

1-1-2003

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Recommended Citation

Y. Ye et al., "Influence of Nanocrystalline Grain Size on the Breakdown Strength of Ceramic Dielectrics," *Proceedings of the 14th IEEE International Pulsed Power Conference, 2003*, Institute of Electrical and Electronics Engineers (IEEE), Jan 2003.

The definitive version is available at <http://dx.doi.org/10.1109/PPC.2003.1277809>

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INFLUENCE OF NANOCRYSTALLINE GRAIN SIZE ON THE BREAKDOWN STRENGTH OF CERAMIC DIELECTRICS*§

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Abstract

In an effort to develop transmission lines with higher energy storage capabilities for compact pulsed power applications, the University of Missouri-Rolla (UMR) and the University of New Mexico (UNM) have undertaken a collaborative approach to developing and studying ceramic dielectrics. At UMR, the electrical breakdown strength (BDS) of TiO₂-based materials is investigated for high energy density applications. The results of research to-date show that dense titania ceramics with nanocrystalline grain size (~200 nm) exhibit significantly higher BDS as compared to ceramics made using coarse grain materials. Processing-microstructure-property relationships in TiO₂ systems are found to play a role with respect to increasing the BDS. At UNM, a pulsed power system is being assembled to perform BDS studies of the ceramic materials produced at UMR. Electromagnetic simulations in support of this work will also be presented. The long-term aim of this research is to enable the fabrication of large sizes of high energy density ceramics for use in pulsed power systems.

I. INTRODUCTION

High Power Microwave (HPM) systems require compact, portable pulsed power in order to operate on mobile platforms [1]. To-date the emphasis in HPM research has been on sources, with modest attention being paid to the pulsed power. Capacitive storage-based pulsed power systems are typically used as HPM source drivers. The requirements on such drivers include maintaining constant impedance (typically of the order of 10-100 Ω) at 0.1-1.0 MV voltages for time scales on the order of 100's ns. Capacitive energy storage-based systems are well suited to meet these requirements.

Figure 1 presents a block diagram of an HPM system. The second block in Fig. 1 represents the temporal compression of the voltage pulse. In order to achieve this and properly match to the electron gun load (third block), a pulse forming line is required. One commonly used line is the so-called Blumlein transmission line.

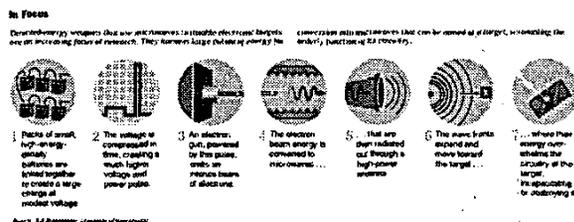


Figure 1. Block diagram of capacitive energy storage-based high power microwave system.

A Blumlein transmission line (or simply a Blumlein) consists of two or more coupled transmission lines (Fig. 2). Rapidly shorting switches cause voltage reversal in half of the lines for a time given by the double transit of an electromagnetic pulse. The resulting characteristic feature of a Blumlein is that the output pulse is roughly equal to the charge voltage of the line. A characteristic of transmission lines is that the voltage pulse length at the load depends on the length of the line. Traditional materials used in such transmission lines include primarily plastic dielectrics, whose dielectric constants range from about 2 – 10. If one could use a material whose dielectric constant were significantly larger, then the length of the transmission line could be significantly reduced for a given voltage pulse length. The challenge lies in finding such a material that also has a high BDS.

* Research at the University of New Mexico supported by an AFOSR/DoD MURI Grant on compact, portable pulsed power.

§ Research at the University of Missouri supported in part by the Center of Dielectric Studies of NSF-I/UCRC program.

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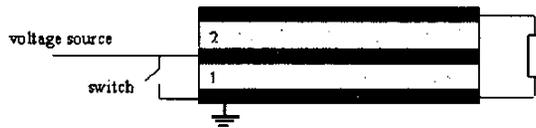


Figure 2. A planar Blumlein transmission line. The full charge voltage appears at the load (indicated on the right).

