor HAW 20/28 cask, it has been found that neutrons can be detected as far as 10 m from their source, and that the energy distribution of neutrons around the casks do not follow a simple linear relationship with the distance from the casks; it fluctuated as shown in column 5 of Table 2 [16]. Butchillier ascribed this to back-scattering from wall, air, floor surface and from other casks in the same building, [17-19].

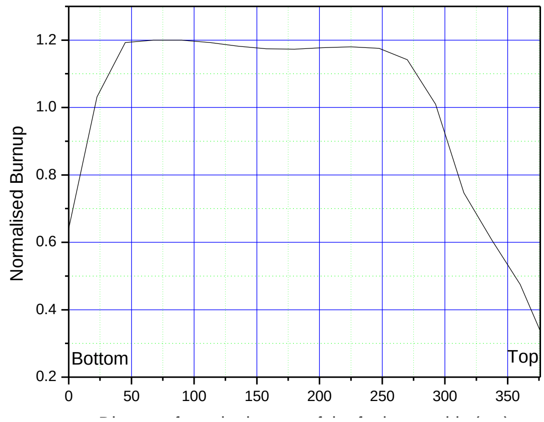

Figure 5: Variation of burnup with height of the fuel assembly

This has further been confirmed by Rimpler and Kralik who reported detecting neutrons as far as 20 m from their original source, not only confirming what was reported earlier by Butchillier but also discovering new information; that how far neutrons can go from their cask depends on the design of the cask and the material out of which they are made. In addition to that, Rimpler also noted that the energy spectrum of the neutrons became harder the further away they moved from the source [7, 8]. This finding seems to support the decrease in k_{eff} noted in 1X4 and also in 2X15 and 3X10 arrays below. The only reason why the 2X2 array does not follow the same trend is because of absence of a shield between the casks.

Depending on whether one applies the three- or four-group theory, the detected neutrons can be regarded as thermal, epithermal or even fast neutrons. In this project a three group theory will be used, and the upper and lower limits of 3-and 4 energy groups are indicated in Table 3 [10, 11].

From this it is evident that not only can neutrons travel that far away from their source, they can also be in the epithermal or thermal range making it quite likely that they can induce fission even though they are that far away from the source if they come into contact with fissile or even fertile material. Hence, even though the neutrons may be in the fast energy range, depending on the array chosen they may potentially induce criticality if the array is such that moderating power is high, and ξ is logarithmic energy decrement and s is the macroscopic scattering cross section. This appears to be the case in the 2X2 array.

The observed neutrons are due spontaneous fission of ^{244}Cm and (α,n) reactions from ^{241}Am, ^{16}O and ^{10}B. The nuclear reactions by which they were generated are [18, 20- 22]:

1) $^{244}_{96}Cm + \alpha \rightarrow {}^{247}_{98}Cf + {}^{1}_{0}n + Q$

2) Spontaneous Fission of ^{244}Cm.

3) $^{16}_{8}O + \alpha \rightarrow \left[{}^{20}_{10}Ne\right]^{*} \rightarrow {}^{19}_{10}Ne + {}^{1}_{0}n$

4) $^{10}_{5}B + \alpha \rightarrow [{}^{13}_{7}N]^{*} + {}^{1}_{0}n \rightarrow {}^{14}_{6}C + {}^{0}_{+1}e$

5) $^{241}_{95}Am + \alpha \rightarrow {}^{244}_{97}Bk + {}^{1}_{0}n \rightarrow {}^{245}_{96}Cm + {}^{0}_{+1}e$

The fact that the neutrons can travel that far combined with the fact that their energy spectrum can fluctuate from fast to thermal energy range is one of the reasons why there is fluctuation in neutron multiplication factor as the distance between the casks is increased from 100 cm to 500 cm.

d) Statistical uncertainties

Statistical uncertainty associated with probabilistic calculations may be due to uncertainty in geometry of the array or any of its subcomponents such as the manufacturing tolerance of diameter of the fuel rod or the density of a certain fuel material. It can also arise due to uncertainty from modeling caused by over simplification of results, e.g. rounding-off, which subsequently contributes uncertainty to the k_{eff} of the system. If statistical uncertainties are not taken into account during the calculations and added to the calculated k_{eff} as a bias (correction) to obtain the benchmark model k_{eff}, the analyst will obtain incorrect results and any trend based on these will also be incorrect because of the error. In this respect, the statistical uncertainties tend to hide the real change between results of two very similar configurations such the effect of random variation of distances among various casks on the k_{eff} and result in incorrect values being reported.

