

EFFECTS OF ALLOYS AND SURFACE TREATMENTS ON ELECTRICAL BREAKDOWN STRENGTH OF WATER AND WATER MIXTURES

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Abstract

Many pulse power machines use purified water as the high voltage dielectric in intermediate storage capacitors. Another candidate dielectric liquid for future pulse power devices is a mixture of ethylene glycol and water, because this mixture can extend the intrinsic time constant from hundreds of microseconds to tens of milliseconds. Longer intrinsic time constant eases the requirements for the charging portion of the pulse power train.

An important characteristic of the liquid dielectric is the energy density. Since energy density varies as the square of the electric field, electrical breakdown is an important physical phenomenon for dielectric mixtures. Our paper examines the effect on breakdown of several alloys and surface treatments for stainless steel and aluminum electrodes in subzero temperature mixtures of ethylene glycol and water and in near zero temperature purified water.

Dielectric Liquids

The use of water as an intermediate energy store in pulsed power devices stems from its favorable physical and electrical properties: high dielectric constant, self-repairability, low cost, ease of purification, handling and lack of health or safety problems. Previous work^{1,2} at NSWC established the need for proper conditioning of the water in order to make valid analyses of electrical breakdown phenomena and to evaluate the efficacy of a capacitor system as an energy store. Deaeration, deionization and the use of nonmetallic, inert materials wherever possible were demonstrated to be necessary. To obtain longer intrinsic time constants, our research demonstrated that cooling increased the useful stress time of water from on the order of microseconds to hundreds of microseconds.

Further research³ at NSWC then showed that mixtures of ethylene glycol and water yielded stress times of tens of milliseconds and were practical high voltage dielectrics. All the techniques used to condition water were found to be useable for the mixtures and were necessary for meaningful analysis.

In both liquids the breakdown strength observed is far less than that predicted from simple theories. Also, it is generally observed that breakdown initiates at the liquid - electrode interface and not in the bulk of the liquid. The role of the interface in electrical breakdown is not well understood.

Electrode Surfaces

The other half of the electrified interface is the surface of the electrode, which is almost always different in material composition than the bulk electrode material. The surface of 304 stainless steel electrodes, which previous investigation⁴

has demonstrated possess superior electrical properties when compared to other common electrode materials, is a combination of various oxides and by-products of the bulk metal. The relatively high chromium content allows the formation of a protective, passive film.⁵ This film can be increased by various passivating techniques and has been found to inhibit charge injection by the surface. Alloys with even higher chromium contents, such as 310, naturally form more protective films. The effect on electrical breakdown and energy storage properties of the passive film had not been previously investigated, to our knowledge.

Aluminum is often the material of choice for intermediate energy stores because it is relatively cheap, light, and easily machined. Previous investigations⁴ had determined the electrical properties of 7075 aluminum alloy - one of the highest strength alloys readily available. The major alloying element in the 7000 series is zinc, which when coupled with a small amount of magnesium results in a high strength alloy, but has moderate corrosion resistance. Anodization is the process whereby aluminum alloy can be given a more passive film. Previous investigation showed that it increased the breakdown strength for this alloy.

Aluminum in all alloys will oxidize in air to form a more or less protective film. The 5000 series exhibits very good corrosion resistance and moderate strength.⁶ The influence of different aluminum alloy surfaces with varying corrosion resistance, and presumably varying protective films, on electrical breakdown and energy storage properties had also not been systematically studied, to our knowledge.

High Voltage Testing

An electrical block diagram for the testing apparatus is shown in Figure 1. The main components are a Marx generator that charges the test cell via a copper sulfate resistor and a high voltage diode. A fast copper sulfate, dividing resistor probe and a capacitance probe transfer voltage signals to an oscilloscope. Figure 2 depicts the dielectric liquid flow. Two separate systems are maintained - one for pure water and one for ethylene glycol - water mixtures. The experimental apparatus has been thoroughly explained in previous papers.^{1,3}

Analysis

The 10% threshold breakdown voltage is defined in the following manner: voltage application for a set of electrodes is started at fields well below breakdown and a set of 10 shots is made at the initial field, then after increasing the applied voltage approximately 10 kV another set of 10 shots is made, this pattern continuing until breakdown; whereupon, the highest field applied to the electrodes that had been successfully held-off for all 10 shots is defined as the 10% threshold breakdown field. This pattern of testing is considered to yield statistically meaningful and repeatable measurements