High dielectric constant ceramics are being developed for a wide variety of applications in electrical engineering, including as a dielectric for high energy density capacitors. Although the dielectric constant of electronic ceramics typically range from a low value of 2.2 for pure SiO_2 up to 30,000 for relaxor ferroelectrics, the parameters of interest to the Blumlein pulse-shaping driver application include high dielectric constant ($\epsilon_r > 100$) and high breakdown strength (>400 kV/cm). The energy density γ (in J/m^3) of a ceramic under the influence of an applied electric field is given by

$$\gamma = \epsilon_0 \epsilon_r E^2 / 2 \quad (1)$$

where ϵ_0 is the permittivity of free space, ϵ_r is the dielectric constant of the ceramic, and E is the applied electric field strength. Typical candidate materials that are being studied in this regard are TiO_2 and BaTiO_3 [2].

II. NANOSTRUCTURED TiO_2 AND DC BREAKDOWN STUDIES

As can be seen from Eq. (1), a high permittivity and a high BDS of dielectric materials result in increased stored energy density (γ). TiO_2 was selected for study as the candidate material for its high BDS (~ 350 kV/cm) and relatively high ϵ_r (~ 110) (compare with $\epsilon_r = 81$ for water). Since the goal of our work is to develop a dielectric with as high an energy storage density as possible, this study is focused on increasing the BDS of the candidate material.

BDS is affected by density, grain size, and defect chemistry of the dielectrics. Single crystal TiO_2 exhibits higher BDS over polycrystalline samples with high grain boundary mobility and residual porosity. Nano-sized materials exhibit a high grain boundary area-to-volume ratio and lower concentration of impurities within the grain boundaries. The samples in this work were prepared by cold isostatic pressing of powders and sintering at various temperatures.

In previous work [3], it was shown that dense nanocrystalline TiO_2 structures (~ 200 nm) exhibited significantly higher BDS than coarse-grained TiO_2 structures (~ 10 μm). This paper concentrates on the results of the nanocrystalline samples compared to the coarse-grained samples in a test configuration designed to measure the intrinsic BDS of the material.

A. Electrical Breakdown Test Setup

At UMR, a dimpled electrode design (Fig. 3) was used in a DC BDS measurement in order to achieve the required high electric field strength. The dimpled sample thickness was ~ 2 mm and the radius of the dimple was ~ 5.5 mm. An optimized process was conducted for dimpled TiO_2 pellets with uniform microstructure. The electrical properties were investigated for both dense nano- (~ 200 nm) and coarse-grained (~ 10 μm) samples. The study confirmed that nanostructured TiO_2 samples exhibited significantly higher BDS compared with coarse-grained structures.

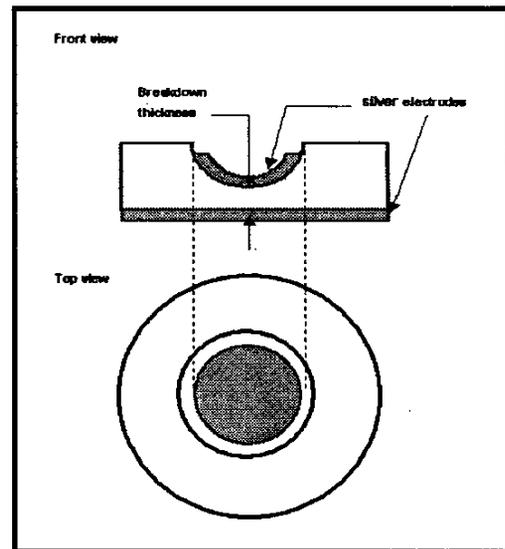


Figure 3. Dimpled electrode design used at UMR.

