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**DETERMINATION OF LINEAR ATTENUATION COEFFICIENTS OF
CHEMICALLY BONDED PHOSPHATE CERAMICS**

By

JASON JOSEPH PLEITT

A THESIS

Presented to the Faculty of the Graduate Faculty of the

MISSOURI UNIVERSITY OF SCIENCE AND TECHNOLOGY

In Partial Fulfillment of the Requirements for the Degree

MASTER OF SCIENCE IN NUCLEAR ENGINEERING

2012

Approved by
Carlos H. Castano, Advisor
Shoaib Usman
Michael Schulz

PUBLICATION THESIS OPTION

This thesis consists of the following two articles which have been submitted for publication as follows:

Pages 2 to 24 have been published in the JOURNAL OF NUCLEAR MATERIALS.

Pages 25 to 37 have been published in THE AMERICAN CERAMIC SOCIETY'S CERAMIC TRANSACTIONS PROCEEDINGS.

ABSTRACT

Chemically Bonded Phosphate Ceramics (CBPCs) is a composite based material formed in a manner similar to cements. CBPCs have shown to be a good structural material with excellent thermal resistance properties. They have also been shown to have potential as a radiation shield. By incorporating various powders into the ceramic the improvement in its radiation shielding properties are examined. The method for obtaining the linear attenuation coefficients for CBPCs as well as MCNP simulated results for gamma and neutrons are presented.

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I would like to thank my advisor, Dr. Castano who provided me with the opportunity to perform this work and provided guidance for my research through both my undergrad and graduate schooling. I would also like to thank Lucas Tucker who by teaching me many different things about neutrons and neutron detection.

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Finally, I would like to dedicate this work to my parents, Joseph and Denise Pleitt. Their constant words of encouragement and gentle pushing have made me into the person I am today and this work is just a small part of the thanks that I can do for all they've done for me.

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1 INTRODUCTION

Chemically Bonded Phosphate Ceramics (CBPCs) are formed in low temperature conditions by mixing a base powder with phosphoric acid. The compound that forms from the cement like processing has the properties of an engineering ceramic with the advantage of being able to be formed with relative ease. An additional advantage is that the compound can be formulated with additives that can enhance the properties of the ceramic without causing many serious issues to the structural properties. In addition the high melting point temperatures and the good thermal resistance have caused the ceramic to be used as a firewall to prevent transformer explosions. Based upon these principles the ceramic was examined using several different additives to determine the improvement of the linear attenuation coefficient for dry cask fuel storage.

Dry cask spent fuel storage involves storing spent nuclear fuel for either above ground or transport in cement casks that are lined with lead gamma shielding. One issue that arises with these casks is that while they are designed to withstand high temperatures for long lengths of time there have been previous accidents that have resulted in temperatures that exceed the current safety design parameters. In order to make sure that no issues arise with current casks design this research examines CBPCs in order to determine how to improve the shielding capabilities so that current casks can survive if an accident exceeded current safety parameters.

PAPER I

Wollastonite Based-Chemically Bonded Phosphate Ceramics with Lead Oxide Contents under Gamma Irradiation

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ABSTRACT

The shielding properties to gamma rays as well as the effect of lead concentration incorporated into chemically bonded phosphate ceramics (CBPC) composites are presented. The Wollastonite-based CBPC was fabricated by mixing a patented aqueous phosphoric acid formulation with Wollastonite powder. CBPC has been proved to be good structural material, with excellent thermal resistant properties, and research already showed their potential for radiation shielding applications. Wollastonite-based CBPC is a composite material itself with several crystalline and amorphous phases. Irradiation experiments were conducted on different Wollastonite-based CBPCs with lead oxide. Radiation shielding potential, attenuation coefficients in a broad range of energies pertinent to engineering applications and density experiments showing the effect of the

PbO additions (to improve gamma shielding capabilities) are also presented. Microstructure was identified by using scanning electron microscopy and X-ray diffraction.

Keywords

Chemically Bonded Ceramics, PbO, gamma radiation, shielding, nuclear applications.

1 INTRODUCTION

Chemically Bonded Ceramics (CBCs) are inorganic solids synthesized by chemical reactions at low temperatures without the use of thermally activated solid-state diffusion (typically less than 300°C) [1]-[3]. This method avoids high temperature processing (by thermal diffusion or melting) which is the normal in traditional ceramics processing. The chemical bonding in CBCs allows them to be inexpensive in high volume production. Because of this, CBCs have been used for multiple applications. They include: dental materials [4], nuclear waste solidification and encapsulation [5], electronic materials [6], composites with fillers and reinforcements [7, 8]. The fabrication of conventional cements and ceramics is energy intensive as it involves high temperature processes and emission of greenhouse gases, which adversely affect the environment [8].

Public acceptance is an important factor in the construction of new nuclear power plants, and addressing potential scenarios for the safe handling and transportation of high level spent fuel is necessary to maintain public's trust. As such, there has been an ongoing debate on the suitability of the current cask design used for transporting spent nuclear fuel (a type of high level waste) across the US due to some accidents in confined spaces.

In particular, two fire accidents in tunnels, the first on April 7, 1982 a tank truck and trailer carrying 8,800 gallons of gasoline was involved in an accident in the Caldecott Tunnel on State Route 24 near Oakland, California [10], and second on July 18, 2001 where a freight train carrying hazardous (non-nuclear) materials derailed and caught fire while passing through the Howard Street railroad tunnel in downtown Baltimore, Maryland [11]. If current designs of nuclear casks had been involved in the accidents, the neutron shielding would have been lost, and depending on the design (lead) the gamma shielding could be partially or totally lost as well [10].

CBPC ceramics for thermal shock and firewalls applications have been designed due to the superior fire resistance [12]. This material outperforms concrete for prolonged contact with fire, and can take much higher temperatures before failing [13]. We propose using a specially formulated CPCB as a combined gamma, neutron, and thermal shield, besides adding substantial structural integrity to the redesigned nuclear cask.

In this research, a CBC formed by Wollastonite powder (CaSiO_3) and PbO powders, mixed with a phosphoric acid formulation (H_3PO_4) in a ratio of 100/120 powders to acid, reacts into a Chemically Bonded Phosphate Ceramic (CBPC).

In CBPCs, when the aqueous phosphoric acid formulation and the Wollastonite powder mixture are stirred, the oxides dissolve and an acid base reaction is initiated, resulting in a slurry that hardens into a ceramic product [14]. Wollastonite is a natural calcium meta-silicate which is mostly used as filler in resins and plastics, ceramics, metallurgy, biomaterials and other industrial applications. PbO was added to the mixture always keeping constant the total amount of oxide powders (Wollastonite + PbO). The PbO was used in order to determine its effect on the attenuation coefficient of the CBPC.

Thus, the mixing of Wollastonite with phosphoric acid produced a composite material itself with brushite ($\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$) and Wollastonite as crystalline structures; and silica and amorphous calcium phosphates as amorphous phases [15]. We expect PbO introduced to the mixture to appear as a crystalline phase, and some as dispersed Pb in the amorphous phosphate matrix.

The sections below will present the compression strength, curing properties, density, gamma attenuation results, XRD and SEM characterization for Wollastonite based CBPCs with PbO contents.

2 EXPERIMENTAL

2.1 Samples manufacturing

CBPC samples were fabricated by mixing an aqueous phosphoric acid formulation and natural Wollastonite powder M200 (from Minera Nyco; see Table 1) in a 1.2 ratio liquid to powder. Also, lead (II) oxide (from Alfa Aesar; see Table 2) was added to the mixture. For all samples, the 1.2 ratio liquid (phosphoric acid formulation) to powders (Wollastonite + PbO) was maintained constant, see Table 3.

Table 1 Chemical composition of Wollastonite powder.

Composition	CaO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	MnO	MgO	TiO ₂	K ₂ O
Percentage	46.25	52.00	0.25	0.40	0.025	0.50	0.025	0.15

Table 2 Chemical composition of PbO powder.

Composition	PbO	Pb	Fe ₂ O ₃
Percentage	99.97	0.03	0.0002

Table 3 Raw materials amounts in the CBPCs composites samples fabricated.

CBPC-%PbO	Wollastonite (g)	PbO (g)	Total powders (g)	Phosphoric acid formulation (g)
CBPC	100	0	100	120
CBPC-2PbO	98	2	100	120
CBPC-10PbO	90	10	100	120
CBPC-50PbO	50	50	100	120

The mixing process of the components was conducted in a Planetary Centrifugal Mixer (Thinky Mixer[®] AR-250, TM). It has been found that when temperature of the raw materials is reduced, the pot life of the composite is increased and the viscosity is decreased, which affects the quality of the composite [14]. Thus, for compression and irradiation samples the raw materials were first stored in a refrigerator for an hour at 3°C in independent and closed containers.

On the contrary, for curing test samples, all raw materials were maintained at room temperature in order to see immediately the effect of the PbO on the pot life. On the other hand, it has been reported [14] that the pot life can be extended by different ways (by thermal processing, aging the raw materials, or putting additives in the raw materials).

