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Disclaimer

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ABSTRACT

Criticality Evaluation of Plutonium-239 Moderated by High-Density Polyethylene in Stainless Steel and Aluminum Containers Suitable for Non-Exclusive Transport. TIMOTHY T. WATSON (Rensselaer Polytechnic Institute, Troy, New York 12180) JOHN SCORBY (Lawrence Livermore National Laboratory, Livermore, California 94550)

Research is conducted at the Joint Actinide Shock Physics Experimental Facility (JASPER) on the effects of high pressure and temperature environments on plutonium-239, in support of the stockpile stewardship program. Once an experiment has been completed, it is necessary to transport the end products for interim storage or final disposition. Federal shipping regulations for non-exclusive use transportation require that no more than 180 grams of fissile material are present in at least 360 kilograms of contiguous non-fissile material. To evaluate the conservatism of these regulatory requirements, a worst-case scenario of 180g ^{239}Pu and a more realistic scenario of 100g ^{239}Pu were modeled using one of Lawrence Livermore National Laboratory's Monte Carlo transport codes known as COG 10. The geometry consisted of ^{239}Pu spheres homogenously mixed with high-density polyethylene surrounded by a cube of either stainless steel 304 or aluminum. An optimized geometry for both cube materials and hydrogen-to-fissile isotope (H/X) ratio were determined for a single unit. Infinite and finite 3D arrays of these optimized units were then simulated to determine if the systems would exceed criticality. Completion of these simulations showed that the optimal H/X ratio for the most reactive units ranged from 800 to 1600. A single unit of either cube type for either scenario would not reach criticality. An infinite array was determined to reach criticality only for the 180g case. The offsetting of spheres in their respective cubes was also considered and showed a considerable decrease in the number of close-packed units needed to reach criticality.

These results call into question the current regulations for fissile material transport, which under certain circumstances may not be sufficient in preventing the development of a critical system. However, a conservative, theoretical approach was taken in all assumptions and such idealized configurations may not be likely to be encountered in actual packaging, transportation, and storage configurations. Modeling of realistic, as-built configurations is beyond the scope of this study.

INTRODUCTION

The Joint Actinide Shock Physics Experimental Research Facility (JASPER) is a two stage light gas gun which studies the effects of high pressure and temperature environments on various heavy metals, such as plutonium-239 in order to support the stockpile stewardship program. As a result of these efforts, waste products are created and must be transported off-site for disposal. Since ^{239}Pu is a fissile isotope it has the potential, given the wrong geometry, moderation, reflection, and spacing, to reach criticality. Such a configuration would be extremely hazardous for the public and for the workers transporting this material, as well as provide for an expensive environmental decontamination effort. At the same time, it is prudent to provide the most cost-effective and efficient method of transport as possible.

In Title 49 of the Code of Federal Regulations, Section 453, Part 173, an allowance exists under which a package of fissile material may be shipped as a non-"exclusive-use" item and, thus, an assigned a Criticality Safety Index (CSI) is not required. Such an exemption would greatly reduce the time and cost necessary for a strict analysis. The regulations state:

> "There is no more than 180 grams of fissile material distributed within 360 kilograms of contiguous non-fissile material. Lead, beryllium, graphite, and hydrogenous material enriched in deuterium may be present in the package, but must not be included in determining the required mass of solid non-fissile material."

This research is intended to verify the adequacy of the federal regulation listed above, and to determine if an actual package, which would contain of only ~100g ^{239}Pu would ever reach critical in varying configurations.

