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**CRADA Final Report
for CRADA Number ORNL91-0056**

**MICROWAVE PROCESSING OF
TANTALUM CAPACITORS***

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AVX Tantalum Corporation

*Research sponsored by the U.S. Department of Energy, Office of Energy Research, Laboratory Technology Applications Division, under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp.

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MICROWAVE PROCESSING OF TANTALUM CAPACITORS

CRADA 91-0056

ABSTRACT

A Cooperative Research and Development Agreement (CRADA) between Lockheed Martin Energy Systems, Inc. (LMES) and AVX Tantalum Corporation (AVX) of Biddeford, Maine, was initiated in October 1991. [Lockheed Martin Energy Research Corp. (LMER) has replaced LMES]. The completion date for the Agreement was March 1996. The purpose of this work is to explore the feasibility of an advanced microwave processing concept to develop higher capacitance tantalum anodes. Tantalum capacitors are used where high reliability is needed (e.g., pacemakers, hearing aids, and military devices). Two types of tantalum powder are used: sodium-reduced powder and electron beam-refined powder. Sodium-reduced powder has higher surface area, but lower purity; electron beam-refined powder has higher purity for working voltages, but somewhat lower surface area. The powder is pressed into pellets using traditional methods and then placed in the microwave furnace for processing. It is of interest to determine if variable-frequency microwave sintering can increase quality while decreasing processing time and decreasing or eliminating surface contamination; these issues must be addressed while retaining the maximum surface area of the anode. Meeting each of these needs will result in a higher quality anodic film, which will thereby increase the dielectric strength. Additionally, microwave sintering might enable us to develop a strong sintered anode without excessive grain growth.

The variable-frequency microwave furnace (VFMF), located at the Y-12 Plant, allows us to study the effects of sintering over a wide frequency range. This novel design uses a high-power traveling wave tube (TWT), originally developed for electronic warfare. By using this microwave source, one can not only select individual microwave frequencies for particular experiments, but also achieve uniform power densities over a large area by the superposition of many different frequencies.

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OBJECTIVES

The long-term objective of this CRADA is to develop microwave sintering as a means of improving the quality and reliability of tantalum capacitors. Process variables explored include microwave power, atmosphere, and casketing/insulation techniques. The goal of studying these variables was to determine the effect on microstructure development, surface chemistry, and, ultimately, device reliability. A parallel objective was to gain fundamental insights into details of the microwave sintering process.

ATTAINMENT OF OBJECTIVES

Tantalum anodes have been successfully sintered using Variable-Frequency Microwave (VFM) processing. In order to initiate microwave heating as well as to provide thermal insulation at high temperatures, various casketing arrangements were used in the microwave furnace. Initially, oxide refractories, such as zirconia bubbles or yttria fiber, were used as insulating materials. Sintering was achieved, but the tantalum reacted with the insulating materials. Also, gases released from the oxide refractories degraded the vacuum. Because cleanliness of the anode is crucial to the fabrication of a good capacitor, it is clear that this casketing system was unacceptable.

In order to minimize material interactions and temperature nonuniformities, the oxides were eliminated from the insulating strategy, replaced by tantalum powder. This replacement with tantalum powder has resulted in several surprising advantages: Not only is thermal insulation provided by the powder, but microwave coupling is improved while permitting some microwave power to penetrate the samples. Also, the tantalum powder eliminates surface contamination by acting as a getter for oxygen. We have also improved the vacuum system. Additionally, a higher surface-area powder (26,000 $\mu\text{fd/V/g}$), representing state-of-the-art products, is being used.

SPONSOR BENEFITS

This project offers several benefits to our Department of Energy (DOE) programs. First, it continues the development and understanding of microwave/materials interactions begun under Fusion Energy support. Second, it helps us to develop new approaches for processing refractory metals, which will benefit the DOE Office of Health and Environmental Research, DOE Defense Program, and the U.S. Department of Defense projects. Third, it provides promising avenues for further research into high-performance capacitors and other energy storage technologies.