2.2 Irradiation tests

Irradiation samples were fabricated using glass molds of 12.7mm diameter and 100mm long. Then, a diamond saw was used to cut samples discs of 0.1mm thick. Samples were then ground using silicon carbide papers of grit ANSI 400. Finally, samples were dried in a furnace at 100°C for 24 hours in order to stabilize the water content. Figure 1 shows the irradiation disks and the compression samples.



Figure 1 typical irradiation (disks) and compression (cylinders) samples.

PbO was used in the samples due to Pb being a common shielding material for most gamma sources. Pb exhibits a high attenuation of both gamma and x-rays and is widely used in shielding applications. In general high Z nuclei and high density materials are more effective at shielding gamma radiation. Pb is a stable high Z material, with high density, and relatively low price.

The samples were tested to determine the linear attenuation coefficients at various energies using a calibrated Europium source with isotopes of Europium-152, Europium-154, and Europium-155. This source presents 14 different peaks giving a wide range of gamma energies to study attenuation. We used six (6) energies. An example of this spectrum can be seen in Figure 2. The samples were then tested to determine the linear attenuation coefficients at various energies using a Europium source that consisted of isotopes of Europium-152, Europium-154, and Europium-155.

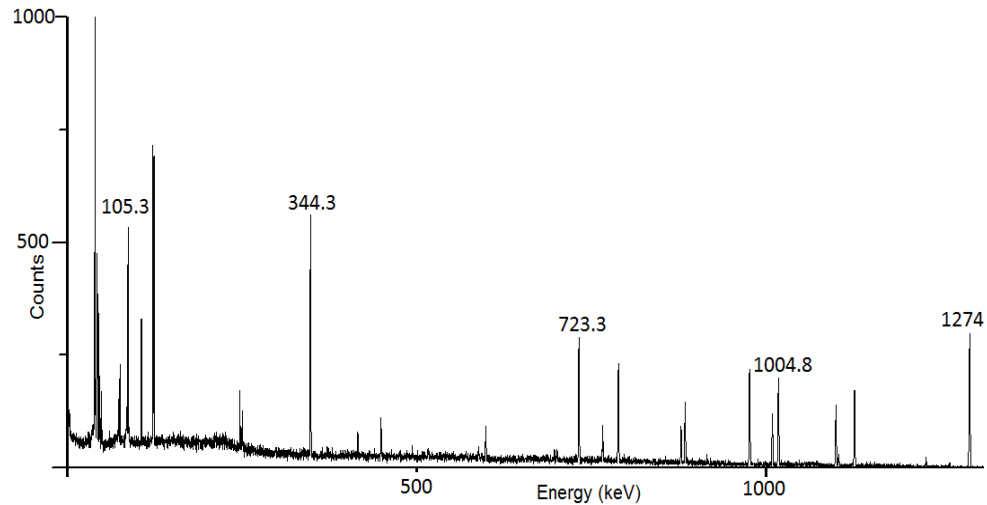


Figure 2 Europium spectrum

Calculating the linear attenuation coefficient is done by using Equation 1 where I_0 is the intensity of the uncollided beam, I is the intensity of the beam after it has passed through the sample, x is the thickness of the sample in cm, and μ is the linear attenuation coefficient measured in cm^{-1} .

$$I = I_0 e^{-\mu x} \quad (1)$$

The setup for irradiation with gamma rays with europium gamma source is represented in Figure 3a. A picture of the actual sample holder is also included in Figure 3b. A high purity Germanium detector was used to obtain peaks for the europium source. The first lead collimator is used to form a beam of gamma particles that will strike the sample and the uncollided particles will pass through and become detected by the High-purity Germanium detector. The particles that are scattered by the sample are absorbed in the second collimator if they are scattered at an angle. However, some slowing of the

particles does occur and these particles are removed from the analysis by subtracting the baselines of the γ -peaks. A picture of the actual sample holder is also included in the picture. All measurements are performed inside a heavily shielded facility (to reduce background) in the Missouri University of Science and Technology Research Reactor (MSTR).

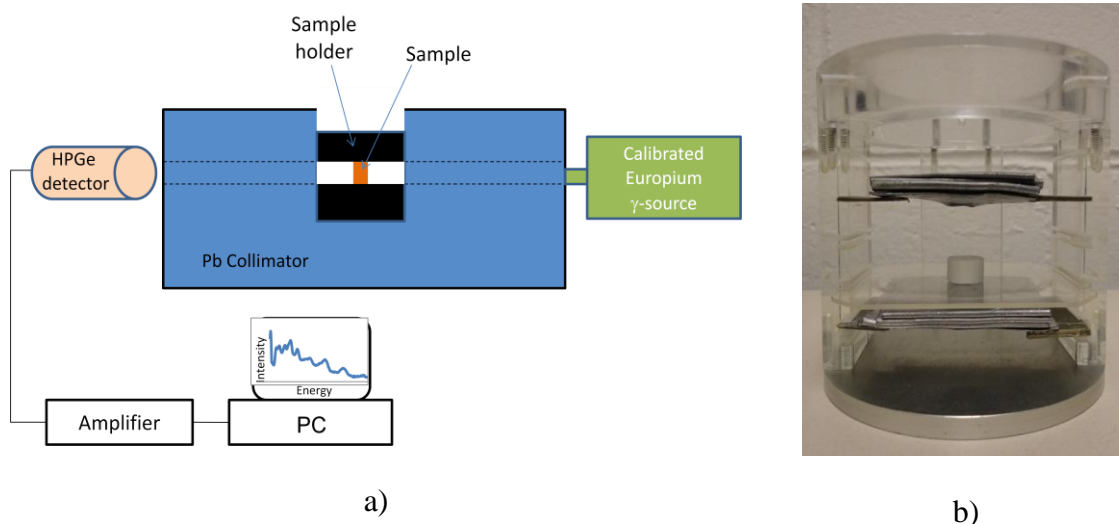


Figure 3 Irradiation setup for gamma rays, a) representation, b) detail of the sample holder.

2.3 Compression tests

For samples without PbO, the raw materials were mixed in the TM for 2min. For samples with PbO, first Wollastonite and the acidic solution were mixed one min, and when PbO was added, they were mixed one more min. Samples were fabricated using glass molds of 12.7mm diameter and 100mm long. Then, a diamond saw was used to cut cylinders of 28mm length. Samples were ground (with silicon carbide papers of grit ANSI 400) using a metallic mold until flat, parallel and smooth surfaces were obtained. The final length

was 25.4mm. Samples were dried in a furnace at 100°C for 24 hours in order to stabilize the water content. Compression tests were conducted in an Instron[®] machine 3382. A set of 5 samples were tested for each composition of Table 3. The crosshead speed was 1mm/min.

2.4 Other Characterization:

To see the microstructure, sample sections were initially ground using silicon carbide papers grit ANSI 240, 400 and 1200 progressively, then they were polished with alumina powders of 1, 0.3 and 0.05 μ m grain size progressively. After polishing, samples were dried in a furnace at 100°C for 24 hours and observed in an optical microscope. For SEM examination, samples were mounted on an aluminum stub and sputtered in a Hummer 6.2 system (15mA AC for 30 sec) creating approximately a 1nm thick film of Au. The SEM used was a JEOL JSM 6700R in high vacuum mode. Elemental distribution x-ray maps were collected on the SEM with an energy-dispersive X-ray spectroscopy analyzer (SEM-EDS). The images were collected on the polished and gold-coated samples, with a counting time of 51.2 ms/pixel.

X-Ray Diffraction (XRD) experiments were conducted using an X'Pert PRO (Cu K α radiation, $\lambda=1.5406$ Å), at 45KV and scanning between 10° and 80°. M200, M400 and M1250 Wollastonite samples (before and after the drying process) were ground in an alumina mortar and XRD tests were done at room temperature.

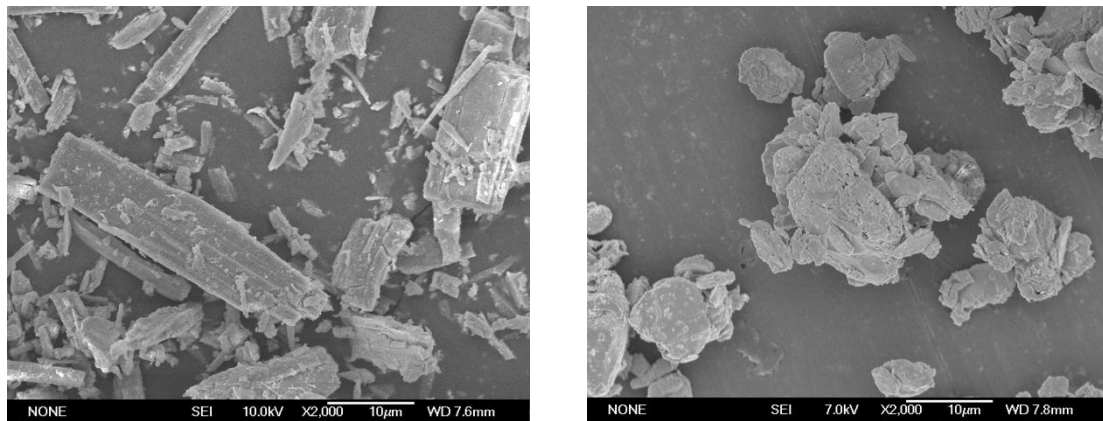
Density tests were conducted over CBPCs with Fly ash as filler. All samples were tested after the drying process described above, in a Mettler Toledo[™] balance, by means of the

buoyancy method. The Dry Weight (W_d), Submerged Weight (W_s), and Saturated Weight (W_{ss}) were measured. The following parameters were calculated:

Bulk volume: $V_b = W_{ss} - W_s$; Apparent volume: $V_{app} = W_d - W_s$; Open-pore volume: $V_{op} = W_{ss} - W_d$; % porosity = $(V_{op}/V_b) \times 100 \%$; Bulk Density: $D_b = W_d / (W_{ss} - W_s)$; and Apparent Density: $D_a = W_d / (W_d - W_s)$. In these calculations, density of water was taken to be 1.0 g/cm^3 .

