Temporal Behavior of the Individual Soft Microparticles: Understanding the Detection by Particle Impact Electrochemistry

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Abstract: Droplets loaded with ferrocene (Fc) were analyzed by the Particle Impact Electrochemistry (PIE) technique using a Carbon Ultramicroelectrode (UME). The technique successfully characterized the size and polydispersity of the emulsion droplets. A 3D random walk lattice simulation with MATLAB was used to model for the random motion and collision dynamics of the droplets. The simulation indicated the stochastic nature of the droplet motion. The smaller droplets are faster compared to the larger droplets. Unlike nanoparticles, the droplet collision associated with adsorption on the surface of the electrode generating a similar spike-like current response to real-time experiments.

Introduction:
- PIE detects the electrochemical current changes upon collisions of individual micro or nanoparticles at the UME surface.
- This technique is a rapid, low cost, and analyze one analyte at a time.
- Quantitative information on the collision and adsorption dynamics of the soft-microparticles (absence of crystalline structure) will enable the PIE to characterize them.

Hypothesis:
- Emulsion droplets (Soft Microparticles) loaded with redox species will react electrochemically on the surface of the disk UMEs.
- The collisional behavior of the droplets with UMEs can be investigated by random walk simulation.

Methods:
- Preparation of 20 mM Fc-toluene in-water emulsion
- Characterize the emulsion by PIE technique
- Investigate collision dynamics by random walk simulation
- Simulate the current

Preparation and stability of emulsion

Electrode: 3 µm Pt-UME (A) in-house made, (B) 20 µm Au-UME (C) 50 µm Au-UME (D) 100 µm Au-UME

Droplet diffusion over migration

Droplet Adsorbed

Collision frequency (Hz)

Ratio of droplet and electrode diameter

Collision frequency (KHz)

Time (s)

Current (nA)

Figure: Schematic diagram of particle blocking experiment. (B) Simulated mass transfer frequency (red dots) as a function of simulation hemisphere radius and frequency (blue dots) for a droplet collision with 20 seconds simulation runtime of a 1.0 µm (diameter) droplet. (C) Observed mass transfer frequency (blue dots) as a function of droplet diameter of different simulation runtimes.

Characterization of emulsion by PIE

Figure: Schematic diagram of Amperometric i-t curve. (A) 20 mM Fc-toluene droplet. (B) Empty droplet. (C) Oxidation current of 1.0 µm diameter 20 mM Fc electrode dropping on a 6.0 µm (diameter) electrode. (D) Change of concentration resulting of multiple collision of the 1.0 µm (diameter) 20 mM Fc electrode dropping on a 6.0 µm (diameter) electrode. (E) Oxidation currents of 1.5 µm (diameter) 20 mM Fc electrode dropping on a 6.0 µm (diameter) electrode. (F) Comparison of simulated and experimental current of a 1.0 µm (diameter) 20 mM Fc electrode dropping.

Conclusion:
- PIE technique successfully detected the polydispersity 20 mM Fc emulsion droplets.
- The droplets:
  - have stochastic motion in the solution.
  - undergo irreversible adsorption on the surface of electrode.
  - oxidize following the bulk electrolysis model.