Report Documentation Page

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1. REPORT DATE JUN 1985	2. REPORT TYPE N/A	3. DATES COVERED -	
4. TITLE AND SUBTITLE Effects Of Alloys And Surface Treatments On Electrical Breakdown Strength Of Water And Water Mixtures		5a. CONTRACT NUMBER	
		5b. GRANT NUMBER	
		5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S)		5d. PROJECT NUMBER	
		5e. TASK NUMBER	
		5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Naval Surface Weapons Center Code: FI2 Dahlgren, Virginia 22448		8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)		10. SPONSOR/MONITOR'S ACRONYM(S)	
		11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release, distribution unlimited			
13. SUPPLEMENTARY NOTES See also ADM002371. 2013 IEEE Pulsed Power Conference, Digest of Technical Papers 1976-2013, and Abstracts of the 2013 IEEE International Conference on Plasma Science. Held in San Francisco, CA on 16-21 June 2013. U.S. Government or Federal Purpose Rights License			
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15. SUBJECT TERMS			
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT SAR
a REPORT unclassified	b ABSTRACT unclassified	c THIS PAGE unclassified	
			18. NUMBER OF PAGES 4
			19a. NAME OF RESPONSIBLE PERSON

in light of the previously reported phenomenon of a "hysteresis-like"⁷ effect when plotting breakdown probability versus applied field. Briefly, this effect implies that a set of electrodes that has been conditioned at lower voltages without breakdown has a lower probability of breakdown than electrodes that have not been conditioned in this manner.

Energy density is a figure-of-merit that can be used to measure capability for energy storage. Energy density, W , is defined as:

$$W = \frac{1}{2} \epsilon \epsilon_0 E^2 \quad (1)$$

where ϵ is the relative dielectric constant, ϵ_0 is the permittivity of free space (8.85×10^{-12} farads/meter), and E is the maximum, average electric field.

However, a better choice for a figure-of-merit is action density. The choice of action density as a figure-of-merit emphasizes the need for adequate hold-off times as well as energy density for future pulse forming lines. Action density also depends on the normalized, effective time constant, $\tilde{\tau}_{eff}$. The normalized, effective time constant is the same as defined by Martin, namely the time the voltage remained above 0.63 of the maximum voltage, except that it is normalized to uniform electrode spacings, peak fields and low voltage intrinsic time constants. This is done to allow fair comparisons between different electrode materials that were tested under slightly different conditions. Details can be found in previous works^{4,8} dealing with charge injection. The reference electrode spacing is 1 cm, the reference peak field is 100 kV/cm, and the reference time constant is 20 milliseconds. Action density, AD, is:

$$AD = \frac{1}{2} \epsilon \epsilon_0 E^2 \tilde{\tau}_{eff} \quad (2)$$

Results

Figure 3 shows the variation in 10% threshold breakdown strength for different electrode materials tested under similar conditions. The nominal test parameters were electrode spacings of 1 cm, 60% ethylene glycol and 40% water (by weight) cooled to -20°C to -25°C , and similar conditioning patterns. The breakdown strength of 310 stainless steel is approximately 14% higher than 304 stainless steel. Recall that 310 had the higher chromium content and the more passive film; it seems that in this case higher breakdown strength accompanied reduced charge injection from the surface. Further theoretical and experimental work is necessary before any cause and effect relationship might be established between these two facts.

For aluminum alloys, anodization has increased the hold-off strength of 7075 by 20%. Highly unusual results were obtained for the bead-blast 5083 alloy. Initial hold-off strength was measured at 192 kV/cm but could not be repeated. Subsequent measurements yielded hold-off values near 150 kV/cm, but finally hold-off fell to a little over 100 kV/cm. The electrodes were bead-blasted once again and hold-off increased to 131 but subsequently fell to the 100 kV/cm level. This is the first case we have noted of such unstable breakdown behavior. Only one measurement was made of

anodized 5083 aluminum and it yielded a hold-off strength of less than 80 kV/cm. Investigations of this alloy will continue.

It should be noted that the breakdown strength of 304 stainless steel, 7075 bead-blasted aluminum and electrolytic tough-pitch copper were approximately the same in ethylene/glycol and water as in pure water.

Figure 4 depicts the action density versus the applied field for the two stainless steel alloys. With the surface bead-blasted, the alloys have only their native passive films and the superiority of the higher chromium content 310 alloy is significant. The action density is higher for 310 than 304 because we observe that the more protective surface has a longer effective time constant - probably because it has less charge injection. However, when the surfaces are passivated in a chromium rich industrial solution, they exhibit similar effective time constants. 310 stainless steel ultimately has the larger action density also because breakdown strength is higher than similarly treated 304.

Figure 5 gives the action density measurements for the two aluminum alloys. Surprisingly, anodization does not yield any benefit in action density at the lower field values indicating that the effective times and probably the charge injection are not affected by the process. At high field values, the increase in breakdown strength for 7075 from anodization yields higher action density. The action density for 5083 aluminum was from the first (and best) measurement. This level of performance has not been duplicated, as the breakdown strength is lower in subsequent measurements.