3 ANALYSIS AND RESULTS

Figure 4a and b show Wollastonite and PbO powders respectively. Wollastonite grains are needle like shaped while PbO grains do not have specific shape.

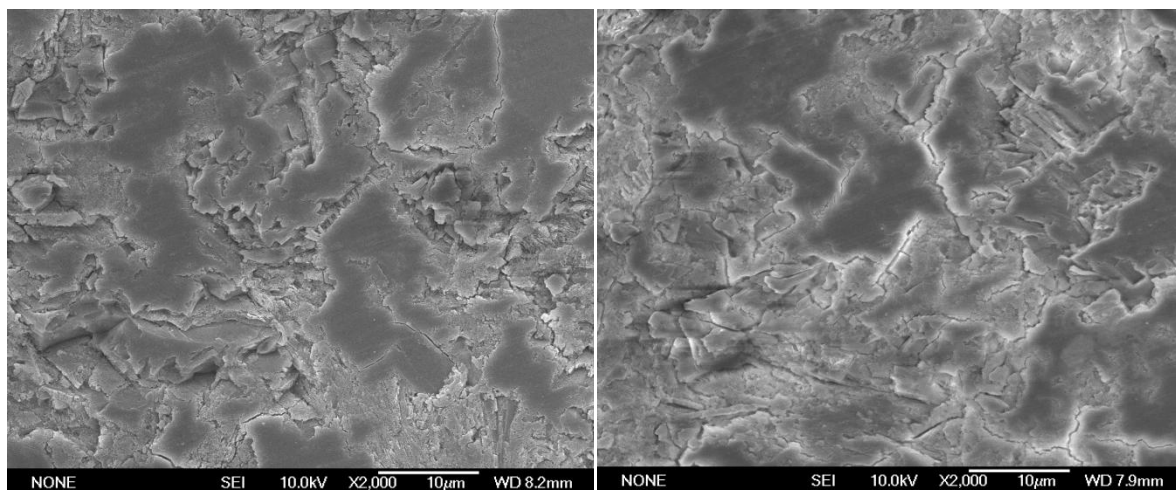


a)

b)

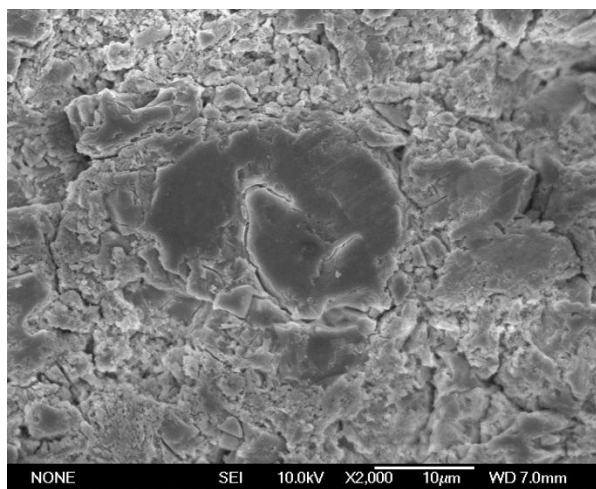
Figure 4 Powders used in the ceramic composite a) Wollastonite, b) PbO

Figure 5 shows SEM cross section images for Wollastonite based CBPCs taken at the same magnification. Figures 5a and b show Wollastonite-based CBPC with 0 and 10 wt%PbO respectively. The grains correspond to remaining Wollastonite, silica and remaining PbO.



a)

b)

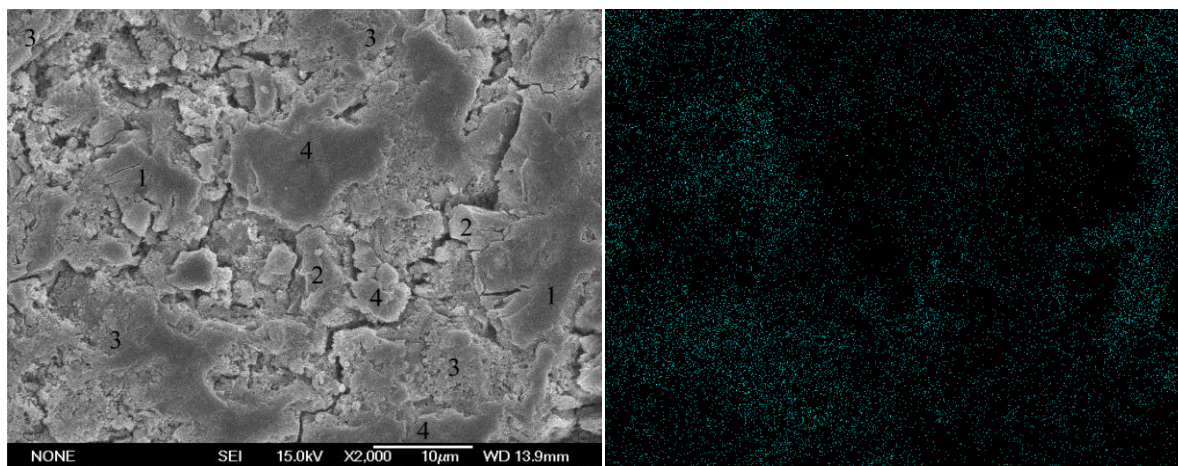


c)

Figure 5 SEM cross section images for Wollastonite based CBPCs, a) Wollastonite-based CBPC, b) CBPC with 10 wt%PbO, c) CBPC with 50 wt%PbO

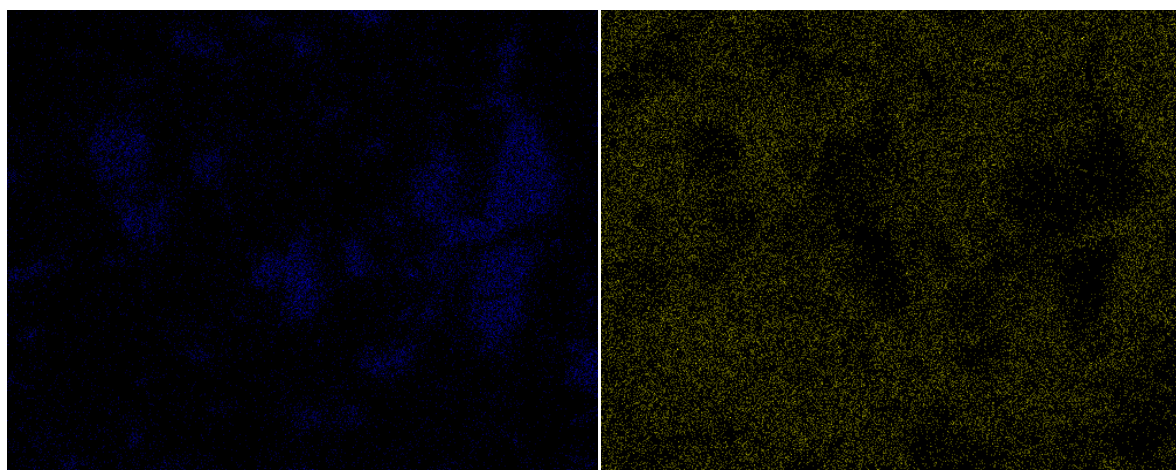
Figure 6 shows X-ray maps for the Wollastonite-based CBPC with PbO. Figure 6a shows a topographical image with phases named by numbers, which have been identified by using the compositional images of Figures 6b-6e. Silica and Wollastonite grains are visible in Figure 6b and c respectively. Figure 6c shows the calcium phosphate based

matrix. Figure 6d shows the P contents, mainly in the amorphous calcium phosphate matrix. Figure 6e shows the Pb content. Some remaining particles of PbO are present as well as PbO in the phosphate matrix which may indicate that some new amorphous phases (not visible by XRD) may be created by the dissolution of Pb in the phosphoric acid formulation. More research is being conducted over this topic.



