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THERMALLY INDUCED EVOLUTION OF MORPHOLOGY ON CERAMIC SURFACES IN A THERMIONIC CONVERTER

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Abstract

The morphology of alumina and scandia ceramics exposed to controlled vacuum and diffusion modes in a thermionic converter has been studied. Evidence for vaporization at a temperature of 1770 K is manifest in the resulting surface morphologies of both ceramics, consistent with reported sample mass loss. Alumina shows intergranular relief with the formation of terrace - step structure on the grain surfaces. Terrace formation is not directly observed on scandia, however the development of vertical structure and maintenance of voids indicates that vaporization is initiated by structure at the grain edges. Extensive Sc_2O_3 re-deposition occurs on the scandia surface, possibly mediated by the presence of molybdenum and tungsten. Evidence exists for refractory metal secondary phase formation in this deposit in the form of $\text{Sc}_6\text{MO}_{12}$ ($M = \text{W}$ or Mo). Alumina also shows evidence for materials' interactions in the form of tantalum assisted vaporization which significantly alters the terrace structure.

INTRODUCTION

Ceramic materials are often used as mechanical spacers in the interelectrode gap of thermionic conversion devices. Scandia (Sc_2O_3) has been the material of choice for the TOPAZ-II thermionic fuel elements. The spacer's primary function is to maintain a critical gap distance at operational temperatures when emitter distortion occurs. These materials are immersed in a Cs plasma and are subjected to a host of energetic, thermal and mechanical stresses. Degradation of these materials can directly impact both the performance and longevity characteristics of a converter. Variations in the mechanical stability of metal oxide insulators can lead to changes in interelectrode gap distances and changes in converter efficiency. Material loss from the spacer through vaporization reactions (Kozlov et al. 1993) will lead to shifts in the interelectrode material balance and possibly changes in the plasma characteristics. As a result, the chemical and physical roles these material play need to be understood.

In this paper, we report on the morphology of scandia and alumina surfaces subjected to vacuum and diffusion mode exposure. The vacuum and diffusion modes for a converter are defined as operation at the operational emitter temperature with the interelectrode gap evacuated or pressurized with cesium, respectively (Baksht et al. 1978). These two modes are the precursors to the ignited mode where electron flux is allowed from emitter to collector. These two modes represent the first step in understanding what processes are expected to take place on the spacer surfaces. Kozlov et al. (1995) have shown that vaporization of both alumina and scandia occur at 1770 K for these two modes. The gravimetrically determined vaporization rates show a trend of an approximate 10-fold decrease on passing from vacuum to diffusion modes (7×10^{-9} to 8×10^{-10} and 6×10^{-10} to 7×10^{-11} $\text{kg/m}^2\text{sec}$) for both alumina and scandia. Ignited mode operation produces an increase in evaporation rate (5-fold relative to the vacuum mode) as well as large-scale microstructural changes for alumina, while scandia shows no discernible change in vaporization rate or microstructure (Kozlov et al. 1995). We have conducted an analogous set of experiments and report on the resulting microstructural evolution. Vaporization from most oxides results in the dissociative formation of the metal species and substoichiometric oxides (Lamoreaux et al. 1987). These processes represent reactions that can be chemically assisted. Our goal is to use this microstructural information, as well as some surface compositional information, to better understand these vaporization reactions within a converter environment. Attempts to identify stable ceramic materials for this application will require an understanding of how microstructure and composition impact these reactions.

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