IDENTIFICATION OF GAS-DIFFUSION PROCESS IN A THICK AND POROUS CATHODE SUBSTRATE OF SWPC TUBULAR SOFC USING AC IMPEDANCE METHOD

Keqin Huang

Siemens Westinghouse Power Corporation
1310 Beulah Rd., Pittsburgh, PA15235, USA

Concentration polarization, as one type of electrochemical polarizations commonly encountered in SOFCs, results from the concentration gradient of reactive species across the thickness of the electrode. A limited gas-diffusion through an electrode is the root-cause of concentration gradient, therefore concentration polarization. For thin-film SOFCs, the thick and porous electrode-substrates are clearly the major barriers for the diffusion of reactive gases to the electrolyte interfaces. For example, a limited H_2-H_2O diffusion through the porous anode substrate could be a cause of concentration polarization for anode-support planar SOFCs whereas a limited O_2-N_2 diffusion through the porous cathode substrate in SWPC tubular SOFC could be a performance-reducing factor. In this paper, we report a direct experimental observation of gas-diffusion process occurred at a thick and porous cathode substrate of SWPC tubular SOFC. The main technique that was used to identify the gas-diffusion process in this study was the AC impedance spectroscopy, in conjunction with the effect of cathodic DC bias, effective O_2-diffusivity and bulk Po_2.

The results indicated that no pore gas-diffusion process could be found on the impedance spectra at lower temperature range regardless of cathodic DC bias level (down to –200 mV), bulk Po_2 and effective O_2-diffusiivity. Figure 1 showed the effect of the applied cathodic DC bias on the impedance spectra at 800^{o}C in air. The electrode resistance decreased as the DC bias increased, indicating the dominance of activation polarization process. In addition, two semicircles appeared to evolve from one big semicircular arc as the DC bias increased. In literature, these two semicircles were ascribed to two elementary steps associated with the oxygen reduction kinetics, with the high-frequency one related to the O^{2-} transfer and intermediate-frequency one related to the O^- surface diffusion. With increasing temperature to above 900^{o}C, however, the pore gas-diffusion process became visible on the spectra. Figure 2 showed a similar measurement to Figure 1, but taken at 1000^{o}C. The pore gas-diffusion process was shown as a semicircle at the lowest frequency on the impedance spectra.

The results of varying bulk Po_2 and effective O_2-diffusivity further confirmed the above findings, *i. e.*, at lower temperatures the pore gas-diffusion cannot be manifested on the impedance spectra within a fixed frequency domain. With increasing the temperature to above 900^{o}C, the percentage of the gas-diffusion resistance was increased as the bulk Po_2 and effective O_2-diffusivity deceased, as shown in Figure 3.

The above results provide good examples of illustrating the relationship between activation and concentration polarizations as the temperature varies. At lower temperature range, the electrode kinetics was predominated by the activation polarization, which has been indirectly verified by the effect of cathodic DC bias. As temperature increases and the activation process becomes thermally activated, the pore gas-diffusion comes into play and quickly becomes dominated at higher current density, low bulk Po_2 and low effective O_2-diffusivity.

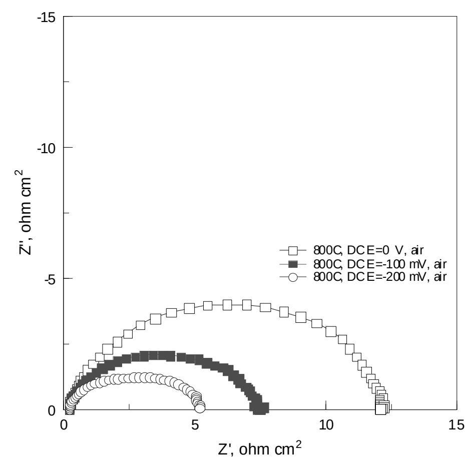

Figure 1 AC impedance spectra taken at 800^{o}C and DC bias in air

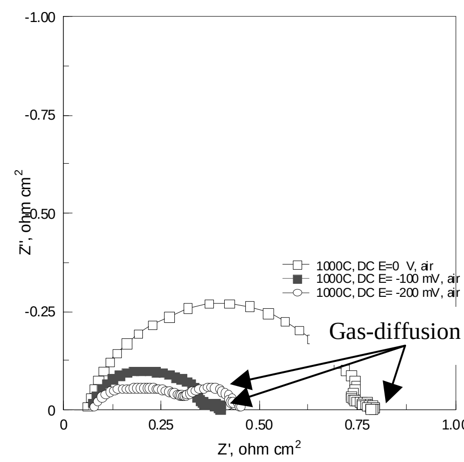

Figure 2 AC impedance spectra taken at 1000^{o}C and DC bias in air

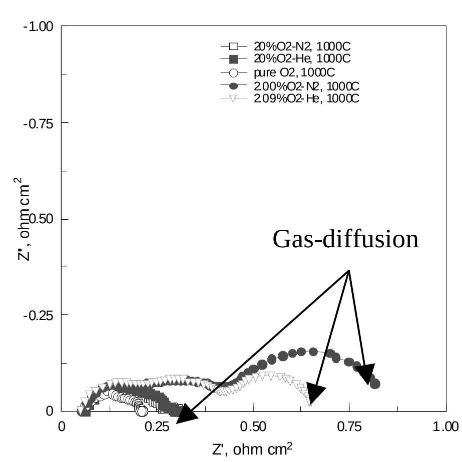

Figure 3 Impedance spectra in He-O_2 and N_2-O_2 mixtures with varied Po_2 at 1000^{o}C