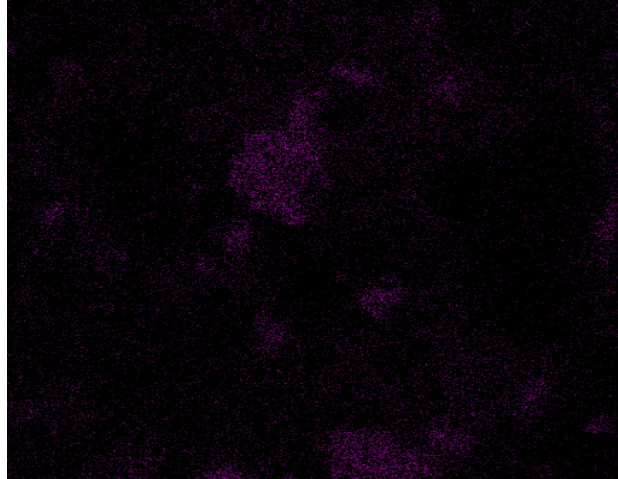
a)

b) Ca map



c) Si map

d) P map



e) Pb map

Figure 6 X-ray maps of CBPCs with PbO, a) topographical image; and b) Si, c) Ca, d) P and e) Pb compositional images respectively. Numbers 1, 2 3 and 4 on part a) correspond to Wollastonite, silica, amorphous calcium and lead phosphates and lead oxide respectively.

Figure 7a shows the compressive strength for the CBPCs with PbO contents. The standard deviation was bigger for the sample with 10%PbO. When either the concentration of Wollastonite or PbO were large, the error bars were low.

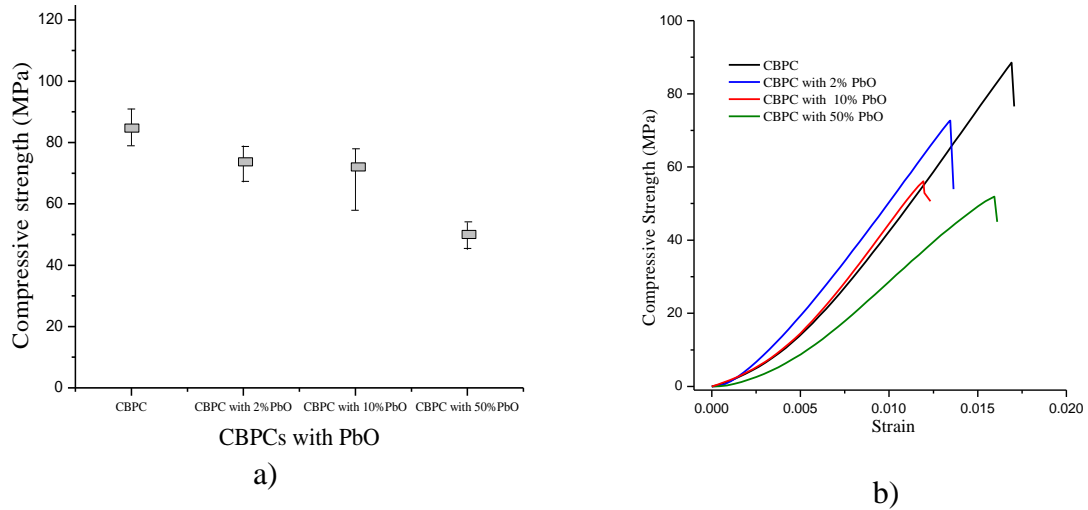


Figure 7 Compressive strength for CBPCs with fly ash, a) error bars, b) typical stress-strain curves

Also, in all cases the compressive strength for the CBPC composites was superior to those values for the regular Portland cement concrete (about 30MPa). Figure 7b shows typical compression curves for the CBPCs with PbO.

Figure 8 shows the XRD patterns for the CBPCs with PbO samples fabricated. The Wollastonite-based CBPC is a composite material itself with several crystalline (Wollastonite and brushite) and amorphous phases (silica and amorphous calcium phosphates) [15]. By adding PbO, only remaining PbO appears. The intensity of PbO peaks increases at the higher concentrations in the composite.

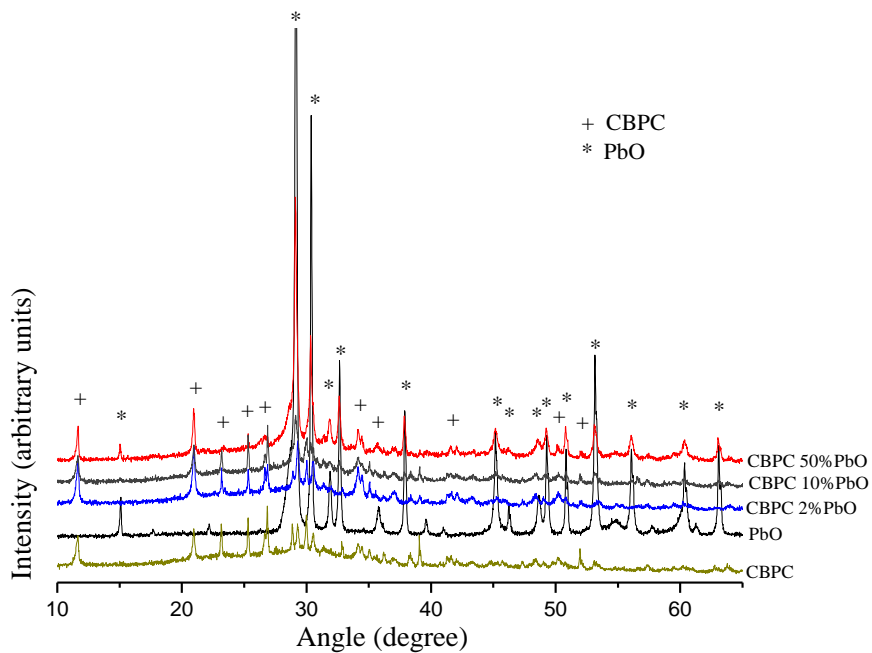


Figure 8 XRD patterns for Wollastonite-based CBPCs with PbO

On the other hand, density results (which are the mean of six measurements) presented in Table 4 show that in general when PbO was increased, the bulk density was increased and the percentage of porosity was decreased.

Table 4 Density tests for Wollastonite-based CBPCs with PbO

Sample		Wd (g)	Ws (g)	Wss (g)	Vb (cm ³)	Vap (cm ³)	Vop (cm ³)	Db (g/cm ³)	Da (g/cm ³)	% Por
CBPC	Mean	0.340	0.179	0.385	0.206	0.161	0.045	1.652	2.112	21.758
	SD	0.031	0.020	0.036	0.017	0.011	0.006	0.024	0.050	1.068
CBPC with 2% PbO	Mean	0.284	0.139	0.320	0.180	0.145	0.036	1.576	1.964	19.786
	SD	0.013	0.013	0.011	0.003	0.002	0.003	0.094	0.090	1.227
CBPC with 10% PbO	Mean	0.323	0.165	0.361	0.197	0.158	0.038	1.644	2.040	19.417
	SD	0.010	0.008	0.012	0.005	0.004	0.002	0.021	0.043	0.792
CBPC with 50% PbO	Mean	0.370	0.231	0.401	0.170	0.139	0.031	2.177	2.667	18.373
	SD	0.004	0.004	0.004	0.002	0.002	0.001	0.026	0.040	0.677

Figure 9 shows the gamma attenuation tests with europium in the CBPCs with different PbO contents. It is observed that the Europium is almost completely attenuated at low energies and that high energy gamma particles are less attenuated in each case.

The energy peaks were evaluated by integrating the area underneath the peaks (number of counts) for each energy. For the low energy peaks attenuation was complete and so the peaks were removed from the analysis. This is not an issue since a shield designed for high energy attenuation will also attenuate lower energies. In fact for the 50 percent PbO composition the attenuation reduced all peaks below 123.1 keV to background suggesting sufficiently high attenuation coefficients for energies below 344.3 keV. At higher

energies there is a decrease in the attenuation coefficients since the gamma particles travel through the sample with greater ease (as expected). Figure 10a shows the change in linear attenuation coefficients for the CBPC with 50wt% PbO. There is a substantial increase in the attenuation of gamma rays (more than 100%) reaching values useful for practical applications.

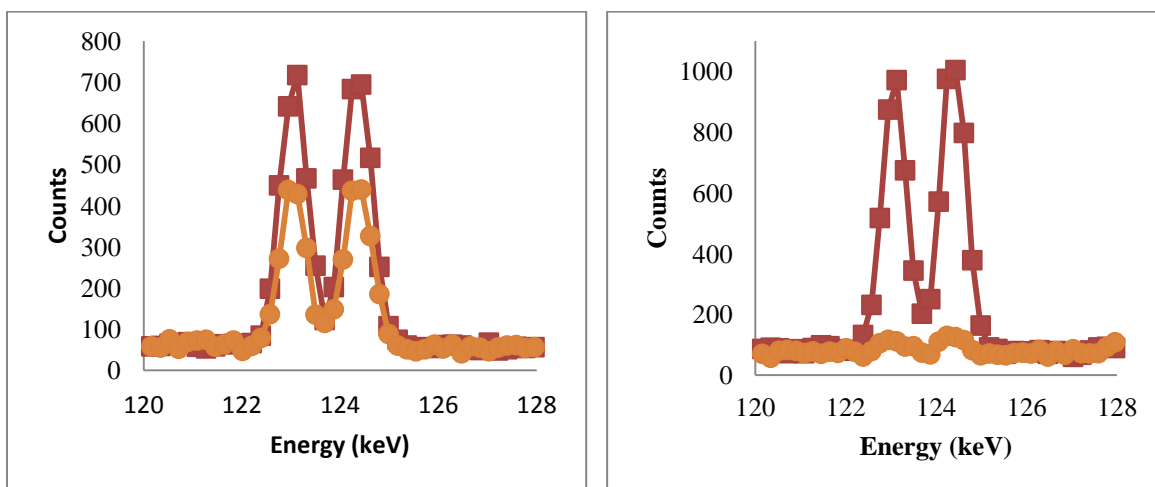


Figure 9 Attenuation tests with Europium in the CBPCs with different PbO contents. Figures a) and b) show a double peak around 121 keV for the 0% and 50% PbO samples respectively, the figures show the samples without any additives in a) and with 1% lead added in b) showing the change in the attenuation at low energies.

