QUANTITATIVE ANALYSIS OF GEORGE LAURENCE'S SUBCRITICAL URANIUM PILE

USING CURRENT REACTOR PHYSICS CODES

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2014

Abstract

In the early 1940s, Dr. George Laurence conducted several experiments to test the possibility of a sustained nuclear fission chain reaction in a carbon-uranium pile. Although the pile was proved to be subcritical, this pioneering experiment was not only significant in nuclear history, but was a valuable reference for Canada's later achievements. In this project, MCNP -- a current reactor physics code -- was utilized to create computer models of George Laurence's uranium pile based on existing literature information and drawings. A quantitative analysis using the modelling results was performed to investigate the impact of material impurities and the arrangement of uranium within the pile on the level of subcriticality. The goal of this project was to create MCNP models of the subcritical pile, calculate the effective multiplication factor ($k_{\rm eff}$), quantify the effect of various parameters on the $k_{\rm eff}$ and compare the calculated results with the experimental results reported by George Laurence. The maximum $k_{\rm eff}$ value 0.7355 was obtained for an experimental configuration using 1 tonne of uranium mixed with 10 tonnes of carbon, without any material impurities. Parametric studies were conducted on both the geometry of the pile and material impurities. It was observed that the shape of the uranium and coke bags did not have a significant impact on $k_{\rm eff}$, but that the spacing of the bags had a significant impact (since this changed the extent of moderation). The relationship between the $k_{\rm eff}$ and the purity of uranium as well as that between the $k_{\rm eff}$ and the boron concentration were observed to be linear.

1. Introduction

1.1 Purpose

The goal of this project is to explore the technical world of reactor physics calculations and the history of nuclear science in Canada. A current reactor physics code was used to model and visualize George Laurence's uranium pile experiments, based on existing drawings and reports. The purpose was to create a model of the uranium pile experiments, calculate the neutron multiplication factor (k_{eff} , the ratio of neutron production and absorption), and perform a quantitative analysis of the impact of material impurities and geometry of the pile on the level of subcriticality.

1.2 Hypothesis

It was hypothesized that the models created with MCNP would achieve a $k_{\rm eff}$ value close to what Laurence calculated for his uranium pile ($k_{\rm eff} = 0.90$). It was also hypothesized that the presence of impurities, such as boron, would have a negative impact the $k_{\rm eff}$ value, since neutrons would be lost by absorption. Furthermore, the separation between uranium bags was also hypothesized to impact the final $k_{\rm eff}$ results, since it would change the extent of neutron moderation.

1.3 Background

George Laurence

George Laurence was a Canadian nuclear physicist (1905-1987). He conducted the world's first experiment on a nuclear chain reaction in a large graphite pile at the National Research Council Laboratories in Ottawa in the early 1940s. He was a pioneer in Canadian nuclear research and became the second president of the Atomic Energy Control Board (which later became the Canadian Nuclear Safety Commission). He worked at Montreal nuclear energy laboratory and the Chalk River Nuclear Laboratories. He directed the development and design of the Zero Energy Experimental Pile (ZEEP) and National Research Universal (NRU), helping Canada become the second country to control nuclear fission in a reactor [1].

George Laurence's Experiment

In the mid-twentieth century, Laurence, along with many other physicists, was interested in the potential power of nuclear fission. By the end of the summer of 1942, George Laurence was able to demonstrate, but failed to sustain, a nuclear fission chain reaction in the natural uranium pile he constructed at the National Research Council Laboratories in Ottawa. Since these experiments were performed during World War II, they were classified and only a small number of scientists were aware of their existence and were involved in conducting them. The experiments failed to produce a sustainable chain reaction (the reaction was 10% away from reaching criticality, according to his calculations), primarily due to the impurities in the uranium oxide (U_3O_8) and carbon used and the limited amount of fissile material (U_3O_8) and moderator (carbon).