MATERIALS AND METHODS

All simulations were performed using Lawrence Livermore National Laboratory's Monte Carlo transport code known as COG 10. This code was developed by Dr. Richard Buck, Dr. Edward Lent, and Dr. Thomas Wilcox. COG 10 was run on the Criticality Safety Section's unclassified workstation, Surya. This system is a rack mounted Sun Fire V480 workstation with 4 Ultra SPARC III Cu 1.2 GHz processors and 8 GB of dynamic memory running under the Sun Solaris 5.8 Unix operating system. This software was installed and verified by Shang-Chih Philip Chou in July 2006. [1]

The geometry selected for simulation was intentionally chosen to both resemble an actual shipping container used for radiological waste transport, and to have a worst-case scenario configuration that would be the most likely to cause criticality. Two separate given quantities of plutonium-239 were modeled in homogeneous mixture with high-density polyethylene in the shape of a sphere. One case consisted of 180g Pu-239, while the other consisted of only 100g Pu-239. This sphere, for a majority of the cases run, was centered in a solid cube of either aluminum (Al) or stainless steel 304 (SS 304). Both materials were selected for modeling, as both can be the main constituents of a typical package assembly. Refer to Figure 5 for a generic COG-generated cross section view of the assembly.

Initial calculations consisted of determining an optimum H/X (hydrogen to fissile isotope concentration) ratio for the maximum allowed amount of fissile isotope in a package. This would provide for the most reactive configuration, and thus the most probable for self-induced criticality. This exercise also determined if a single unit would be critical. The optimized unit was then set in an infinite array, as a precursor to individual finite array simulations. If an infinite array was sub-critical in which no neutron leakage was present, then a finite array in which considerable neutron leakage was present would never go critical. Finite arrays consisting of 2, 4, 8, and 27 units were then simulated for the potentiality of stacking units during transport. Finally, the spheres of ^{239}Pu and polyethylene in the finite arrays were offset to their nearest neighbors, rather than centered in their individual cubic containers.

Determination of the safety or hazard of a given unit configuration was based on the calculation of k-effective. This is the ratio of the number of neutrons in the present generation of activity to the number of neutrons in the previous generation of activity. This value indicates the rate of increasing or decreasing neutron population in a given system, and thus its emission of neutrons to the surrounding environment. A critical system is defined in which k-effective is equal to 1, that is, the rate of neutron production equals the rate of loss. Such a system is unfavorable as it presents a serious radiation danger to anyone around it.

For each simulation 2000 neutrons were run in each of 200 generations. The first 10 generations were skipped from calculation. This removes any bias due to the artificial distribution created for the initial generations. Version 6.7 of the Evaluated Nuclear Data File from Brookhaven National Laboratory was used for neutron cross section data since it is the most recent cross section library included in the software.

RESULTS

The rise to maximum reactivity for a sphere of 180g ^{239}Pu and high-density polyethylene (HDPE) is shown in Figure 1. Initially, only small incremental amounts of HDPE are required to greatly increase k_{eff} for the unit. The maximum occurs when the H/X ratio is approximately 1022, after which the reactivity decreases linearly. The data collected for this optimized unit is shown in Table 1. The calculated k_{eff} for a stainless steel type 304 container is 0.8755, while for aluminum it is 0.7939. The radius of the sphere is approximately 11 cm, with a mass of 5.6 kilograms. The simulation of an infinite array of these two optimized units demonstrated the need for finite array analysis, as both were super-critical. Infinite arrays of the SS 304 contained unit and aluminum contained unit had k_{eff}'s of 1.0793 and 1.1274, respectively.

Figure 2 illustrates the increase in reactivity for two, four, eight, and twenty-seven centered unit finite arrays. None reached a critical condition. Figures 6, 7, and 8 are cross section views of the finite arrays in question. Offsetting of the spheres in their given cubes was then considered, such that their proximity to other spheres is maximized. Figures 9 and 10 illustrate a cross section view of the offset finite arrays. The results of these calculations are found in Table 4. Much fewer units were required to reach criticality. The four unit and eight unit SS 304 contained offset finite arrays and the eight unit Al contained offset finite array were considered for separation analysis.