Figure 10a shows the change in linear attenuation coefficients for the CBPC with 50wt% PbO. There is a substantial increase in the attenuation of gamma rays (more than 100%) reaching values useful for practical applications.

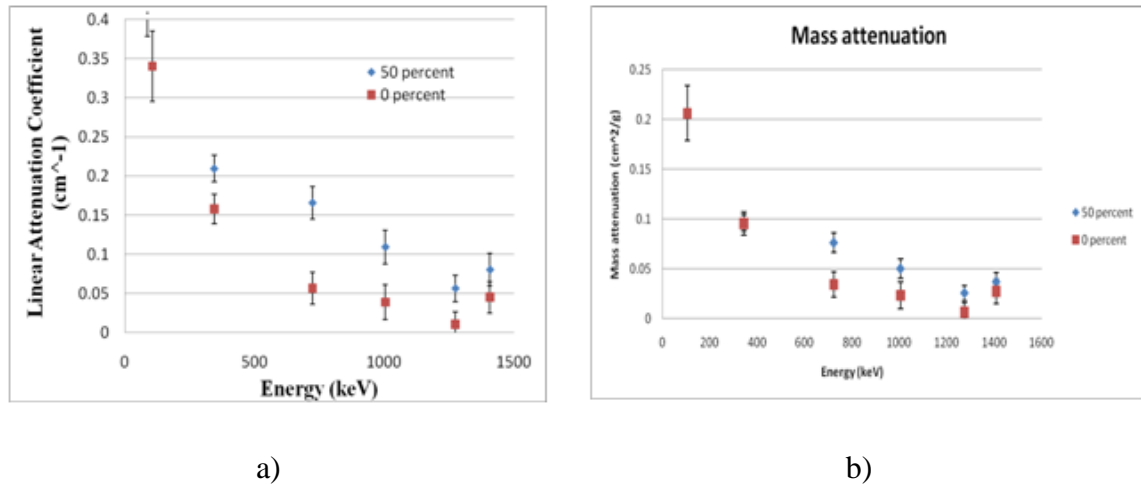


Figure 10 a) Linear attenuation coefficient, b) mass attenuation coefficient.

The manufacturing of Wollastonite based CBPCs with PbO contents has been successfully made in this research. Several previous results showed that changing the mixing order of the components has a significant effect on the final properties. It has been found that mixing first the components of the Wollastonite-based CBPCs and then putting the PbO contents decreases the variability of the compressive strength when it is compared with samples where all components were mixed at the same time. Also, the SEM shows that Pb was dissolved into the amorphous phosphate matrix, probably producing amorphous lead phosphates mixed with amorphous calcium phosphates and silica, products of the wollastonite-based CBPCs.

Other materials that appear in the composites are crystalline: Wollastonite and probably brushite. For the compression tests, it was observed that the scattering was low in general for CBPC-PbO samples when compared to other ceramics and cements. Also, that compressive strength for the CBPC composites was superior to those for the regular Portland cement concrete.

With respect to the shielding capabilities, the addition of Pb to the samples increased the linear attenuation coefficient substantially. At 1 MeV the value almost triples (from 0.039 to 0.109 cm^{-1}). The linear attenuation coefficient of our 1 at. % doped CBPC reached 14% of the value for lead (0.109 vs 0.77 cm^{-1} for lead) up from 5% for the ceramic without doping. Part of this increase is due to intrinsic increase of the mass attenuation coefficient due to using a higher Z material (Pb) and part is due to the increase in apparent density (Da increased from to 2.1 to 2.7 g/cm^3). Also notice that due to the weight of Pb 1 at. % Pb is equivalent to 11.4 wt. % Pb. The net improvement of attenuation coefficient for 1MeV gamma radiation was 180.9% (the improvement at different energies was 32.4, 193.8, 180.9, 44.9, and 78%).

When comparing CBPC to other common gamma shields a few things are noticed. It is clearly equivalent to both concrete and iron for stopping low energy gammas. With the addition of the PbO the mass attenuation coefficient compares favorably to other common gamma shields as shown in Table 5. At .1 MeV the mass attenuation coefficient is better than concrete yet doesn't perform as well as Iron or Lead. At higher energies the coefficient performs equally with iron and concrete. When the error calculations from the measurements are factored into this table CBPC behaves as well if not better than concrete.

Table 5 Mass Attenuation Coefficients for some common Gamma Shields (16)

Mass Attenuation Coefficient (cm ² /g)					
Energy (MeV)	CPBC	CPBC (50% PbO)	Concrete	Iron	Lead
0.1	0.161		0.167	0.334	5.337
0.3	0.096	0.096	0.107	0.105	0.373
0.8	0.034	0.076	0.071	0.066	0.084
1	0.024	0.050	0.064	0.060	0.068
1.5	0.027	0.037	0.052	0.049	0.051

The improvement of the linear attenuation coefficient implies that the 11.43cm (4.5”) to 14.61cm (5.75”) gamma shield made of lead in some nuclear casks ([10] and [13]) can be replaced with a ceramic shield of our material of 84cm (33.1”) to 107cm (42.1”). This is still big for transportation applications, but it is sufficient for above ground storage. The results are encouraging to pursue further improvements.

4 SUMMARY AND CONCLUSION

The effect of PbO in the attenuation coefficient and compressive strength of Wollastonite based-CBPCs has been presented. In all cases, Wollastonite and the acidic solution were mixed first, and then, PbO was added. It was found in this research that mixing in this order extends the pot life when is compared with the samples made by mixing all the components at the same time. On the other hand, in all cases the compressive strength for the CBPC composites was superior to those values for the regular Portland cement concrete (about 30Mpa). Even in the case of CBPC with 50%PbO. These results show promissory this class of materials not only to be a superior radiation shielding solution but also to play an important role in the infrastructure of new nuclear plants.

Also, it is important to emphasize that the Wollastonite-based CBPC is a composite material itself with several crystalline (Wollastonite and brushite) and amorphous phases (silica and amorphous calcium phosphates as was confirmed by the SEM and was reported before [14]). The XRD showed that by adding PbO, only remaining PbO appears. X ray maps showed that there is Pb contents dissolved in the amorphous calcium phosphate matrix, which can be in the form of amorphous lead phosphates. This characteristic enables this materials to encapsulate different nuclear wastes since it can tailored the composition of the matrix by introducing new species and also it can be controlled the amount of phases and it distribution to optimize other properties like mechanical ones for structural applications. Since these CBPCs start from a liquid (mixture of calcium oxides with a phosphoric acid formulation) solution, almost any reinforcements can be introduced, which could enable these ceramics with wastes (introduced the microstructure [5]) to be basements for specific infrastructure developments.

On the other hand, it was obtained in this research that the attenuation coefficient was substantially improved by the incorporation of Pb in our CBPCs. The improvements at energies above 300 keV ranged from 32 to 193.8%. Further improvements are necessary to optimize the composition and density of the ceramic such that improved strength is achieved with maximum incorporation of Pb into the ceramic matrix. Finally, a procedure to maximize the amount of Pb in the material that minimally degrades its mechanical and thermal properties, and also to incorporate boron as neutron shielding material is under development and will be published in a journal in future.

5 ACKNOWLEDGEMENTS

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PAPER II

RADIATION SHIELDING SIMULATION FOR WOLLASTONITE-BASED CHEMICALLY BONDED PHOSPHATE CERAMICS

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ABSTRACT

Neutron and gamma attenuation on Wollastonite-based Chemically Bonded Phosphate Ceramics (Wo-CBPCs) is studied with Monte Carlo simulation (MonteCarlo N-Particle, MCNP). Wollastonite-based CBPC is a composite material with several crystalline (Wollastonite and brushite) and amorphous phases (silica and amorphous calcium phosphates). The effect of lead incorporated into the ceramic as a gamma shield has been examined across various gamma energies as well as neutron attenuation of the ceramic. Besides this simulation effort, neutron activation analysis was also performed experimentally at the Missouri University of Science and Technology Reactor (MSTR) to determine the activation by neutrons of the ceramic. In the past, it was found that the attenuation coefficient was substantially improved by the incorporation of Pb in the Wo-CBPC. The improvements at energies above 300 keV ranged from 32 to 193.8%¹. In the

present simulation, we found that the addition of PbO increased the neutron attenuation by 180 to 400% and accurately predicted gamma attenuation values for photon energies above 1000 keV.

