Pore-Free Alumina Used in Capacitors Decades Before Lamp Envelope Application

To The Editor:

I enjoyed reading the June 1996 issue of *MRS Bulletin*, which focused on the topic of "Electroceramic Thin Films." The paper on transparent aluminum oxide lamp envelope development (i.e., "Lucalux Alumina: The Ceramic That Revolutionized Outdoor Lighting") by J.E. Burke is particularly well-written and informative.

I would like to point out, however, that, while lamp envelopes were the first mass-produced, *free-standing* objects made from thin, transparent, and pore-free alumina, much thinner films of this material were in use decades earlier as the dielectric material in aluminum electrolytic capacitors.

The first description of transparent, pore-free, dielectric quality alumina films produced by anodizing aluminum substrates (generally in the form of a thin foil) appears in U.S. Patent #2,062,464, which was issued to Dr. Julius Edgar Lilienfeld on December 1, 1936. Lilienfeld employed essentially saturated solutions of boric acid at temperatures between 90 and 100°C.

A much less expensive method of obtaining high-quality crystalline alumina films from dilute citric acid solutions maintained at approximately 90°C was developed in the 1970s. This method and the greatly superior dielectric properties of crystalline alumina films generally were described by C. Crevecoeur and H.J. de Wit in a paper, entitled, "The Influence of Crystalline Alumina on the Anodization of Aluminum," which was given at the 1978 Spring meeting of The Electrochemical Society [May 21–26, Seattle, Washington, Abstract No. 174].

Both of the above anodizing methods rely on aqueous anodizing solutions and produce large amounts of hydrated oxide (in addition to the hydrated oxide already present on the aluminum surface) which tends to dissolve in the electrolyte. Up to 1/3 of the aluminum consumed in the production of anodic alumina dielectric films is ultimately lost through dissolution in aqueous solutions.

Additionally, Crevecoeur and de Wit describe a unique loss of dielectric strength (or "depolarization") which occurs with aluminum-oxide films during anodizing due to increasing oxygen pressure *within* the films (from electrolytic decomposition of incorporated hydrated alumina) causing film cracking. The incorporation of hydrated oxide must be carefully avoided for the production of stable capacitor dielectric films.

During the mid 1980s, I discovered that transparent, pore-free dielectric films could be produced on aluminum by anodization at room temperature in polar, aprotic solvent solutions of phosphoric acid, as is described in my British Patent, GB 2 168 383A.

During the late 1980s, James Clouse and I extended this work with the finding that transparent, pore-free, nonhydrated dielectric anodic aluminum-oxide films containing very little non-alumina material (as indicated by FTIR [Fourier transform infrared] analysis) can be produced in polar, aprotic solvent solutions of the amine salts of phosphoric or boric acid in combination with one or more carboxylic acids (those forming insoluble aluminum salts). These solutions were found to produce the desired films over a very wide temperature range, from approximately -65°C to over 125°C, with appropriate application of current and were patented as electrolytic capacitor electrolytes (U.S. Patents, #4,812,951 and #5,160,653).

It is interesting to note that the first application of high quality alumina films is still the largest, with well over 10 billion aluminum electrolytic capacitors sold worldwide in 1995.

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1996 MRS Fall Meeting/ICEM-96
December 2–6, 1996
Boston, Massachusetts

PLENARY SESSION

John P. McTague
Vice President of Technical Affairs, Ford Motor Company

"Where in the World is Science and Technology Going?"

Monday, December 2
6:00 p.m., Salon E
Boston Marriott

John P. McTague directs the operations of the Ford Research Laboratory, Environmental and Safety Engineering Staff, and the Technical Strategy Office.

Before joining Ford in 1986, McTague was Acting Science Advisor to President Reagan. He was also appointed to President Bush's Council of Advisors on Science and Technology.

IUMRS INTERNATIONAL FORUM ON MATERIALS RESEARCH AND EDUCATION POLICY

Monday, December 2
1:30 – 5:30 p.m., Cape Cod/Hyannis Room
Boston Marriott

The purpose of the first IUMRS Forum is to discuss policies and plans on the future of research and education that are of mutual interest in the global community. It is hoped that through this forum there will be closer collaboration and cooperation among scientists and governments in developing new materials science and technologies which will benefit humankind. IUMRS will sponsor similar forums in other parts of the world in subsequent years.

Speakers include: (Keynote) **Marye Anne Fox**, *Reshaping Graduate and Undergraduate Education in Materials Science and Engineering;* **Lorenzo Gomez**, *Materials Research in Mexico and Trilateral Collaboration with USA and Canada;* **Ryo Imoto**, *Overview of Research andTechnology for Advanced Materials in Japan;* **Lih J. Chen**, *Materials Research and Education in Taiwan;* **Jean Pierre Massue**,*TBA; Hengdi Li,Materials Research Programs in China;* **Rustum Roy**,*Proposed Innovations on Materials Policies;* and **Chong-Oh Kim**, *New Materials Research and Education in Korea.*

PUBLIC AFFAIRS FORUM

Tuesday, December 3
8:00 – 9:00 a.m.
Boston Marriott

"The Future of Condensed Matter and Materials Physics"

The Board on Physics and Astronomy is currently undertaking a series of reassessments of all the branches of physics as the foundation for a new physics survey. As part of this project, a Committee on Condensed Matter and Materials Physics has been established under the leadership of Venkatesh Narayanamurti of the University of California at Santa Barbara. The committee has been working since June on a study that will include an illustrative recounting of major recent achievements; identification of new opportunities, needs, and challenges facing the field; and articulation—for leaders in government, industry, and universities, and for the public at large—of the important roles played by the field in modern society. An especially urgent issue to be addressed in the study is how to maintain the intellectual vitality of condensed matter and materials physics, and its contributions to the well-being of the United States, in an era of limited resources.

This interactive forum will feature a panel of materials researchers who are members of the Committee on Condensed Matter and Materials Physics. They will give a brief report on the status of the study and engage in a dialogue with MRS members about the issues facing the materials research community. Community input will be vital to the success of the study. Come make your voice heard!

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