The 100g ^{239}Pu and HDPE spheres were then analyzed. Figure 3 illustrates a more rigorous attempt to identify the optimum H/X ratio. As the peaks became well defined, one can see a slight difference between the SS 304 and aluminum contained units. For the stainless steel type 304 unit, the optimum H/X ratio ≅ 1100. The sphere radius is approximately 9.3 cm with a mass of 3.3 kilograms. The calculated k_{eff} resulted in a value of 0.7395. For the aluminum unit, the optimum H/X ratio ≅ 1550. The sphere radius is approximately 10.4 cm with a mass of 4.6 kilograms. k_{eff} for this unit was calculated at 0.6465. Tables 2 and 3 present all pertinent information about the above two optimized units. When infinite arrays were modeled, both container types proved to be sub-critical. The SS 304 contained unit array achieved a k_{eff} of 0.9314 while the aluminum contained unit array achieved a k_{eff} of 0.9379. Centered finite arrays were therefore not modeled. Instead, offset finite arrays were modeled with the optimized units. The data collected from these simulations can be found in Table 5. The 8 unit SS 304 contained offset finite array was considered for separation analysis.

Separation analysis consisted of incrementally moving the offset spheres away from an "in-contact" configuration, to determine the threshold distance at which they would sub-critical. Figure

4 presents this data. The 180g ^{239}Pu 4 unit SS 304 contained finite offset array reached a conservative sub-critical value of 0.9500 when the separation distance between spheres was ~ 5.0 cm. For the 180g ^{239}Pu 8 unit SS 304 contained finite offset array, 0.9500 was achieved at a distance of ~12.5 cm. For the 180g, 8 unit, Al contained finite offset array case, 0.9500 was achieved when separation reached ~7.2 cm. Finally, the 100g, 8 unit, SS 304 contained finite array, a separation of ~1.0 cm is necessary for k_{eff} to equal 0.9500.

DISCUSSION AND CONCLUSION

Potential sources of error in this study include idealized geometry configuration, and the use of only 200 generations for simulation. This was chosen only for conserving the time necessary to simulate the above geometries. The use of stainless steel type 304 proves to be more effective at reflecting neutrons and would thus provide a more reactive transport unit. The requirement as written in the Code of Federal Regulations is, in this specific case, not adequately conservative to prevent the assembly of a critical system. An offset finite array of 180g plutonium-239/HDPE units as illustrated in Figure 10 is a system that would present an extreme hazard to anyone around it. No single unit will ever achieve criticality without some disturbance or reassembly of the configuration, along with the presence of other units. It should be noted that all efforts were made for a conservative safety evaluation and that other controls such as stacking restrictions or spacing requirements are means of included safety but are beyond the scope of this study. Additional modeling should be conducted to verify the results of this study and to further analyze the adequacy of federal regulations concerning fissile-package transport. Potential accident conditions such as fire, flooding, and damage to the container were not under study, nor were any other credible abnormal conditions associated with criticality safety analysis.

ACKNOWLEDGMENTS

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REFERENCES

[1] Chou, Shang-Chih Philip, Installation and Verification of the COG 10 Code on Unclassified Workstation, Surya. Hazards and Control Department. Criticality Safety Section, Lawrence Livermore National Laboratory, Livermore, CA. CSAM06-102. July 19, 2006.

TABLES

Table 1- Calculated Data for Optimum 180g Pu-239 Units

Material	Density [g/cc]	Mass [g]	Volume [cc]	Specific Gravity [g/cc]	H/X Ratio
Pu-239	19.8400	180.0000	9.0726	0.0322	1022.5308
HD-Polyethylene	0.9670	5400.0000	5584.2813	0.9654	
			C	0.8266	
			H	0.1387	
Radius of Sphere [cm]	Total Volume [cc] (SS304)	Total Volume [cc] (Al)	Length of Side [cm] (SS304)	Length of Side [cm] (Al)	
11.0119	51865.8474	138926.6872	37.2930	51.7919	
Average K_{eff} for SS304:	0.8755				
Average K_{eff} for Al:	0.7939				

Table 2- Calculated Data for Optimum SS 304 Encased 100g Pu-239 Unit

Material	Density [g/cc]	Mass [g]	Volume [cc]	Specific Gravity [g/cc]	H/X Ratio
Pu-239	19.8400	100.0000	5.0403	0.0299	1100.0000
HD-Polyethylene	0.9670	3227.2866	3337.4215	0.9655	
			C	0.8268	
			H	0.1388	
Radius of Sphere [cm]	Total Volume [cc] (SS304)	Total Volume [cc] (Al)	Length of Side [cm] (SS304)	Length of Side [cm] (Al)	
9.2753	49614.9554	136675.7951	36.7455	51.5107	
Average K_{eff} for SS304:	0.7395				
Average K_{eff} for Al:	0.6348				