1 INTRODUCTION

Chemically Bonded Ceramics (CPCs) are inorganic solids generated by chemical reactions at low temperatures. This method avoids high temperature processing which allows the ceramics to be processed inexpensively in high volume production. The ceramics have also been shown to have high thermal shock resistance and have been used in firewall applications², nuclear waste and encapsulation³, electrical materials⁴, as well as composite materials⁵. This research focuses on the simulation of previous formulations with MCNP simulations to evaluate the effectiveness of the experiment in determining linear attenuation coefficients and to determine how to optimize the neutron attenuation values. Various models have already been used to determine shielding properties of concrete with additives^{6,7}, as well as designing optimal shielding properties of concrete ratios⁸. Also there has been studies examining both gamma ray and neutron attenuation of composite materials in MCNP simulations as well as optimization studies on ceramic samples to improve their shielding properties^{9,10}. The linear attenuation value is an important parameter to calculate the maximum amount of radiation released from a shielded source. In order to make sure that the public remains below their dose limit an analysis of linear attenuation coefficients for gamma with both experimental data and MCNP simulations was performed as well as MCNP simulations for neutron attenuation.

In a previous paper it was found that incorporating PbO into CBPC improved the linear attenuation coefficient for gammas from 32 to 193.8%¹ at different energies. The current simulations will help us improve the formulation for both gamma and neutrons should other additives be incorporated in the structure.

2 EXPERIMENTAL DETAILS

In a previous study an experiment was performed using a calibrated Europium Source that contained Europium-152, Europium 154, and Europium-155 which provided a wide range of energies to test for Linear Attenuation coefficients. Due to decay of the Europium source only 7 energy values were evaluated due to the clarity of their peaks. A high purity Germanium Detector was used to determine the energy values of the peaks. The lead collimators were placed to generate a gamma beam and can be seen in Figure 1 for both the theoretical set-up and the actual set-up.

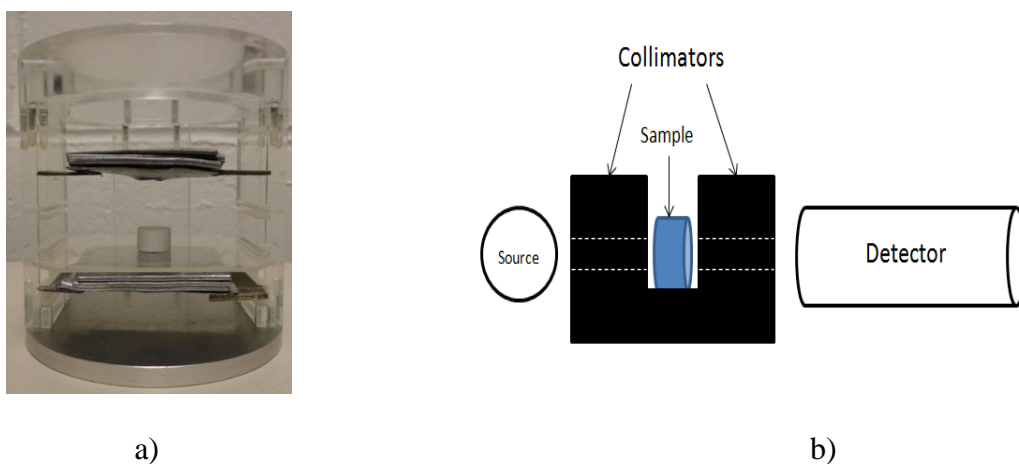


Figure 1. a) experimental set-up for Gamma Rays, b) representation

The linear attenuation coefficient is determined from equation 1 where I_0 is initial flux, I is flux with sample included, x is the thickness of the sample in cm, and μ is the attenuation coefficient in cm^{-1} .

$$I = I_0 e^{-\mu x} \quad (1)$$

Using this equation one can determine the linear attenuation coefficient by solving for μ in cm^{-1} . One thing to note about this is that this equation is energy dependent, different energies result in different linear attenuation values and therefore a graph of μ versus energy or tabled values are regularly used to present the information. Using these values one can determine the necessary thickness of shielding for a variety of radiation energy sources.

The energy spectrum for the calibrated Europium source is shown in Figure 2.

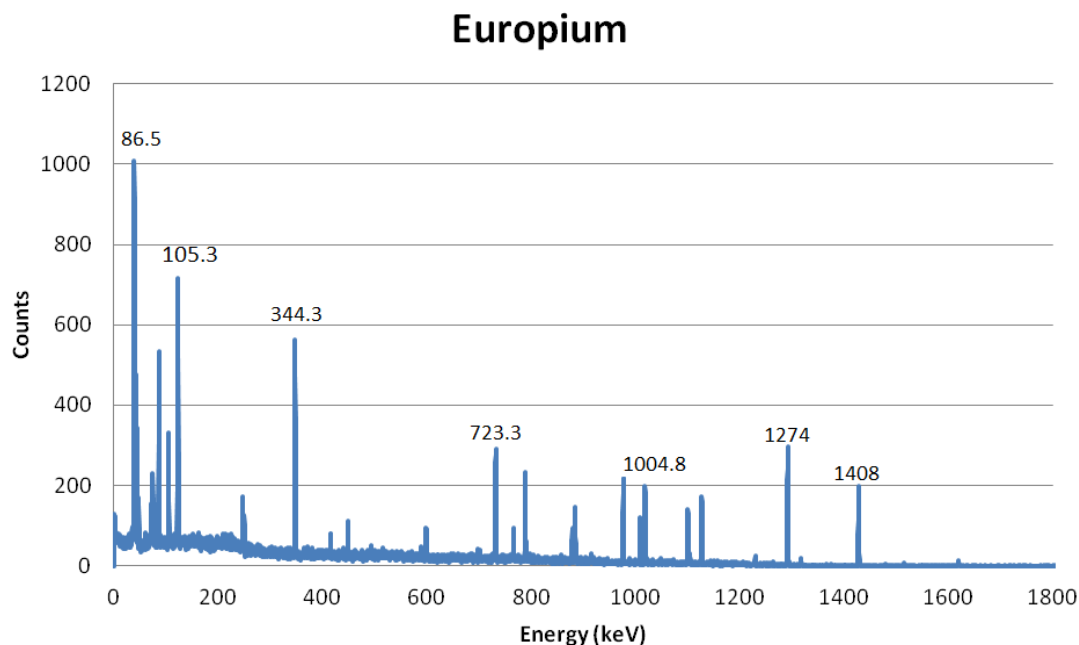


Figure 2. Europium Energy Spectrum

Using the Europium spectrum one can obtain many different energy peaks that can be used to determine a linear attenuation graph for various materials. Once this graph is constructed one can use equation 1 to determine the minimum thickness of a radiation shield to achieve the necessary requirements for dose limits. In order to determine the peaks the results from the experiment integrated from the peak height across 10 energy channels with an average width of 0.1 keV.

3 SIMULATION DETAILS

MCNPX was used for simulating gamma attenuation by the CBPCs. MCNPX is a general purpose Monte Carlo transport code that can track thousands of particles over large energy ranges(0 to 150MeV)¹¹ and has been benchmarked for various uses¹². The simulation used the same basic set-up as the experiments, however the source was

approximated as a point source due to the difficulty in determining the exact location of the various europium particles in the source. The samples used were 2 cm thick samples of the same cylindrical shape of the manufactured samples. The samples were estimated to be homogeneously mixed. In order to add porosity of the substance into the simulation, the calculated porosity of the sample was used to add in air pockets to the sample, however for calculating the linear attenuation the initial thickness of the sample was used. The model for CBPC with and without PbO added is summarized in Table I.

Table I. MCNPX inputs

Height (cm)	Porosity (%)	Modeled Height (cm)	Density (g/cm ³)	Diameter (cm)	Atomic Composition (%)
2	21.76	1.56484	2.112	1.1	47.4- O 36.8- H 5.2- Si 5.2- Ca 5.2 -P
2	21.76	1.56484 cm	2.667	1.1	41.2- O 32.6- H 4.6- Si 4.6- Ca 4.6 -P 11- Pb

For the neutron simulations a similar setup as the measurements for the gamma ray detection was used. A neutron beam of varying energies from thermal to fast (1/40eV to 2.5 MeV) was used to determine the attenuation coefficient for the ceramic. Since reactors are usually used to determine neutron shielding details¹³ and we plan to measure the neutron attenuation using a Pu-Be neutron source we studied the shielding capabilities at energies up to 2.5 MeV. The same compositions as well as the air pocket addition for porosity were included in the simulations.

The model was examined for gamma attenuation energies for a Europium source containing 7 energy peaks at 86.5, 105.3, 344.3, 723.3, 1004.8, 1274, and 1408 keV. These give a wide spectrum of energies useful for obtaining a linear attenuation graph.

For the 7 neutron energies chosen ranging from thermal energies to 2.5MeV, 500 million particles were run in order to reduce the error of the MCNP simulation for each run.

4 ANALYSIS

The experiment used a Europium Source to determine the linear attenuation coefficient for Chemically Bonded Phosphate Ceramics (CBPCs). The original experiment was also examined in MCNPX to verify the results of the measurement¹. The measured porosity of the sample was included in the calculation as additional uniformly distributed air pockets. The results measured the flux over 10 energy channels averaging 0.1 keV for each of the initial energies from Europium to obtain the uncollided flux of the particles. The results from both MCNP and the actual measured values are plotted in Figure 3 for CBPC without lead added.