e) Neutron importance

The concept Neutron Importance was first introduced in reactor physics by Soodak, in order to better described and understand this concept, one has to study the behavior of a neutron at position **x** at the time *t* and its progeny at some time later. The progeny in this context refers to all those neutrons that trace their origin back to the original neutron through fission, scattering, or even after a change in position of the original neutron**.** This implies that the number of progeny can either be more or less than the number of original particles depending on whether the system is multiplying (fission and scattering) or dividing (absorption or capture). To take into account the possibility that the progeny can potentially be more or less than their parent neutrons, one has to speak about the *probable* number of progeny, which will allow the progeny represented to be less or equal to one [23].

Notwithstanding lack of unified 'meaning' of the 'Importance' concept, in 1965 Jeffrey Lewins [23] defined neutron importance as follows: "*The importance N^{+}(**x**,t), is defined as the expected or probable contribution of one particle at **x** at time t to the meter reading at time t_f. Thus a particle is "important" to the (future) observable reading."*

From this it can be inferred that the importance no longer solely depend on the detector position at the time the meter is read but rather on the combination of detectors at time t_f and the progeny which will have diffused outwards from **x** in the interval from *t* to t_f. The distribution of detectors around the source will therefore play a significant role in defining the final boundary conditions.

Thus the importance of a neutron in a system such as square array of casks depend on its location **x** in that system; "neutrons near the outer surface have a higher probability of leaking out of the system without leaving any progeny behind and therefore without affecting the meter reading. Hence the neutron importance will be lower near the outer surface than those near the centre of the array" [23].

Also, if as a result of fission the neutron density of the system is increasing with *time*, the progeny of the first neutron to be released will always be higher in number than the progeny of the second generation particle. The earlier neutron therefore will have a greater effect on the meter reading than its progeny; which implies that the neutron importance decreases with time.

If an independent neutron source S is taken into consideration, it has been established that in a linear system of sources, the behavior of neutrons and its progeny will not be affected by other neutrons or the independent sources of neutrons thus the importance equation is independent of the source S. In non-linear systems however, this equation must be modified since the systems properties (fission, scattering, absorption) and therefore the way in which progeny will propagate in the system will be influenced by the density and therefore indirectly by the source [23].

In the context of spent fuel casks stored in different geometrical arrangements, in the first instance arranged in a linear array in such a way that Cask A is a source of neutrons to Cask B which then acts as a detector of neutrons from A and vice versa; in either case they have a certain sensitivity associated with certain reaction or energy range. The above section implies that if the casks are in linear array the behavior (fission, scattering etc.) of the neutrons and its progeny will not be affected by other neutrons or the independent source of neutrons S because of the self-shielding effect. However, in non-linear system such as in a 2X2 Array the independent source has an effect on the behavior of neutrons and its progeny, therefore this equation must be modified since the system properties (fission, absorption, scattering) and therefore the way in which the progeny propagates will be influenced by the density and therefore indirectly by the source. This is the fundamental difference between the results obtained in 1X4 array vz those of 2X2 Array.

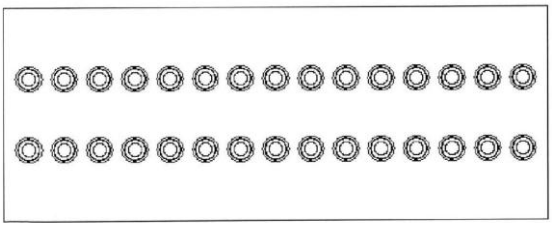

Figure 6: Top view of 2X15 Casks

3.0 Thirty (30) casks: 2x15 vz 3x10 storage array

3.1 2X15 array

This study was originally aimed at finding the least reactive storage matrix of 33 casks, but because the cask storage building was not big enough to accommodate so many casks, the model was reduced to 30 casks. In the first scenario, the casks were arranged in 2X15 array, because of the limitations of the size of the building, the distance between the casks was only changed along the y-axis only. The grid xy-co-ordinates of the locations of the casks in the cask storage building are summarized in Table 4 and the diagrammatic representations of the casks are shown in Figure 6.