B. DC Breakdown Results

The BDS properties of nanostructured TiO_2 and coarse-grained TiO_2 were investigated for DC breakdown using the dimpled configuration. The results were higher values as compared with those measured in a set-up using planar electrodes [3]. As can be seen from Fig. 4, the nanostructured TiO_2 shows a higher BDS (1096 kV/cm) compared with the coarse-grained TiO_2 (550 kV/cm). A constant failure rate is assumed in this analysis. For this case, a straight line that crosses zero would be expected for this plot intersecting at the mean value for the BDS (σ_{01} - coarse grained TiO_2 and σ_{02} - nanocrystalline TiO_2). In addition, the effect of sample thickness was studied to see if this had any effect on the measured data not conforming better to the Weibull distribution. Figure 5 shows the results of the thickness study. Due to the strong dependence on thickness, the intrinsic BDS value for nanostructured TiO_2 may be even higher than the characteristic value seen in the previous figure.

III. PULSED BREAKDOWN STUDIES

A. Development of Pulsed Testing Capability

One of the important uses of this high energy density dielectric material will be in pulsed power systems. To

this end, we have started a project to measure the BDS of these same types of materials under pulsed conditions. The design of this test fixture is based upon the Sandia National Laboratories (SNL) work performed a few years ago on ceramic material breakdown [4]. The samples will be placed between electrodes in a de-ionized water container that is then placed in an oil bath for high voltage operation.

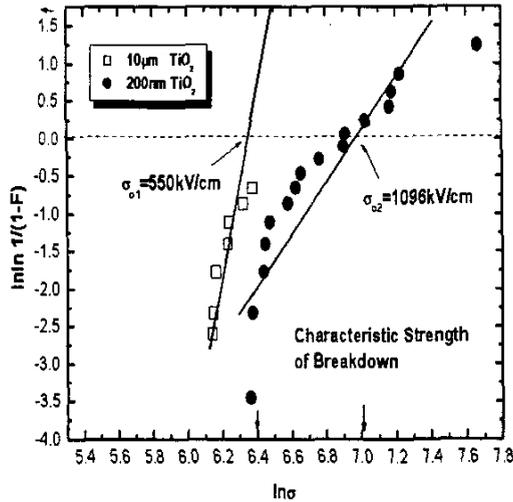


Figure 4. Weibull distribution of breakdown strength (σ) for nanocrystalline- and coarse-grained TiO_2 .

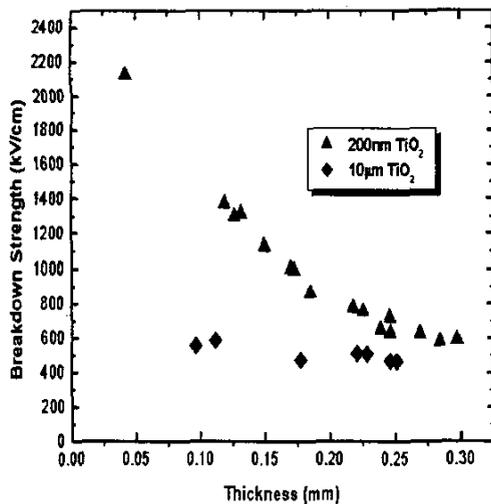


Figure 5. BDS as a function of dielectric thickness for nanocrystalline- and coarse-grained TiO_2 .

The high voltage will be supplied by a 3-element system consisting of a pulse modulator box, an 8-kV transformer and a 100 kV transformer. The features of each of these components are listed below:

- a. Pulse modulator box:
 - 110 VAC input
 - 0 - 600 V pulse output
 - One shot trigger
 - IGBT solid state design.
- b. 8 kV pulse transformer:
 - 0 - 600 V pulse input on primaries (5 turns – 2 in parallel)
 - 0 - 4 kV pulse output on secondaries (80 turns total).
- c. Stangenese pulse transformer (24:1 ratio):
 - 0 - 4 kV pulse input on the primary
 - 0 - 100 kV pulse output on the secondary.

The properties of the resulting waveform is as follows:

- Fast rise time (~ 100 ns)
- Pulse width set at $1.5 \mu\text{s}$ (but will be adjustable in the future)
- 1 – 10 Hz repetition rate
- Maximum voltage of 100 kV.