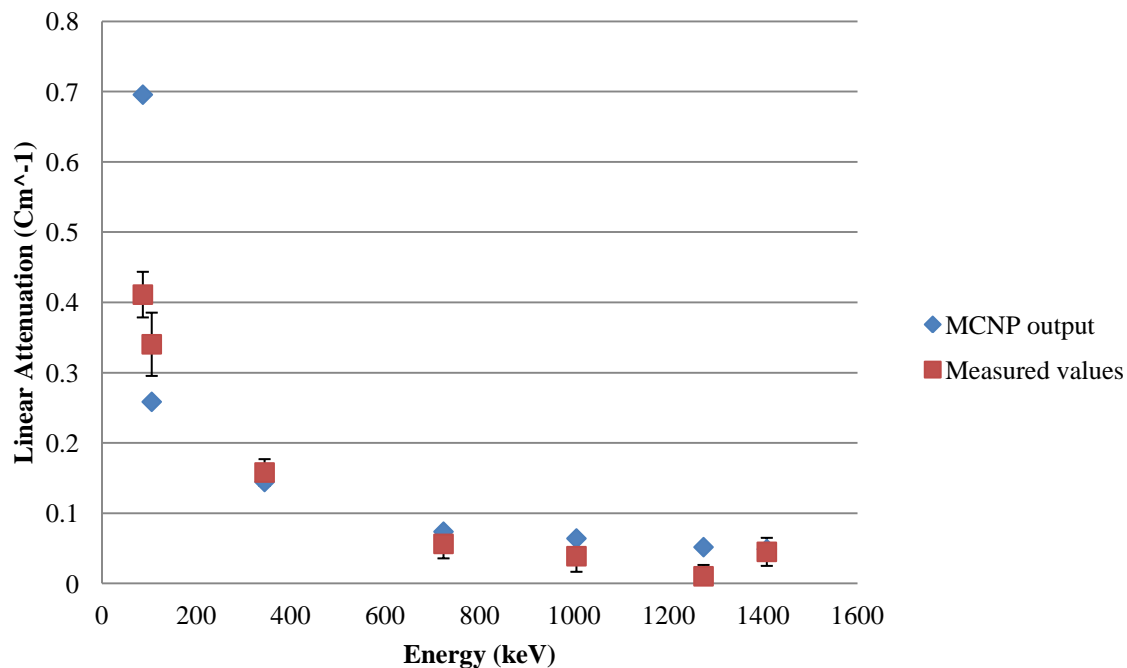


Figure 3. MCNP output and measured Values for a Europium Source and CBPCs with no PbO content

The simulations were performed with MCNPX evaluating the energy peaks within a 10keV range of the initial energy. Porosity of the substances was included to obtain results that were similar to measured results for the Europium gamma source used. For most values the MCNPX output was slightly higher than the measured value which is likely due to the porosity of the CBPCs since a change of just 1% porosity causes a 5% change in attenuation values. For the lower energy values ($E < 1000$ keV) values are close to the Compton edge which could cause differ notably from the calculated values. The error for the simulations was around 0.025% for all of the simulated values. For 1274.5 keV there is a large difference between the simulated and measured values. This had been noted in the previous study that the data point obtained appeared to be an

outlier¹ causing the discrepancy between simulated and actual values. The simulations were also performed for CBPCs with PbO added and are shown in Figure 4.

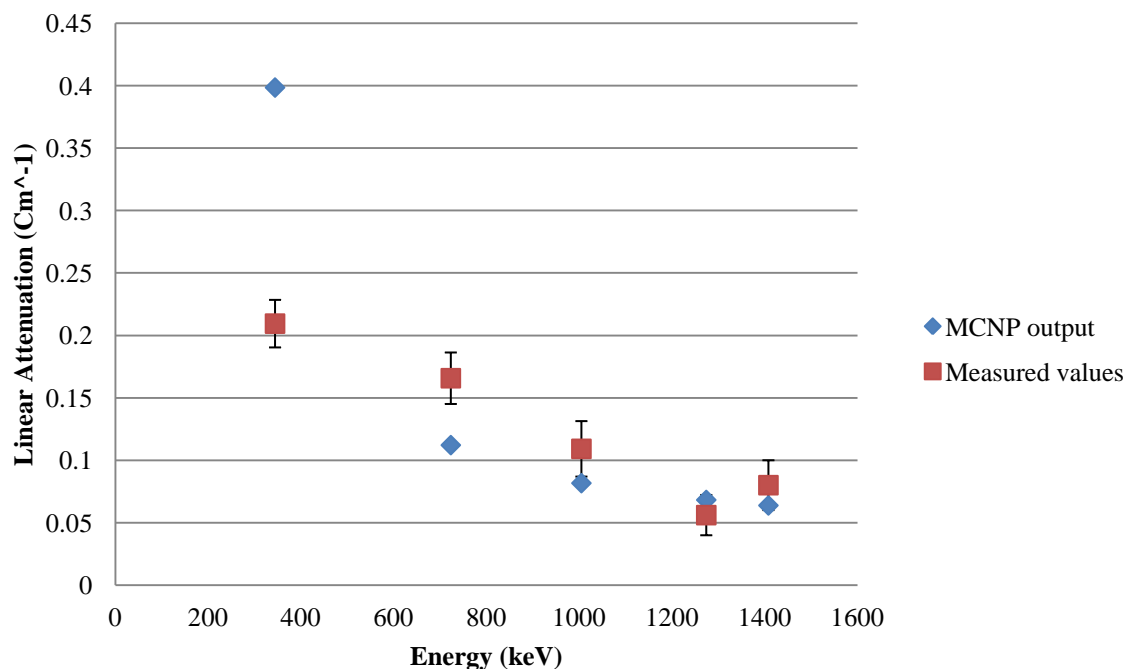


Figure 4. MCNP output and Experimental measured values with 50 wt% PbO added in

One thing to note is that there is only five gamma peaks in this sample set as compared to the content the CBPC without PbO added in. The lower energy peaks (XX keV and XX keV) are missing since they had been reduced to background and a gamma peak could not be determined in the low energy ranges thereby removing the data points. The MCNP outputs for 50 wt% PbO mixture added into the ceramic tended to be lower than the actual measured values of the ceramic excepting the 1274.5keV and 344.3keV energy peaks. For the 344.3keV the likely cause is due to the scattering of higher energy

peaks since the initial calculation required over 20 energy channels to be integrated together to obtain the peaks as well as baseline subtraction to eliminate background noise, this would result in a measured value that is lower than its actual for lower energy gammas. At 1274.5keV the energy values are within one standard deviation of each other which suggest that the simulation is accurately predicting the measured results. For the most part as the energy increased the differences between the simulations predictions and the measured results became smaller which suggests that there were errors in the simulation of the linear attenuation coefficients likely due to differences in the porosity of the simulated CBPCs and the experimental. These errors are likely due to accidental beam adjustment as well as scattering from higher energy gamma rays. The scattering error cannot be adjusted for, however making sure that the gamma source remains stationary in between sample loadings would improve the accuracy of these measurements.

For the neutron attenuation test with MCNP varying energies ranging from thermal to fast were used to determine the attenuation coefficients for neutrons. The energy spectrum for CBPC without any PbO and as well as the spectrum with PbO included is shown in Figure 5.

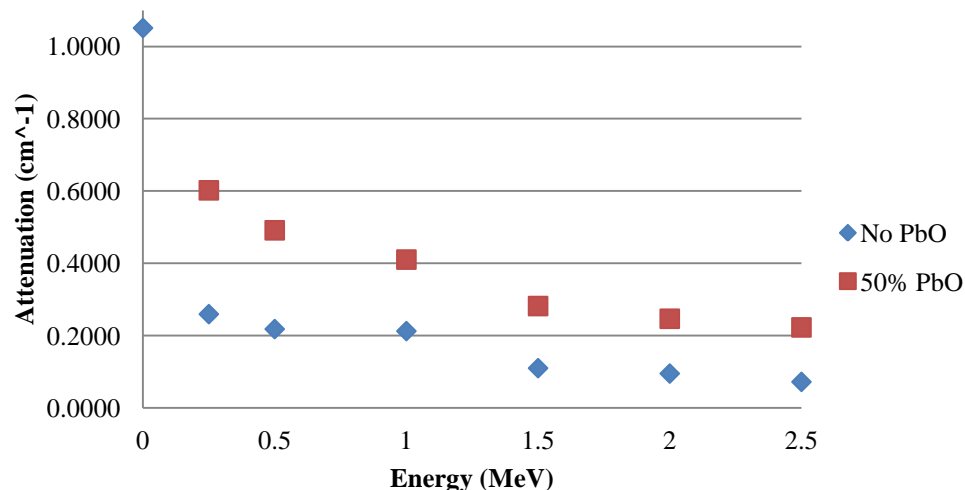


Figure 5. Neutron Linear Attenuation for CPBC

As can be seen CPBC attenuates neutrons very well with a value above 1 for thermal energy neutrons. When PbO was added to the system, the thermal energy attenuation increased to 4.8 cm^{-1} increasing by about 400%. At the higher energy values for fast neutrons CPBC performs decently with most values between 0.2 and 0.3 cm^{-1} . The addition of PbO caused an increase ranging from 180 to 300% for the attenuation values.