Table 3- Calculated Data for Optimum Al Encased 100g Pu-239 Unit

Material	Density [g/cc]	Mass [g]	Volume [cc]	Specific Gravity [g/cc]	H/X Ratio
Pu-239	19.8400	100.0000	5.0403	0.0212	1550.0000
HD-Polyethylene	0.9670	4547.5401	4702.7302	0.9660	
			C	0.8271	
			H	0.1388	
Radius of Sphere [cm]	Total Volume [cc] (SS304)	Total Volume [cc] (Al)	Length of Side [cm] (SS304)	Length of Side [cm] (Al)	
10.3970	50980.2642	138041.1039	37.0795	51.6816	
Average K_{eff} for SS304:	0.7283				
Average K_{eff} for Al:	0.6465				

Table 4- 180g Pu-239 Offset Spheres Arrays

Array Configuration [close-packed units]	K_{eff} (SS304)	K_{eff} (Al)
1	0.8755	0.7883
2	0.9348	0.8497
4	0.997	0.9333
8	1.087	1.0652

Table 5 - 100g Pu-239 Offset Spheres Arrays

Array Configuration [close-packed units]	K_{eff} (SS304)	K_{eff} (Al)
1	0.7395	0.6465
2	0.8049	0.7014
4	0.8783	0.7752
8	0.9752	0.8992