TECHNICAL DISCUSSION

The insulation configuration employed is such that the anodes are buried in tantalum powder inside a boron nitride crucible. The boron nitride crucible is surrounded by alumina bubbles in an outer boron nitride crucible. Temperature measurements are made with an Accu-Fiber™ probe and an optical pyrometer. A soak temperature of 1500 to 1600°C is typically held for 30 min at a vacuum of approximately 0.133 MPa (10^{-6} Torr). The frequency is 5.5 to 6.5 GHz, sweeping over the 1-GHz range 4000 times per second. The forward power input is approximately 1kW.

Microwave processed samples are sent to AVX for anodization and capacitance testing. Samples processed in the VFMF are strong and have been successfully anodized. Gross contamination has been eliminated, and electrical characteristics are acceptable. However, data suggest that some actual specimen temperatures were slightly higher than indicated by probe; more accurate temperature control has been implemented. Most remarkable are the ripples that appear in the microstructure (Fig. 1). These ripples only appear in microwave processed samples and may act to increase the surface area and may, thereby, increase the capacitance. The origin of this effect is unknown, but it suggests the possible existence of microscopic arcing or plasma formation within the pore spaces. Such phenomena could be helpful by encouraging rapid sintering without coarsening and/or by cleaning away surface impurities.

INVENTIONS

Two invention disclosures arose as a result of this CRADA. The first invention was issued as U.S. Patent No. 5,184,286. The second invention was filed as U.S. Serial No. 08-708424 on September 5, 1996 ("Improved Method of Making Tantalum Capacitors," A. D. McMillan, R. E. Clausen, and W. F. Vierow).

PLANS FOR FUTURE COLLABORATION

We will continue to collaborate on an informal basis and will, of course, continue to work together on the prosecution of the pending patent. AVX will complete an analysis to determine if microwave-processed anodes will be introduced into their line. We will also discuss possible collaborative opportunities in related product niches.

CONCLUSIONS

The CRADA between LMER and AVX has been completed successfully. Tantalum anodes were successfully sintered in a VFMF while maintaining acceptable electrical characteristics. Difficulties with casketing techniques have been overcome by the use of tantalum powder as insulation. The powder not only acts as insulation, but also removes some impurities by acting as an oxygen getter. Ripples on the microstructure of the anodes provide more surface area to the anodes and are unique to microwave processing.

SINTERING TANTALUM CAPACITOR ANODES USING VARIABLE-FREQUENCY MICROWAVES

Problem: Tantalum capacitor anodes can be sintered using microwave heating provided the insulation casket contains lossy materials to initiate heating from ambient temperatures. Previous work relating to U.S. Patent 5,184,286 was plagued by contamination from the insulating materials used. Work is ongoing to eliminate surface impurities and to minimize coarsening.

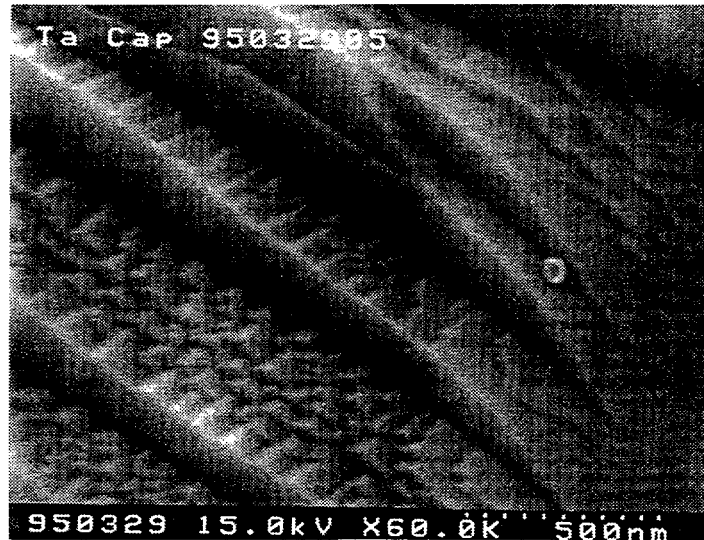


Fig. 1. Ripples in microstructure.

Solution: We have discovered that powdered tantalum can serve as the casketing material, providing thermal insulation during sitnering while acting as a good getter of impurities, particularly oxygen. Anodes sintering using this novel method exhibit unusual ripples on the individual tantalum grains. These ripples increase the surface area of the anode and, therefore, increase the capacitance. This improved casketing technique is the subject of a joint patent disclosure between Oak Ridge National Laboratory and AVX Tantalum Corp.

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