Electrochemiluminescence confined in nanofluidic electrochemical transducers

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Abstract

In annihilation electrogenerated chemiluminescence (ECL), tris(bispyridine)ruthenium(II) Ru(bpy)32+ can be oxidized and reduced at two electrodes biased at constant high and low overpotentials, respectively. The generated species Ru(bpy)33+ and Ru(bpy)31+ recombine (i.e., annihilate) in solution to form an excited light-emitting state Ru(bpy)32+*. After emission of a photon, Ru(bpy)32+ is regeneratedand can repeat the cycle of annihilation and light emission. This reaction scheme is among the most important in ECL and has been researched, e.g., for optofluidic devices. This annihilation reaction and, thus, the intensity of light emission, are limited by the diffusion time in between the two electrodes. For the first time we employ transparent *nanofluidic* thin-layer cells for an annihilation scheme. Using 10 mM Ru(bpy)32+ in acetonitrile, we generate Ru(bpy)33+ and Ru(bpy)31+ at opposite Pt electrodes positioned at the walls of a nanofluidic channel with a height (=inter-electrode distance) of 100 nm. Here, the diffusion time from the electrode surfaces to the center of the nanochannel is reduced to just microseconds. We demonstrate highly efficient light generation at the nanoscale and image the area of light emission in the nanofluidic transducer. Analysis of the generated electrochemical currents reveals that reactions can be limited by kinetics, and that nanofluidic transducers can be used to determine reaction rates.

Keywords: Electrochemiluminescence, Redox cycling, nanogap transducer, nanofluidics

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