FIGURES

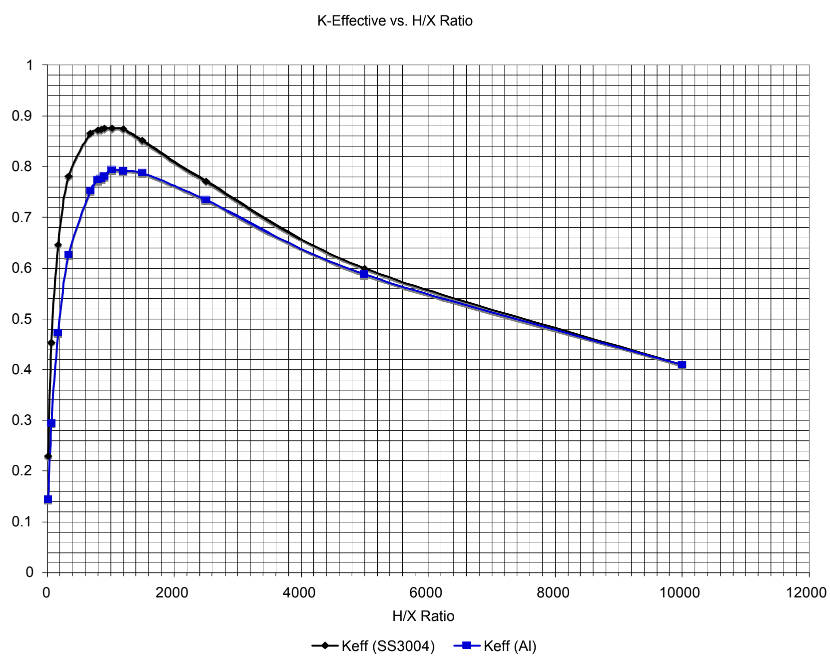

Figure 1- Optimum H/X Ratio Calculations for 180g Pu-239 Cases

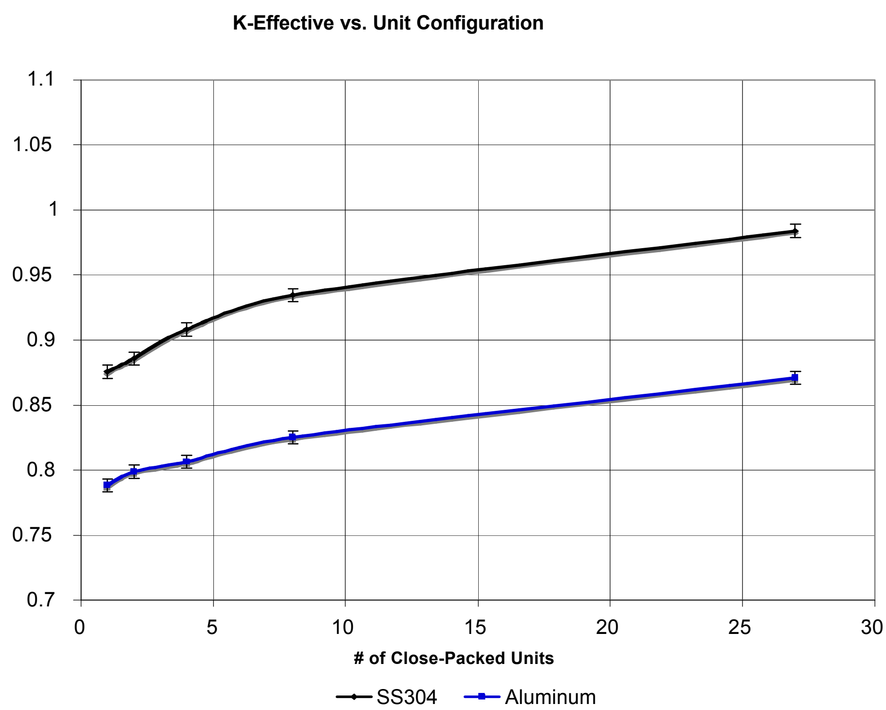

Figure 2- Approach to Criticality for 180g Pu-239 Finite Arrays

