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### ACCEPTED MANUSCRIPT

# Aryldiazonium Salt Derived Mixed Organic Layers: from Surface Chemistry to Their Applications

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#### Abstract

Formation of multicomponent layers *via* electrografting of aryldiazonium salts has been recognized by materials scientists and electrochemists as a powerful tool due to their high capability for covalent surface modification with multiple functionalities. The past decades have seen significant progress in fabrication and application of surfaces decorated with layers containing more than one component, so called mixed layers. This progress is based on advances in the understanding of aryldiazonium saltbased electrochemistry and their surface chemistry properties. In this critical review, we summarize the different strategies for controlling formation of the mixed layers, especially with respect to surface composition, spatial distribution and surface morphology whereupon the focus shifts to the diverse range of applications in which mixed layers are employed. The limitations of existing strategies and perspective opportunities will also be presented.

Keywords: mixed layers, aryldiazonium salts, electrografting, surface chemistry, sensing applications

#### 1. Introduction

Electrochemical reduction of aryldiazonium salts have attracted considerable attention for surface functionalization with covalent bonding at the substrate–aryl interface since Pinson and co-workers described the reaction mechanism for the modification of carbon electrodes by aryldiazonium salts in 1992 (Scheme. 1) [1, 2]. It has been demonstrated to be an efficient way to introduce many types of functional groups to decorate, not only a variety of carbon surfaces (glassy carbon [3], graphite [4, 5], screen printed carbon electrodes [6], carbon nanotubes [7] and diamond [8]) but also metals [9], silicon [10-12], and indium tin oxide [13, 14] with a reaction time scale of seconds to minutes. A surface can be functionalized with either one type or multiple types of aryldiazonium salts, which are defined as single-component or mixed-layers respectively. The modification and application of single-component surface

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