Laurence designed the pile in a cylindrical wooden bin, 140.3 cm in radius and 270 cm in height (as shown in Figure 1). The bin was covered with a layer of 7 cm paraffin wax, which was used as a neutron reflector. In the centre, there was a horizontal tube to introduce a source of neutrons which drove the pile. The source of neutrons was 200 milligrams of beryllium mixed with a few grams of radium compound inside a metal tube about 2.5 cm long. The whole structure was filled with ten tonnes of calcined petroleum coke powder (98% carbon) in paper bags and several hundred bags of natural uranium oxide totalling one tonne. The sacks of uranium and coke occupied a space that was roughly spherical, a shape that is the intersection area of two 1.33 m-radius spheres with centers separated by a horizontal distance of 66.5 cm [2]. Although heavy water is a much better moderator than carbon, it was not readily available in large quantities in the early 1940s; hence, Laurence used relatively inexpensive and commonly found coke instead of heavy water as a moderator for his pile experiments.

Laurence was aware that due to the impurities in the coke and uranium, the reaction would not be sustained and the pile would be subcritical. He eventually achieved a calculated k_{eff} of 0.90. Nonetheless, Laurence's experiment was a great reference for the design and construction of Canada's first nuclear reactor, ZEEP. Laurence deserves honourable mention in any historical account of Canadian science for being one of the first in the world to experiment with large-scale subcritical reactors.

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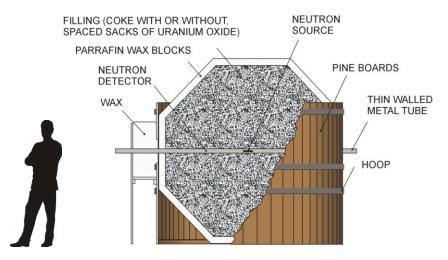


Figure 1. George Laurence's uranium pile [3].

1.4 Application.

This project is both history- and theory-based. The information and assumptions made in the project provide a database for historical accounts. The results of the project will provide detailed and valuable information for Laurence's experiment in regards to material impurities and uranium bag arrangement. The analysis of results yields important information for conjecturing the missing history of the pile.

2. Experimental Methods, Material and Equipment

2.1 Equipment

The program that was used in this project is the Monte Carlo N-Particle Transport Code (MCNP). MCNP is a software package for simulating nuclear processes and calculating the effective neutron multiplication factor (k_{eff}). A visual editor was utilized to create 3-D computer models and visualize Laurence's uranium pile based on existing information and assumptions.

2.2 Experimental Procedure

- 1. Read through related materials and gather background information;
- 2. Make assumptions and adjustments;
- 3. Create input files based on the assumptions and compiled information;
- 3. Use the MCNP visual editor to visualize computer models;
- 4. Run the created models using MCNP and calculate the k_{eff} value;
- 5. Compare and analyse results. Make charts or graphs based on the results.

Repeatedly conduct the above procedure until the desired results are achieved and enough information is obtained to make a quantitative analysis.

3. Theory

3.1 The Effective Neutron Multiplication Factor (keff)

The k_{eff} is the ratio of the neutrons produced by fission to the number of neutrons lost through absorption or leakage. In any one generation, neutrons are produced from fissioning of fissile materials (e.g., 235 U, 233 U or 239 Pu), while other neutrons are lost by being absorbed by various atoms or by leaking out of the system. The value of the k_{eff} determines how a nuclear chain reaction is proceeding, i.e., if the system is subcritical, critical or supercritical.

$$k_{eff} = \frac{Production}{Absorption + Leakage}$$

Subcritical System

A system is said to be subcritical when $k_{eff} < 1$, that is, when the rate of neutron loss is greater than that of neutron production. In this case, the system cannot sustain a chain reaction, and any beginning of a chain reaction dies over time.

Critical System

A system is said to be critical (multiplication factor $k_{eff} = 1$) when the rate of neutron loss is equal to that of neutron production. The neutron population and reaction rate remain constant in this case and the chain reaction can be sustained.

Supercritical System

A system is said to be supercritical when $k_{eff} > 1$, that is, the rate of neutron production is greater than that of neutron loss. The neutron population and reaction rate increase with each generation; the chain reaction thus can no longer be controlled.