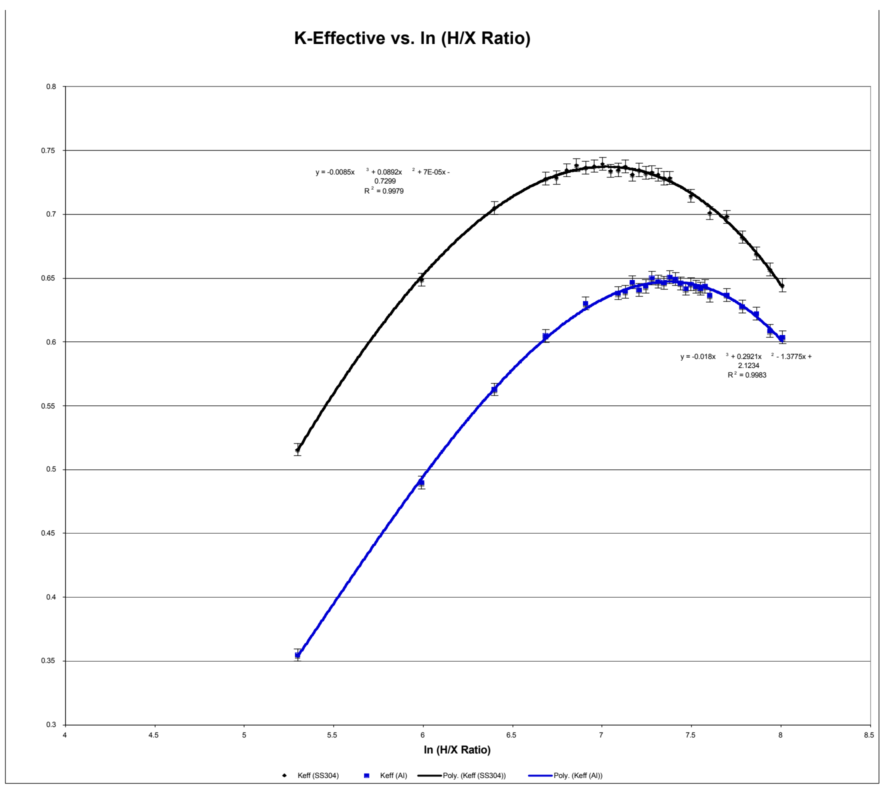

Figure 3- Optimum H/X Ratio Calculations for 100g Pu-239 Units

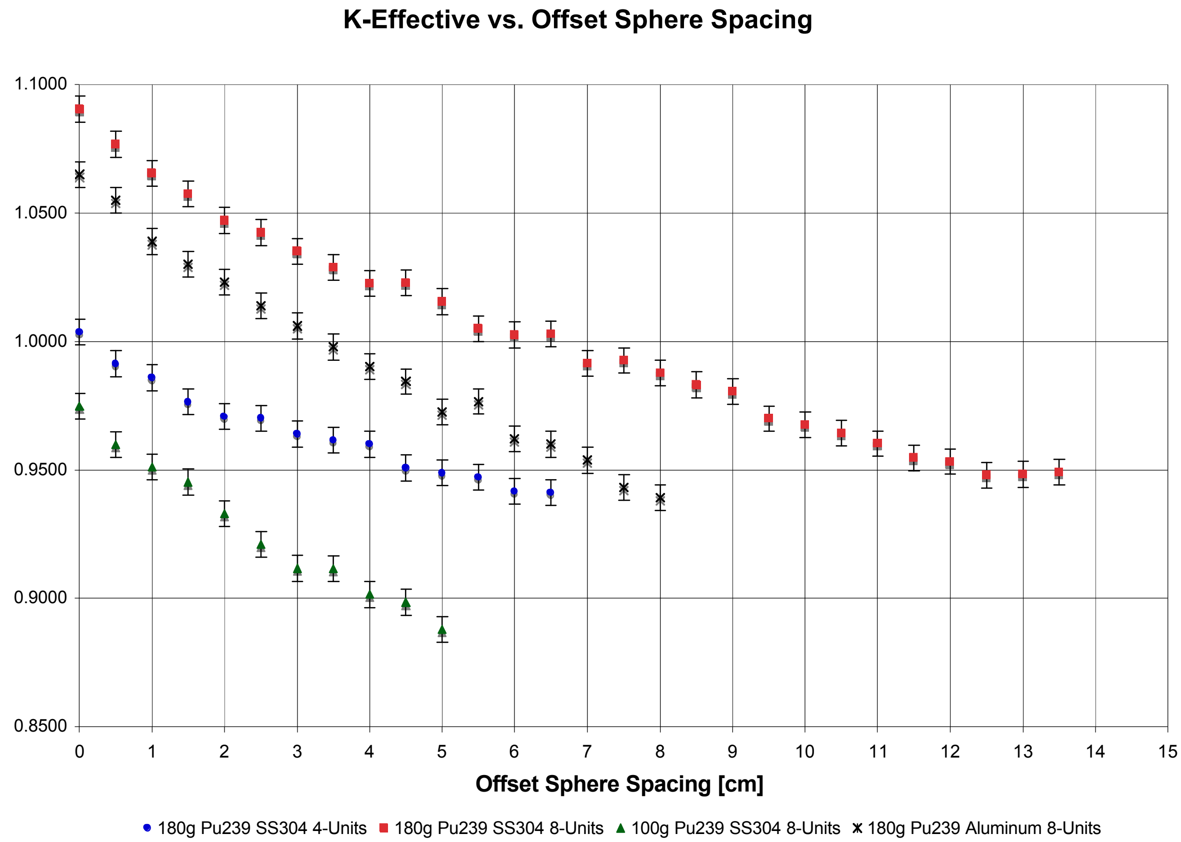

Figure 4 - Return to Sub-Criticality for Finite Arrays as Function of Sphere Separation

