

**CERAMICRETE: A NOVEL CERAMIC PACKAGING SYSTEM FOR
SPENT-FUEL TRANSPORT AND STORAGE**

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ABSTRACT

This presentation summarizes our efforts to develop and apply chemically bonded phosphate ceramic (Ceramicrete™) technology for radiation shielding applications. The specific application being targeted is a packaging system for spent-fuel transport and storage. Using Ceramicrete technology under ambient conditions, we can produce dense and hard ceramic forms that incorporate second-phase material. Ceramicrete inherently is a superior shielding material because it contains large amounts of bound water in its crystal structure and can be cast in any shape.

A parametric study was conducted on Ceramicrete that contained second-phase additions of metals and other ceramic powders. Results of various standardized tests that included mechanical performance and shielding from neutrons are presented. The fabrication of complex shapes and structures by Ceramicrete technology is discussed. Ceramicrete is compared with other currently available shielding systems that are based on concrete and polymers.

INTRODUCTION

Over the past several years, room-temperature-setting Ceramicrete technology has been developed at Argonne National Laboratory-East (ANL-E) as a part of the Department of Energy-(DOE-EM) Mixed Waste Focus Area to treat and stabilize mixed wastes that contain volatile (RCRA) and radionuclide contaminants. This technology has been successful in stabilizing RCRA metals and radionuclides (^{238}U , ^{241}Am , ^{137}Cs) and has been implemented for field application at ANL-West. In addition, Ceramicrete™ technology has been scaled up to allow fabrication of 55-gal waste forms from soil wastes. Currently, these immobilization systems are being used to stabilize various low-level mixed wastes throughout the DOE complex and in the commercial sector.

Ceramicrete, a phosphate-based material formed by acid-base reactions under ambient conditions, can attain strengths almost three times that of cement concrete. As demonstrated by our previous work, because the Ceramicrete process is aqueous-based, it can incorporate high loadings of second-phase additions; hence, it can be tailored to exhibit properties that are based on the intended end use.

Because Ceramicrete contains significant amounts of bound water and boron, it is expected to effectively slow neutrons and thus provide attenuation of neutrons. Moreover, Ceramicrete can incorporate high loadings of heavy metals, a desirable capability for gamma-ray shielding. In

this regard, high-density borated Ceramicrete can lead to a low-temperature, cost-effective technology for providing nuclear shielding. Thus, the objective of this study was to demonstrate the use of Ceramicrete technology in conjunction with boron compounds to develop low-temperature-forming components and/or products with high radiation-shielding capacity.

FABRICATION OF BORATED CERAMICRETE

Ceramicrete is fabricated by acid-base reactions [1,2]. Its binding phase is obtained by reacting calcined magnesium oxide powder with a solution of dibasic potassium phosphate. The reaction that occurs can be represented by



The resulting phase (known as MKP) is extremely stable, with a solubility product of 2.4×10^{-11} under ambient conditions [3]. During the reaction, water enters the structure as water of hydration. In addition, fly-ash up to 50 wt.% is added to the binder mix to enhance the structural integrity of the set product.

Fabrication of bench-scale forms involved additions of various second phases including boron carbide and hematite. The added isotopic boron carbide was fabricated and supplied for this work by Eagle Picher. The neutron cross section of ^{10}B is ≈ 4000 barns for slow neutrons; hence, it is an excellent element for attenuation of neutrons. Similarly, the objective of hematite additions was to enhance photon attenuation properties of the set Ceramicrete product.

Processing involved mixing the dry powders first, then adding water and mixing with a spatula to form a homogeneous slurry. This slurry was mixed for ≈ 20 min and then poured into plastic molds. The slurry hardens into a hard ceramic in ≈ 2 h. The samples were allowed to cure for two weeks before they underwent testing. In the final set product, net ^{10}B was ≈ 7 wt.% and bound water was 21 wt.%.

PHYSICAL PROPERTIES OF BORATED CERAMICRETE

Density of the fabricated borated Ceramicrete was measured by the fluid-displacement method. Typical values for the density ranged from 3 to 3.2 g/cm^3 . Open porosity was <0.2 vol.%. It is clear that the matrix properties are significantly improved with fly ash additions. A scanning electronphotomicrograph of borated Ceramicrete is shown in Fig. 1; it shows that the matrix is very dense. In addition, second-phase additions, which are well bonded to the Ceramicrete, are also present.

