7 - 11 JULY 2024
NANOSMAT Barcelona

15th International Conference on Surfaces, Coatings and Nanostructured Materials
www.nanosmat2024.com

ABSTRACT:

On the Role of Electrochemical Instabilities in Electrolytic Plasma Processing

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Electrolytic plasma technology encompasses a group of high-voltage electrochemical processes featuring spectral light emission at the interface of metallic working electrode with ionically conductive electrolyte. The onset of light emission in these processes is usually associated with two types of behaviour observed in transient polarisation curves of corresponding systems; active-passive transitions that lead to the appearance of diffuse glow and passive-active ones resulting in a filamented type of discharge. Empirically, these two phenomena are treated separately, one associated with separation of the working electrode from electrolyte by a continuous film of vapour-gaseous products of electrolysis and the other with a dielectric breakdown of insulating surface oxide film. However this does not explain the appearance of active-passive transitions and soft sparking phenomena in the systems whose behaviour is dominated by the presence of dielectric oxides grown under pulsed reversed polarisation conditions. In this work, we examine electrolytic plasma processes from the viewpoint of electrochemical instabilities theory which considers the interfacial charge transfer as a competition between a faster process that accelerates the main electrochemical reaction and a slower inhibiting process. Within certain ranges of uncoupled electrolyte resistance, this competition leads to the appearance in the polarisation curve of a region with a negative differential resistance (NDR) which is responsible for the development of the unstable behaviour. We further attempt to identify the nature of instabilities in electrolytic plasma processes using active diagnostic tools recently developed in our group. Our findings indicate that electrolytic plasma processes can be influenced by non-electrochemical phenomena, such as diffusion of intercalated species in the barrier layer, which may be responsible for collective discharge behaviour independent of the direction of electric field. Importantly, control over this collective behaviour can enable uniform treatments of irregular 3D shape surfaces without application of auxiliary electrodes that are not always practical in electrolytic plasma processing.