Figure 6 presents initial data indicating the output of the 8 kV transformer driven by an IGBT-switched pulse modulator. The rise time is 180ns and the peak voltage is 4 kV.

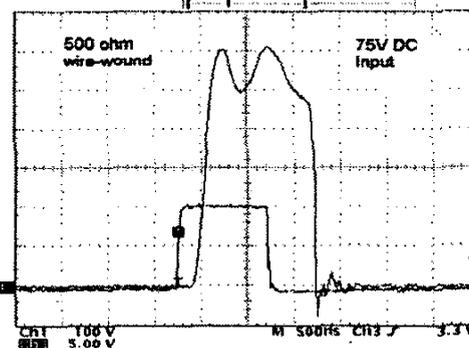


Figure 6. Test output of 8 kV transformer driven by IGBT pulse modulator.

The status of this aspect of the research is that the high voltage transformer is presently being installed in the system (Fig. 7) and testing of the output stage of the modulator will commence shortly.

B. Electromagnetic Simulation Studies

In Fig. 8, we present the results of an HFSS™ (Agilent/Ansoft) calculation to explore the electric field distribution between the center and outer conductors of a Blumlein configuration. In this configuration a voltage waveform is launched between the top and center conductors. A portion of the waveform is transmitted and

propagates between the center conductor and the bottom conductor. A portion of the waveform reflects back upstream in the top section. Finally, after a double transit time, the full line charge voltage appears at the load (shown to be at the end of the line where the coordinate axes are defined in the sketch). The importance of such calculations is to ultimately include the effects of resistivity grading at the outer ends of the line (where the dielectric and liquid interact) and to guide the design of "tailored" dielectrics. Our ceramics effort, described here, is one part of an overall program to find the best materials and suitable geometries that leads to compact and portable pulsed power. (Further information on electromagnetic modeling and recent results using the Microwave Studio™ suite of codes can be found in a companion paper presented at this conference [5].)

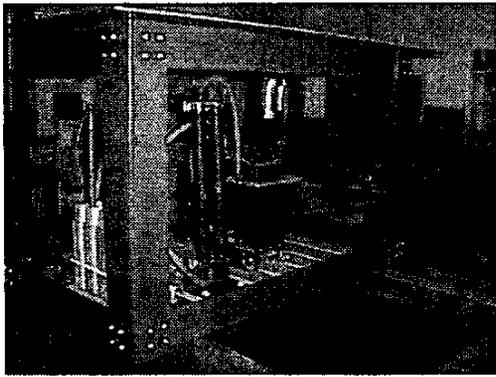


Figure 7. Photograph of the high voltage transformer (copper colored) being installed in the modulator.

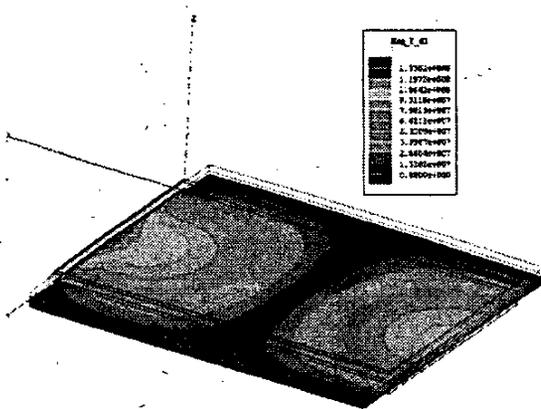


Figure 8. Results of an HFSS™ (Agilent/Ansoft) calculation of a Blumlein at UNM.

IV. CONCLUSIONS

Material studies to-date have suggested that nanocrystalline TiO₂ has higher BDS properties compared with structures made from coarser-grained TiO₂. However, this hypothesis has yet to be validated for pulsed power applications. It is the goal of this program to determine whether ceramics manufactured in this fashion will be a practical material for use in compact pulsed power systems. We hope to be able to report soon on the ability for this material to withstand microsecond-long pulses with fast rise times (~100 ns). Long-term challenges include the ability to manufacture nanocrystalline-grained ceramics in large panel geometries.

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