The compressive strength of the samples was evaluated by testing cylindrically shaped borated Ceramicrete samples on an Instron Universal tester. The strength of such ceramics increases from the original ≈ 4000 psi in the phosphate matrix to 7500-9000 psi for borated Ceramicrete. For comparison, the compressive strength of commercial concretes for nuclear shielding applications is 4000 psi. Thus, borated Ceramicrete exhibits significantly enhanced mechanical properties and is expected to be extremely durable.

Preliminary measurements of thermal conductivity have been conducted on borated Ceramicrete. The thermal diffusivity was measured on thin sections (3 mm) of the samples. These experiments were conducted at Virginia Polytechnic Institute with a laser flash technique. Thermal diffusivity values were $0.0054 \text{ cm}^2/\text{s}$. These values compare quite well with those of concrete, which are reported as $0.006 \text{ cm}^2/\text{s}$. This result is expected because both borated Ceramicrete and cement concrete are inorganic materials with a somewhat similar bonding structure. At the present time, we are working on several additions to borated Ceramicrete to further enhance its thermal diffusivity and conductivity values.

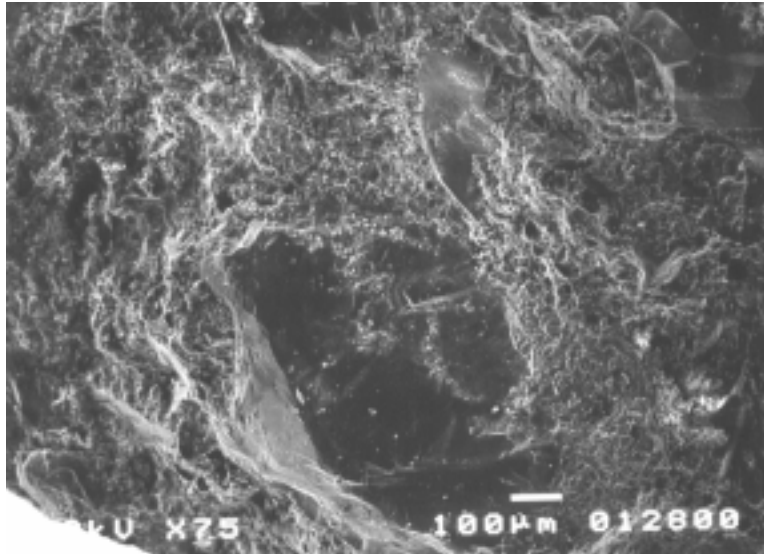


Figure 1. SEM Photomicrograph of Borated Ceramicrete.

NEUTRON SHIELDING

Borated Ceramicrete specimens were exposed to neutrons at the ANL-E Intense Pulsed Neutron Source. A 2.54-cm-thick sample was placed in the neutron beam. The incident (I_0) and transmitted (I) intensities of the neutrons were measured. The ratio of the transmitted and incident intensities was 0.000396. These values were put into the equation below to determine the linear absorption coefficient (μ) for borated Ceramicrete:

$$I/I_0 = \exp(-\mu t). \quad (2)$$

In the equation, t is the thickness of the sample. From the above equation, the linear absorption coefficient of borated Ceramicrete was determined to be 3.08 cm^{-1} . Typically, the linear absorption coefficients of cement grout formulations is 0.06 cm^{-1} [4]. This implies that a 2.24-cm-thick piece of borated Ceramicrete can provide the same shielding characteristics as a 109-cm-thick piece of cement concrete. In other words, the performance of borated Ceramicrete is almost 50 times better than that of ordinary cement concrete. Experiments are currently planned and are being conducted to establish the photon-shielding properties of borated Ceramicrete.

SUMMARY

Preliminary results obtained from borated Ceramicrete material that was developed for nuclear shielding applications, are presented. Borated Ceramicrete is a dense, hard, high-strength material that can be cast in-place under ambient conditions. The material has excellent neutron-shielding capability. Ongoing work includes further development to enhance photon-shielding properties and conduct long-term durability evaluations.

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REFERENCES

1. A. D. Wilson and J. W. Nicholson, *Acid-Base Cements*, Cambridge Univ. Press (1993).
2. D. Singh, A. S. Wagh, J. Cunnane, and J. L. Mayberry, *J. Environ. Sci. Health A32*(2) (1997) 527-541.
3. A. W. Taylor, A. W. Frazier, and E. L. Gurney, "Solubility Products of Magnesium Ammonium and Magnesium Potassium Phosphates," *Trans. Faraday Soc.*, Vol. 59 (1963) 1580-89.
4. W. H. Steigelmann, "Radioisotope Shielding Design Manual," USAEC Report NYO-10721, July (1963).