3.2 3X10 array

In the second scenario the same casks were now arranged in 3X10 array and as in the previous scenario, the initial distance between middle row and the outer two rows was 100 cm and in every subsequent run the gap was increased by 50 cm along the y-axis and the k_{eff} noted. Figure 7 below shows a top view of the casks in a 3X10 array.

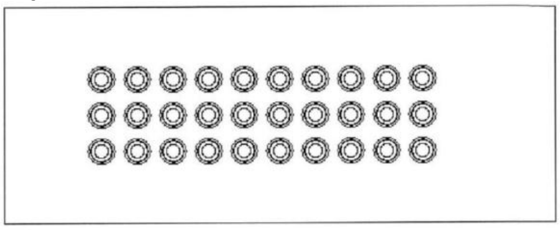

Figure 7: Topview of a Cask Storage Building with 30 casks in a 3X10 Array.

The neutron multiplication factor vz separation gap of both storage Arrays (2X15 vz 3X10) are plotted in Figure 8 for comparison. It is observed that there is decrease in k_{eff} in both arrays with an increase in distance; however the decrease in2X15 is much slower than in 3X10. This can be accounted to the fact a 3X10 has a middle row as acting as a shield between the outer two rows, which is the reason why there is a much more rapid decrease in k_{eff} with distance compared to the 2X15 array.

Table 4. Co-ordinates of a 2X15 Storage Matrix

Y-axis								438.6							
	28	24	20	16	12	8	4	388.6	1	5	9	13	17	21	25
	-2720.2	-2331.6	-1943	-1554.4	-1165.8	-777.2	-388.6	0	388.6	777.2	1165.8	1554.4	1943	2331.6	2720.2
	27	23	19	15	11	7	3	-388.6	2	6	10	14	18	22	26
								-438.6							
								X-Axis							

Table 5. Major Actinides together with their Isotopic Correction Factors (ICF) used in this project.

Nuclide	ICF	$T_{1/2}$	σ_a	σ_γ	σ_f
			barns		
U-234	0.635	2.46×10^5 y	103.47	99.8	0.465
U-235	1.085	7.038×10^8 y	687.0	98.8	587
U-238	0.992	4.468×10^9 y	2.75	2.73	2.7×10^{-6}
Pu-238	0.856	9×10 y		540	17.9
Pu-239	1.076	2.411×10^4 y	1020	269.3	748.1
Pu-240	0.945	6.564×10^3		289.5	0.064
Pu-241	1.087	14.290 y	1378.0	362.1	1011.1
Pu-242	0.848	3.733×10^5 y	18.5	18.5	0.002
Am-241	0.609	432.2 y	587	587 533[1)] 54[2)]	3.24

1) $\sigma_{\gamma(g)}$= cross section leading to ground state;
2) $\sigma_{\gamma(m)}$ = cross section leading to metastable state of a product.

4.0 Spent fuel

Taking credits for burnup: major actinides

There are many different types of nuclides produced during the depletion of the fuel in the reactor referred as different levels of BUC, but only a few of them are important to criticality [24]. They are often into three sets: ***Major Actinides only, Actinides + Minor Fission Products, Actinides + Principal Fission Products***. In this project will only Major Actinides will be considered and are listed in Table 5 together

with their Isotopic Correction Factors etc. Burnup credits in this context means taking credit for the buildup of Fission Products such as ^{239}Pu, ^{155}Gd and ^{149}Sm which have a significantly high cross section and as a result can absorb neutrons from the system, permanently removing them from the system thus resulting in a decrease in neutron multiplication factor [24-27].