3.2 Nuclear Cross Section

The nuclear cross section is the effective area the nucleus presents to a neutron of a particular energy. The larger the effective area of the nucleus, the greater the probability for a reaction to occur between the incoming neutron and this nucleus. For example, the larger the absorption cross section of an atom of a material, the more likely it is that the neutrons will be absorbed by that particular material [4]. In our case, boron is an important element since it has a high absorption cross section; hence, absorbing the neutrons that are needed to fission ²³⁵U and result in a nuclear chain reaction.

3.3 Neutron Source

The neutron source Laurence used in his uranium pile was a mixture of a few grams of beryllium and 200 milligrams of radium. He placed them in the center of a brass tube that went through the center of the pile. Radium spontaneously emits alpha particles, which are absorbed by beryllium resulting in neutrons being produced via an (α, n) reaction as beryllium-9 becomes carbon-12 [4]. This neutron source was modeled using the Ra-Be neutron spectrum shown in Figure 2. This figure shows the relationship between neutron density and neutron energy for the mixture of radium and beryllium assumed to be used in the subcritical pile experiments.

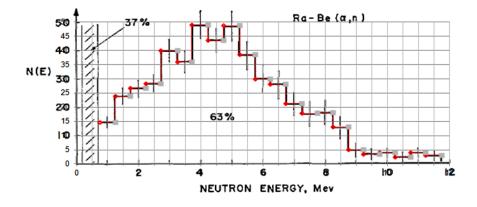


Figure 2. Neutron spectrum of a Ra-Be Source [5]

3.4 Moderation and Thermalization

Since slower travelling neutrons (e.g., speeds of ~1-2 km/s) are more likely to be absorbed by ²³⁵U and the neutrons produced from fissioning of ²³⁵U are fast neutrons (e.g., speeds of ~10,000 km/s), their speed must be significantly decreased. ²³⁵U has a high probability to fission (i.e., a high fission cross section) with thermal neutrons while ²³⁸U, due to its higher critical energy,

does not fission easily with slow neutrons. The process of reducing the energy of a neutron to thermal energy is referred to as thermalization, slowing down, or moderation. The materials that are used for moderation are called moderators. The neutron source Laurence used emitted high-energy neutrons. Figure 2 shows that the average neutron energy from the Ra-Be source was ranging from 3 MeV to 6 MeV, while thermal neutrons have energies under 1 eV [6]. Thus, moderators play a crucial role in sustaining a continuous nuclear reaction. This is also why the quantity of moderating material between the uranium bags can have a significant impact on the k_{eff} value.

Commonly used moderators include light water (H_2O) , heavy water (D_2O) , and carbon. Laurence chose carbon as a moderator since it was the only fairly inexpensive and common material that met theoretical requirements. Canadian Deuterium Uranium (CANDU) reactors use heavy water as a moderator, for the reason that D_2O has a smaller neutron absorption rate; thus, allowing the solution to have a moderating ratio (a measure of moderating efficiency) 80 times higher than that of regular light water and also allowing natural uranium to be used as fuel [7].

4 Results

4.1 Assumptions

When creating the computer models, some assumptions needed to be made. When doing parametric studies on materials, it was assumed that the uranium oxide (U_3O_8) had 2% - 5% impurities. The main impurity that was considered was boron since boron has a high absorption cross section. Other impurities that were considered included SO_4 , SiO_2 , Ca and Fe. Calcined petroleum coke, which is used as the moderator instead of pure graphite, was assumed to have several impurities including H, Ni, S and O.

Other assumptions regarding the size and arrangement of the paper bags within the pile also needed to be made. The actual paper bags were not modelled since it was assumed that their impact on the neutron multiplication factor would not be significant when compared to the effect of boron impurities in the uranium and the coke. In the models, it was also assumed that the calcined petroleum coke was placed uniformly around uranium bags instead of being packed into individual paper bags. When creating rectangular uranium bags, the assumption that the bags have a square base with a height twice the length of one side of the base was also made. In his report, Laurence mentioned that the average spacing between the bags was 16.35 cm, but he did not specify whether this distance is from the edge of one bag to the edge of the other or from the center of one to the center of the other, hence both scenarios were investigated.