Conclusions

- (1) Alloying and surface treatment can affect the electrical breakdown strength.
- (2) 310 stainless steel is superior to 304 stainless steel.
- (3) 5083 aluminum exhibited very high hold-off strength but is erratic.

Acknowledgements

This work is supported by the NSWIC Independent Research Program. The authors would also like to express their appreciation to Mr. K. Chilton and Mr. L. W. Hardesty for their able and arduous assistance in making the experimental measurements.

References

1. D. B. Fenneman and R. J. Gripshover, "Experiments on Electrical Breakdown in Water in the Microsecond Regime," *IEEE Trans. Plasma Science*, PS-8, September 1980, p.209.
2. D. B. Fenneman, "Water Capacitors for Pulse Power Applications," Proceedings of Symposium on High Energy Density Capacitors and Dielectric Materials, ed. C. W. Reed, Washington, D.C., National Academy Press, 1981.
3. D. B. Fenneman, "Pulsed High-Voltage Dielectric Properties of Ethylene Glycol/Water Mixtures," *J. Appl. Phys.*, 53(12), December 1982, p.8961.

4. V. H. Gehman, Jr., D. B. Fenneman and R. J. Griphover, "Electrode Surface Effects on Unipolar Charge Injection in Cooled Liquid Dielectric Mixtures," *Digest of Technical Papers, Fourth IEEE Pulsed Power Conference, 1983*, p.316.

5. J. D. Redmond and K. H. Miska, "The Basics of Stainless Steels," *Chemical Engineering*, October 18, 1982, p.79.

6. American Society for Metals, *Source Book on Selection and Fabrication of Aluminum Alloys*, 1978, p.1-20.

7. F. J. Zutavern, M. T. Buttram and M. W. O'Malley, "Dielectric Breakdown Distributions of Large Dielectric Constant Liquids," *IEEE Conference Record of 1984 Sixteenth Power Modulator Symposium*, p.273.

8. M. Zahn, S. Voldman, T. Takada and D. B. Fenneman, "Charge Injection and Transport in High Voltage Water/Glycol Capacitors," *J. Appl. Phys.*, 54(1), January 1983, p.315.

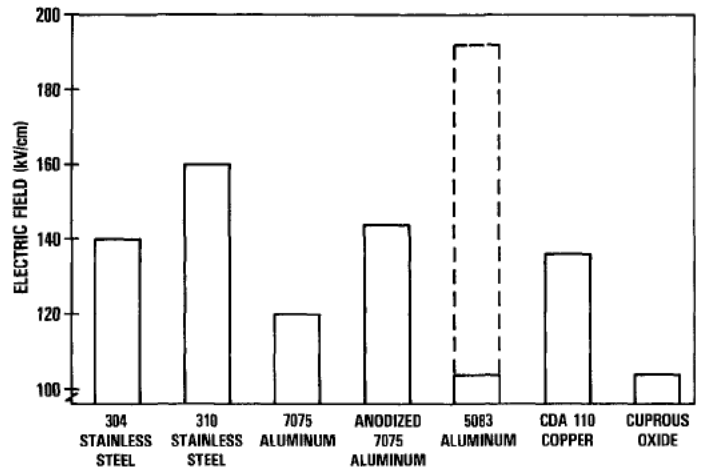


Fig. 3. The 10% probability of breakdown threshold for various electrode materials in a cooled 60% ethylene glycol/40% water dielectric mixture.