For The 50 wt% PbO there was a 458% increase in the attenuation of thermal neutrons and around a 200% increase in the attenuation of fast neutrons. This increase corresponds similarly with the increase of the gamma linear attenuation values. PbO values from 0.5 to 2.5 MeV all exist in the resonance region for Pb, it is likely that this region is causing issues with the output (sudden changes in attenuation values). For the other major elements (Si, Ca, O, and P) they are likewise in the resonance region however due to the lower cross-sections for these elements there is no effect observed. The likely cause for the increase in attenuation is due to the increased density of

hydrogen which has the highest cross section for the elements in this region which resulted in improved neutron attenuation.

5 CONCLUSIONS

From the calculations and validation experiment performed it is clear that MCNP can be used to predict linear attenuation coefficients for ceramics so long as the porosity and density are well known and taken into account correctly. It is believed that the variations in the values of the coefficients is caused by errors in both the experiment and the porosity added into the simulation. With just 1% increase in the porosity the attenuation decreased by 5%. Other than low energy outliers due to inherent uncertainty in the experimental measurements MCNP can arrive at close (20% or lower) to the experimental values for linear attenuation coefficients.

For the gamma attenuation simulations, MCNP arrived within 0.02 cm^{-1} to the measured values for the higher energy gammas and was off anywhere from 0.1 cm^{-1} to 0.3 cm^{-1} in predicting low energy values. This is likely due to issues that were created by the experimental procedure and apparently not due to issues with the simulation. When PbO was added there were some issues in predicting the values obtained from the experiment for energies below 1000 keV. This is also likely due to inherent uncertainty in the measurements.

The increase in the neutron attenuation for CBPC is likely due to the increase in the density and total composition of hydrogen in the simulations. Since Hydrogen has a very large cross section compared to the other elements in the CBPC the overall increase in the total attenuation is due to the increase in the overall density of hydrogen in the

sample. The addition of PbO to CBPC showed a substantial increase in both measurements of the attenuation coefficient for both neutrons and gamma shielding purposes.

The results from the research show promising results for the improvement of shields made of CBPCs for both gamma and neutron attenuation with the addition of additives. By adding other additives and observing the changes in shielding properties it is hoped that the shielding capabilities of this ceramic can be optimized even further.

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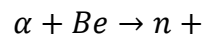
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PAPER III

Neutron Attenuation Measurements of Chemically Bonded Phosphate Ceramics

1 Introduction

In order to determine the shielding effect of CBPCs a method for measuring the neutron attenuation for thermal energy neutrons was developed. In order to allow for quick measuring of the attenuation coefficients a He-3 neutron detector was used. This detector was connected to a lower level discriminator to reduce noise and a scaler to obtain counts. A PuBe neutron source was used at the Missouri University of Science and Technology Reactor. The production of neutrons from the PuBe sources occurs when the alpha particle released from the plutonium react with beryllium in the source producing neutrons according to the reaction shown in Equation 1.



This produces neutrons with a wide energy range; the neutron energy spectrum for PuBe can be seen in Figure 1.

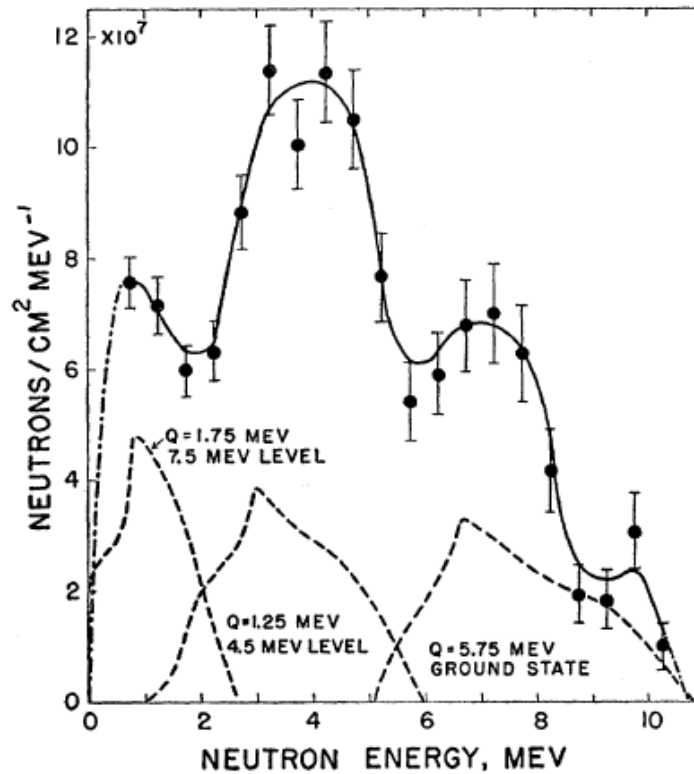


Figure 1: PuBe Neutron Energy Spectrum [1]

Since the spectrum of PuBe contains more fast neutrons than thermal neutrons a method for thermalizing the neutrons was used. The average distance for thermalizing a fission neutron in water is about 27cm^2 [2]. Therefore the source was suspended in water in a container of low density polyethylene with a distance to the wall of water of 9cm. The sample was placed on a platform with the edge of the sample being between 1 to 2 cm away from the polyethylene wall. The samples were then measured using a He-3 detector with 10 minute count rates that were measured with and without a 0.6mm cadmium shield placed over it. The cadmium shield absorbed the thermalized neutrons that got through the sample and the difference measured gave the neutron attenuation for the samples. A picture of the setup is shown in Figure 2.

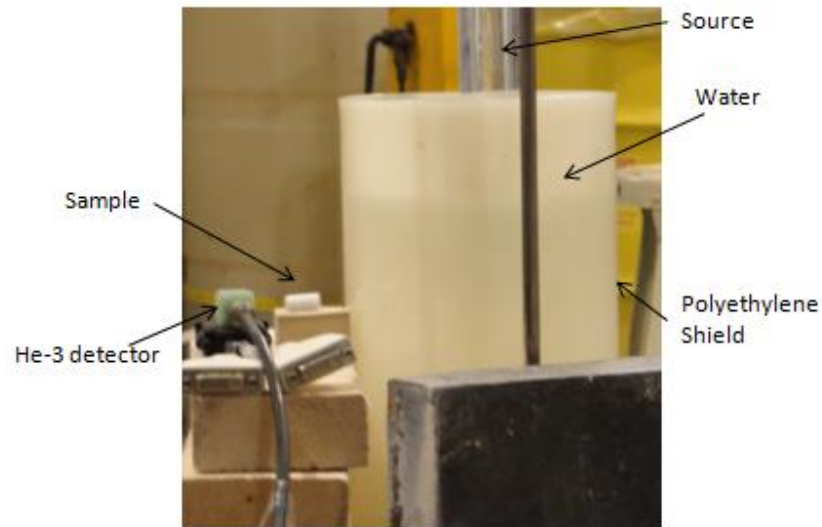


Figure 2: Neutron Attenuation Setup

2 Results and Analysis

After performing the experiment for several different CBPCs the results are summarized in Table 1.

Table 1: Thermal Neutron Attenuation Values for CBPCs

	Thickness (cm)	Linear Attenuation (cm ⁻¹)	Error
20% Boron	2.774	0.089	0.017
0%	1.664	0.060	0.018
50% PbO	1.284	0.045	0.023

From the results the addition of boron improved the attenuation of CBPCs by 48% for thermal neutrons. However the addition of PbO caused a notable decrease in attenuation

decreasing the value by 25%. However these results have fairly large error values still and further testing on neutron attenuation is recommended.

One of the reasons the error is so high is likely due to the continued thermalization of neutrons from the sample. While the sample is blocking lots of the neutrons some are likely still being thermalized by the sample. Another possible error is that the source location could have variations in its position. This would come about since the source had to be repositioned several times over the course of the experiment due to making a safer working environment.

3 Conclusions

The addition of Boron Oxide to CBPC shows significant improvement in neutron attenuation of the ceramic, however the addition of PbO to the ceramic showed a significant drop in neutron attenuation. For applications of this ceramic as a shielding material a balance between Boron Oxide and Lead Oxide needs to be determined in order to function as an effective neutron radiation shield.

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CONCLUSIONS

While the PbO based CBPC showed definite improvement in the linear attenuation coefficient for gamma irradiation (32 to 193.8%), it also showed a significant decrease by 25% for neutron attenuation. For the current cask designs to be replaced this would mean that a shield of 88 to 134 cm would have to be used which would be unusable in current cask design.

Boron Oxide based CBPC showed a significant increase in its ability to attenuate thermal neutrons with an improvement in attenuation of 48% over the base ceramic however the increase in attenuation still needs to be examined in greater detail.

VITA

Jason Joseph Pleitt was born on August 26, 1987 in Lombard, Illinois. The son of Joseph and Denise was raised in both Lombard and Warrenville Illinois graduating from Wheaton Warrenville South High school in May of 2006. He then attended Missouri University of Science of Technology and earned his Bachelor's in Nuclear Engineering in 2006. Wanting to continue his education he remained at Missouri University of Science and Technology and earned his M.S. in Nuclear Engineering in May 2012.