Since the only report published by George Laurence on these experiments [2] did not include all the necessary design information, several assumptions were made about the structure of the pile. For example, the top of the pile is assumed to have a truncated conical shape, a reflection of the shape he described for the bottom of the pile.

Laurence also mentioned that the coke was loosely packed. However, when creating the models, the assumptions had to be made on whether air is between each bag, uniformly mixed with coke or on the top of all the materials, in order to maintain the total reported amount of moderator (i.e., 10 tons).

4.2 Discussions

4.2.1 Parametric studies on the geometry of the pile

Appendix A shows some of the models that were tested and their calculated k_{eff} values. All the models in Appendix A used pure materials and are based on different geometry configurations and design assumptions.

An inspection of Appendix A shows the definite evidence that the hypothesis is partially wrong. Even without material impurities, it is extremely difficult to obtain a k_{eff} close to 0.90, due to the lack of sufficiently detailed experimental information. Most of the models that have been created based on the information in Laurence's report are only able to achieve a k_{eff} of around 0.71. However, certain evidence has shown that every small adjustment on geometry can have impact on the k_{eff} .

It is shown in Figure 3 that the shape of uranium bags can affect $k_{\rm eff}$ even though not significantly. If the shape of the uranium bag is the only changed parameter (i.e., the arrangement of the bags, number of uranium bags and material compositions are maintained), the spherical uranium bag model has a small advantage when compared to the cubic uranium bag model with the same volume. The reason could be that a sphere has a significantly larger surface area than the cube with the same volume.

Model number	Spacing Between Bags	Moderator	Lattice Shape	Bag Shape	Mass of Bag (g)	Bag Number	k _{eff}	Мо	dels
5		Carbon-Air		Sphere			0.7199		• •
6	Edge to Edge	Uniform Mixture	Square	Cube	3780	265	0.6738		

Figure 3. Impact of bag shape on k_{eff}

It is hypothesized and also shown in Laurence's report that the double bag model (265 bags) achieves a higher k_{eff} than the single bag model (529 bags), yet the following models proved that this statement is not necessarily true based solely on the information provided. Figure 4 shows that bag sizes do not have a significant impact on the k_{eff} value and sometimes, a single bag model can yield a better result than a double bag model. This could be due to the fact that, if the system is under-moderated, the absorption of neutrons by ^{238}U is increased when the double bags are modeled. This increased absorption rate results in a lower k_{eff} value.

Model number	Spacing Between Bags	Moderator	Lattice Shape	Mass of Bag (g)	Bag Number	keff	1	Models	
21		Air above		1890	529	0.7339			
22	P.J.	densely packed pile Carbon-Air Uniform Mixture		3780	265	0.7278			
1	Edge to Edge		Uniform	Hexagonal	1890	529	0.7239		
4				Uniform		3780	265	0.7226	

Figure 4. Impact of bag numbers on keff

When calculating the total volume of Laurence's pile using the given information reported in Section 1.3, it is found that the pile has a far greater volume than all materials combined. Since Laurence also mentioned that the coke is loosely packed, it is interpreted that significant amount air is mixed into the pile. Therefore, three configurations – "Air above densely packed

pile", "Carbon-air uniform mixture" and "Air between carbon hexagons"--are used and compared. As Figure 5 shows, "Air above densely packed pile" tends to have the best result, "uniformly mixed carbon" next and "air between carbon hexagons" models third. However, the resulting difference is dependent on each model investigated. For example, for the edge to edge spacing models, the differences are only 0.0052 and 0.0041, while in the center to center models, the differences are 0.0091 and 0.0443.

Model number	Spacing Between Bags	Moderator	Lattice Shape	Bag Shape	Mass of Bag (g)	Bag Number	keff	Mod	lels
12		Air Above Densely Packed Pile					0.7320		
8	Edge to Edge	Carbon-Air Uniform mixture	Hexagonal	Cube	e 1890	529	0.7268		
9		Air between C hexagons					0.7227		
7		Air Above Densely Packed Pile					0.6480		
10	Center to Center	Carbon-Air Uniform mixture	Hexagonal	Cube	1890	529	0.6389		
11		Air between C hexagons					0.5946		

Figure 5. Impact of Carbon-Air arrangements and uranium bag distance on k_{eff}

The above examples suggested some possible factors that have impacts on the $k_{\rm eff}$ value. However, there is only one main factor that affects the $k_{\rm eff}$ value the most from a geometrical perspective. In Figure 5, it is observed that "edge to edge" models achieve better results than "center to center" models because in "edge to edge" models, the larger quantity of the moderator slows down neutrons more effectively.