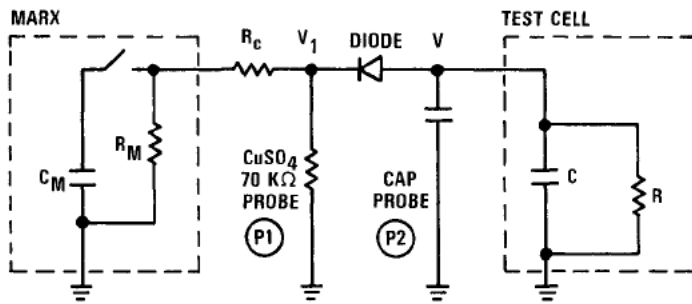


Fig. 1. Electrical diagram of the liquid dielectric experimental apparatus

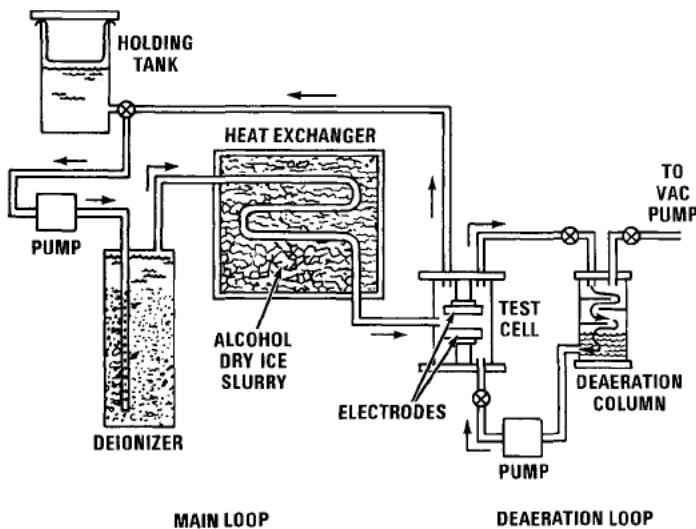


Fig. 2. Liquid flow and conditioning diagram for the experimental apparatus

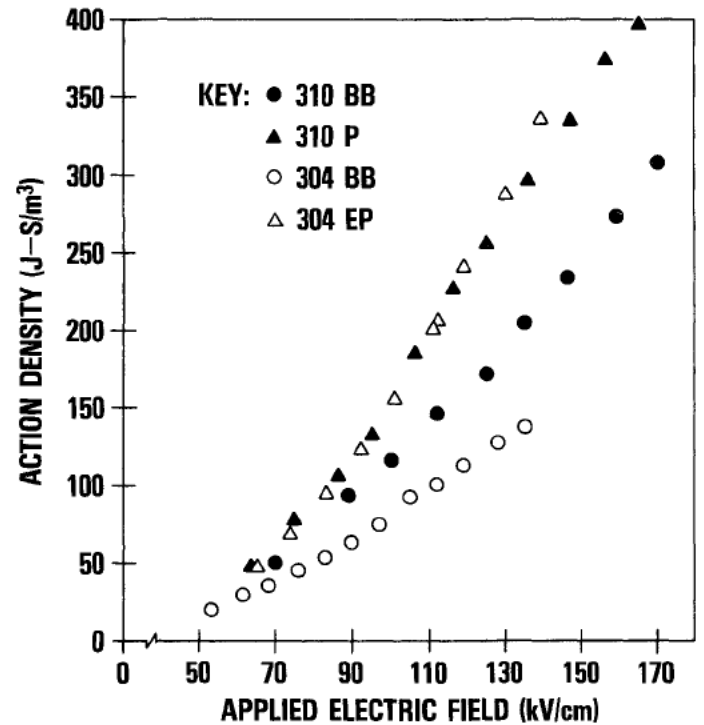


Fig. 4. Action density vs. applied field for stainless steel alloys.
 BB = Bead blasted,
 EE = Electro-polished, passivated,
 P = Passivated

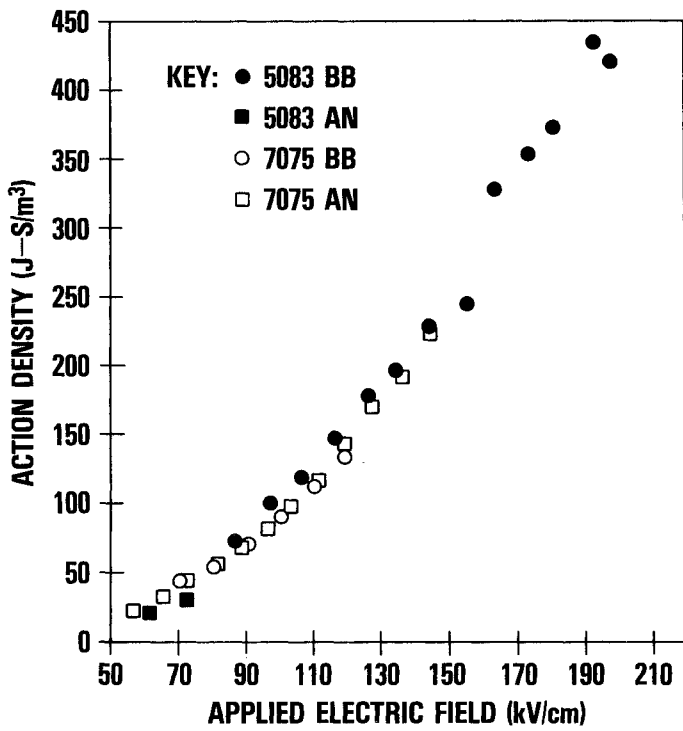


Fig. 5. Action density vs. applied field for aluminum alloys.
 BB = Bead blasted,
 AN = Anodized