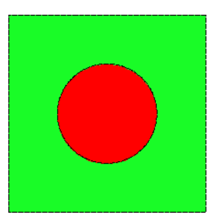

Figure 5 - Cross Section of Single Unit

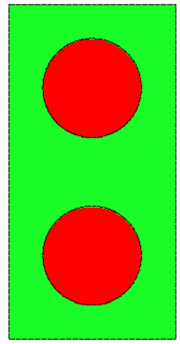

Figure 6 - Cross Section of Two Unit Centered Finite Array

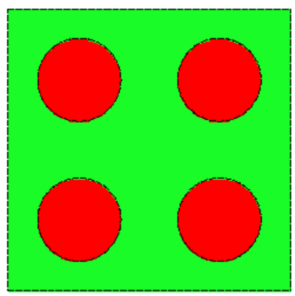

Figure 7 - Cross Section of Four and Eight Unit Centered Finite Array

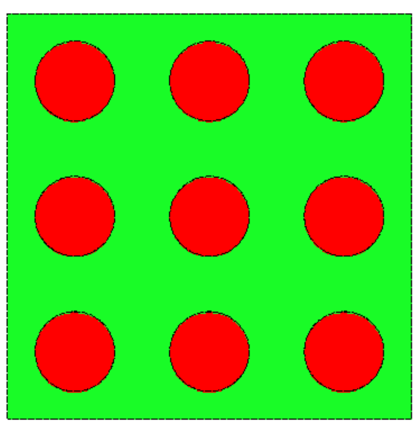

Figure 8 - Cross Section of Twenty-Seven Unit Centered Finite Array

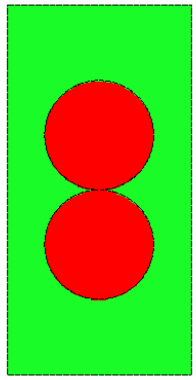

Figure 9 - Cross Section of Two Unit Offset Finite Array

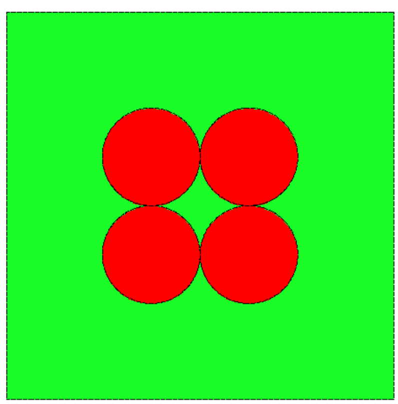

Figure 10 - Cross Section of Four and Eight Unit Offset Finite Array

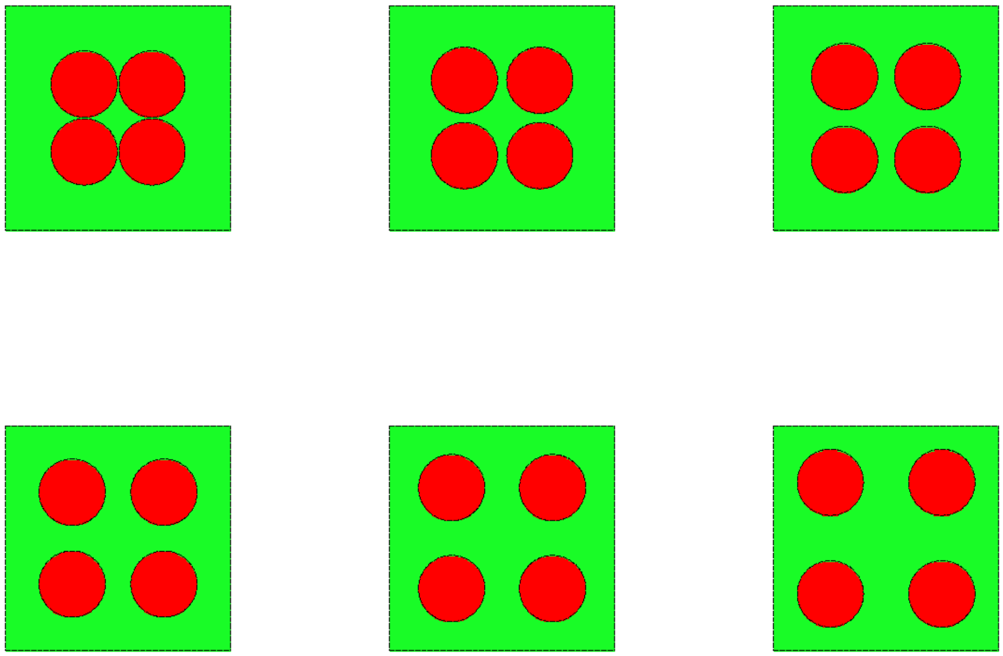

Figure 11 – Cross Section of Four Unit Offset Finite Array During Separation Analysis