The quantity of the materials has the most significant effect on the k_{eff} . In Figure 6, it is suggested that the larger the quantity of uranium bags, the higher the k_{eff} value. This finding also matches Laurence's report "Certain evidence suggested that the quantity of oxide in each unit could be increased to considerable advantage" [8].

Spacing	Moderator	Lattice Shape	Amount of Uranium	k _{eff}	М	lodels
	Carbon-Air Uniform mixture		20% uranium	0.4787		
Edge to Edge	Carbon-Air Uniform mixture	Hexagonal	1 ton uranium	0.7114		
	Air in between		1.5 times uranium	0.8347		

Figure 6.
Impact of uranium quantity on k_{eff}

4.2.2 Parametric studies on materials impurities

When conducting parametric studies on material impurities, boron is considered a major impurity due to its high absorption cross section. Figure 7 shows the impurities that were considered when creating the models and their associated weight percentages. Despite the variety of the impurities, boron is the only independent value that was considered, while the other impurities are treated as having constant weight percentages among the impurity compositions. The uranium oxide purity and the boron composition in carbon and U_3O_8 were varied. The results for each case are illustrated in Figures 8 – 10.

Uranium oxide

Material	Weight percentage (%)
U_3O_8	95
SO_4	3.125
SiO ₂	1.25
Ca	0.3125
Fe	0.3125

Carbon

Material	Weight percentage (%)
С	98.0763
Н	0.1395
0	0.0199
Ν	0.2193
S	1.1961
Ni	0.1744
V	0.1744

Figure 7. Material impurities and compositions in uranium oxide and calcined petroleum coke [9].

For all the graphs and calculations below, the model used has a relatively high $k_{\rm eff}$ value of 0.7355 without any material impurities. The model is an edge to edge example with single bags. The air is above all the fillings and the wax (model 23 in Appendix A). Uranium impurities ranged from 2% to 5% and boron concentration ranged from 0 ppm (parts per million) to 6 ppm are input to generate the graphs.

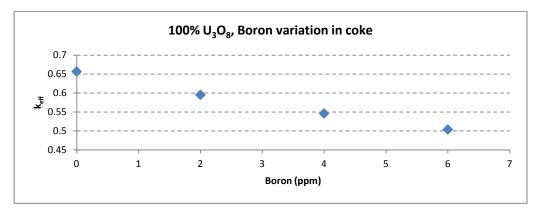
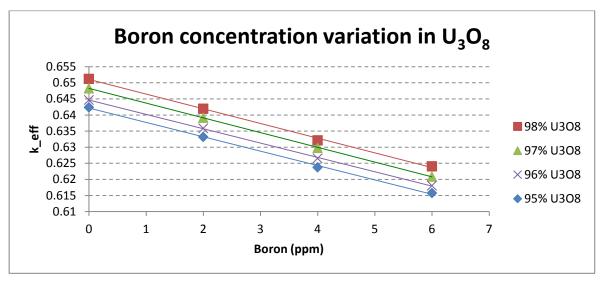


Figure 8. k_{eff} vs. Boron concentration in coke with 100% U_3O_8

Figure 8 shows the relationship between boron concentrations in coke and k_{eff} values when uranium is pure. The graph evidently shows a linear function with an average slope of -0.0255 which indicates that every 1 ppm of increase in boron would cause a 0.0255 decrease in k_{eff} value.



 $\label{eq:figure 9.} Figure 9. \ k_{eff} \, vs. \\ Boron \\ concentration in \\ U_3O_8$

The four lines in the Figure 9 indicate the four different cases of uranium oxide purity. They are parallel to each other and show the consistent linear relationship between boron concentration in the uranium and k_{eff} value. It is also observed that when the boron concentration is constant, the k_{eff} increases proportionally with uranium purity. Each percent increase in uranium purity leads to an average increase of 0.05 in k_{eff} .