The results of the analysis plotted in Figure 9 show that irrespective of the type of the array chosen, the k_{eff} is higher at lower burnup than is the case at higher burnup. This is due to the fact that fission product yield is directly related to burnup (Eq 1); the higher the burnup is, the more fission products are produced, which will subsequently lead to a higher absorption of neutrons and the corresponding decrease in k_{eff} as shown in Figure 9 [35, 36, 39]:

$$y_i^n = \frac{\sum_{g=1}^{G}\sum_{m} y_{i,g}^{n,m} N_m \sigma_{f,g,m}\phi_g}{\sum_{g=1}^{G}\sum_{m} N_m \sigma_{f,g,m}\phi_g}$$

$$y_c^n = \frac{\sum_{g=1}^{G}\sum_{m} y_{c,g}^{n,m} N_m \sigma_{f,g,m}\phi_g}{\sum_{g=1}^{G}\sum_{m} N_m \sigma_{f,g,m}\phi_g}$$

where:

y_i^n = independent fission yield of nuclide n;

y_c^n = cumulative yield of nuclide *n* for energy group g;

N_m = atomic density of fissile nuclide *m;*

$\sigma_{f,g,m}$ = microscopic fission cross section of nuclide *m* for energy g;

ϕ_g = neutron flux at energy g.

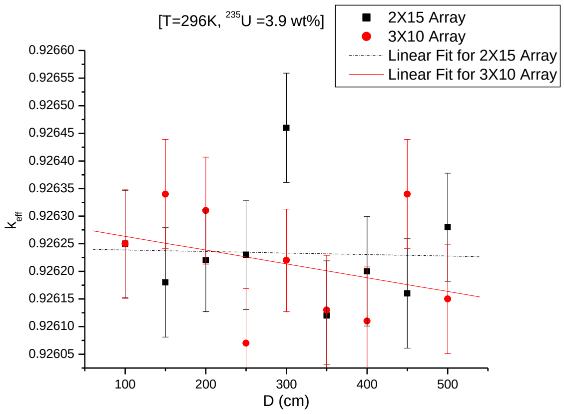

Figure 8: 2X15 vz 3X10 Storage Array

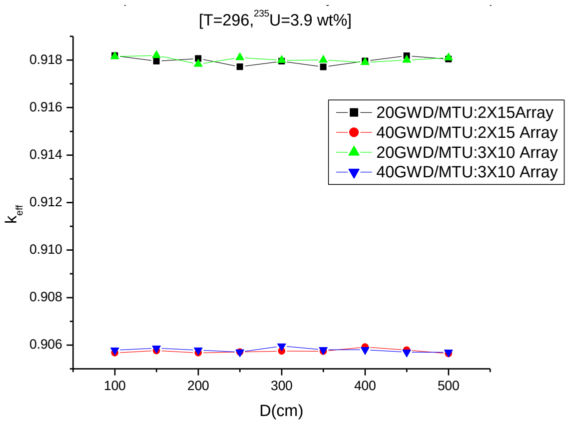

Figure 9: Effect of degree of Burnup on criticality

5.0 Conclusion

For a given number of *N* casks, the k_{eff} of the array (k_{array}) at the distance *d* and enrichment *e*, depends only on how the casks are arranged relative to one another:

- If *N* and *e* are constant and the array is such that there is at least one row acting as a shield between one another, then the k_{array} will only depend on the amount and type of shielding between them. However, if the array is such that there is no shielding between rows of the arrays, then k_{eff} will only depend on *N* in which case it will be higher than when they shield one another.
- If in a given *N* and *e*, *d* is increased the k_{eff} will decreases with an increase in *d*; the rate of decrease in k_{eff} is much higher in arrays where is shielding compared arrays where there is no shielding.

As can be seen on the graphs, there is a huge uncertainty on the k_{eff} of the system at any given separation gap (*d*). This may be due to the error of 10E5 which was included at the beginning of Keno input parameter.

By changing the type of the storage array from *nxn* to *nxm* (e.g. from 1X4 to 2X2 Array) one will significant change the neutron importance of the system, thus allowing those neutrons which would not have leaked out of the system to leak out, thus decrease the neutron importance and vice versa. In a given Array, by changing the distance among adjacent casks, will result in lower neutron density and subsequently lower neutron importance, hence in lower corresponding k_{eff}. When burnup credit is taken into account, higher burnups results in lower k_{eff} compared to their lower burnup counterparts.

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