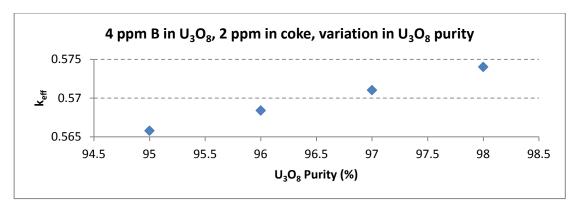


Figure 10. k_{eff} vs. U_3O_8 purity, with 4 ppm Boron in U_3O_8 , and 2 ppm Boron in coke

Figure 10 shows the relationship between uranium purity and k_{eff} value with 4 ppm boron in uranium oxide and 2 ppm in coke. The graph again shows a linear relationship between the two. The 4 ppm boron in uranium and the 2 ppm boron in coke are the impurities commonly accepted in Laurence's materials. But, as shown in the graph, even with 98% pure uranium, the model is only able to achieve a k_{eff} value of 0.574, far from the expected value of 0.90.

4.3 Error Analysis

4.3.1 Parametric Studies on Pile Geometry

The ambiguity of Laurence's report [2] and the limitation of available detailed information are two main sources of error. In the report, Laurence did not mention exactly how the uranium bags are arranged. A 16.35 cm spacing is reported; however, it is not clear whether this spacing is edge to edge or center to center, and if the spacing is maintained in x-, y- and z-direction concurrently. Laurence also mentioned two different sizes for uranium bags and various sizes of carbon bags, yet did not mention the exact sizes and shapes of the bags.

The models created so far ignore the effect of the paper bags Laurence used to pack uranium and coke. Since the paper bags were not modeled, the potential neutron absorption of the materials in the paper bags was ignored. Furthermore, when creating the model, the coke was spread around the uranium uniformly instead of packing coke into bags. These assumptions could also have a small, although not significant, impact on the final results.

In some of the models, air was mixed uniformly with the carbon which is less likely to happen in real situation.

Another possible error that causes this project's result to differ from that of Laurence's might be his own miscalculation. When Laurence was calculating the $k_{\rm eff}$ value, he reports comparing two different methods –"the American" Method and "Halban's" method. The American method gave him a $k_{\rm eff}$ value of 0.90 while the result of the "Halban" method was only 0.71. He described in detail how he calculated the value using the "Halban" method, but did not give much description on the calculation using the American method. Furthermore, the report does not give much explanation on the reasoning behind choosing 0.90 over 0.71, except saying that "since the measurements in the wax shell were inaccurate, owing to low intensities, the value 0.90 found by the American method is regarded as more accurate" [10]. Therefore, there is a possibility that the actual $k_{\rm eff}$ value Laurence obtained is closer to 0.71 than 0.90.

4.3.2 Parametric Studies on Material Impurities

The impurities and their weight percentage used might differ from the ones actually present in Laurence's experiment. The assumption that all impurities except for boron has constant weight fraction is also one of the possible sources of error. Furthermore, very limited information was found on the extent of impurities that could have been present in the actual U_3O_8 and coke materials used in the experiments.

4.4 Conclusion

Based on all the modeling results, it is observed that most models based on Laurence's report can only achieve a k_{eff} around 0.7 without the impurities. With the impurities of 4 ppm boron in uranium and 2 ppm in coke, the model is only able to achieve a k_{eff} value of 0.566, far from what Laurence reported. It is concluded from the graphs in Section 4.2.2 that the increase in the k_{eff} value is inversely proportional to the boron concentration in coke and uranium, but directly proportional to the increase in uranium purity. The uranium bag sizes and the shape of the uranium bags do not have a significant impact on the k_{eff} . Only the spacing between uranium bags has the most dominant and obvious effect on the k_{eff} in regards to the geometry of the pile.

4.5 Future Research

The next steps of this project should focus on parametric studies on both material impurities and geometries. The future work should involve finding more solid references to reduce assumptions and misinterpretations of Laurence's experiment. It is still uncertain what had a significant impact on the k_{eff} value, except for the distance between the uranium bags. More models should be created to investigate the precise impact and reason of impact behind each of the geometry changes. More research should also be done on finding out the exact k_{eff} value of Laurence's pile, since the standards and methods Laurence used to obtain his $k_{eff} = 0.90$ might be different form nowadays' standards and calculations.

5 Acknowledgements

The author of this paper would like to thank Atomic Energy of Canada Limited for providing resources and equipment needed. She would also like to thank Jeremy Whitlock (AECL), Laura Blomeley (AECL) Morgan Brown (AECL), Darren Radford (AECL), Jim Arsenault and Fred Boyd for providing support and help gathering information for the project. Duncan Stewart (NRC) and Terry Jamieson (CNSC) are also thanked for their financial report. The NRC is especially thanked for its interest in this project and invitation. The author appreciates the guidance and supervision from Ruxandra Dranga and Rebecca Carrington; James Xu is also acknowledged for his input in this project and excellent partnership. Brandon Whitley (DRSA), Chloe Lampman (DRSA), Shawna Kunkel (DRSA), Chantel Goodman (DRSA), John Wills (DRSA), and Margo Ingram (DRSA) are thanked for their support and help. This project was organized and supported by Deep River Science Academy and its organizers are acknowledged for making this project possible.

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Appendix A: Parametric Study on Pile Geomotry

Spa cing		Moderator	Lattice Shape	Bag Shape	Bag Type	k _{eff}	XZ View	XY View
ETE	1	Carbon-Air Uniform Mixture	Hexagonal	Rectangle	Single	0.7239		
ETE	2	Carbon-Air Uniform Mixture	Hexagonal	Sphere	Single	0.4787		
ETE	3	Carbon-Air Uniform Mixture	Hexagonal	Sphere	Single	0.7114		
ETE	4	Carbon-Air Uniform Mixture	Hexagonal	Sphere	Double	0.7226		
ETE	5	Carbon-Air Uniform Mixture	Cube	Sphere	Double	0.7199		
ETE	6	Carbon-Air Uniform Mixture	Cube	Cube	Double	0.6738		
CT C	7	Air Above Densely Packed Pile	Hexagonal	Cube	Single	0.6480		
ETE	8	Carbon-Air Uniform Mixture	Hexagonal	Cube	Single	0.7268		
ETE	9	Air between Carbon Hexagons	Hexagonal	Rectangular prism	Single	0.7227		
CT C	10	C-air Uniform Mixture	Hexagonal	Cube	Single	0.6389		
CT C	11	Air between Carbon Hexagons	Hexagonal	Cube	Single	0.5946		

ETE	12	Air Above Densely Packed Pile	Hexagonal	Cube	Single	0.7320	
ETE	13	Air in Between	Hexagonal	Cube	Double	0.8347	
ETE	14	Air in Between	Hexagonal	Rectangular prism	Double	0.8346	
ETE	15	No Air	Cube	Cube	Double	0.8467	
ETE	16	Carbon-Air Uniform Mixture	Hexagonal	Rectangular prism	Single	0.7223	
ETE	17	Carbon-Air Uniform Mixture	Long Hexagonal	Rectangular prism	Single	0.7235	
ETE	18	Carbon-Air Uniform Mixture	Hexagonal	Rectangular prism	Single	0.7249	
ETE	19	Carbon-Air Uniform Mixture	Hexagonal	Cube	Single	0.8172	
ETE	20	Carbon-Air Uniform Mixture	Hexagonal	Cube	Double	0.7198	
ETE	21	Air above densely packed pile	Hexagonal	Rectangular prism	Single	0.7339	
ETE	22	Air Above Densely Packed Pile	Hexagonal	Rectangular prism	Double	0.7278	
ETE	23	Air Above Densely Packed Pile	Hexagonal	Rectangular prism	Single	0.7355	

ete: Edge to Edge

ctc : Center to Center

Single: 529 bags each with 1890 g

Double: 265 bags each with 3780 g

Model Number	XZ View	XY View
1		
2		
3		

