558g Kinetics of Deposition of Reactive Chromophores in Layer-by-Layer Films for Second Order Nonlinear Optical Materials

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Organic films of oriented chromophores can be used to fabricate electro-optic modulators for telecommunications applications. Low molecular weight chromophores can be integrated into films by a layer-by-layer deposition process using electrostatic interactions and covalent bonding. The deposition process involves aqueous solutions of ionic chromophores and a polycation at ambient conditions with individual immersion times as low as 2 minutes/monolayer. The chromophore used was Procion Brown (PB) while poly(allylamine hydrochloride) provided both the amine groups for reaction with the triazine ring of PB and for electrostatic interaction with the sulfonic acid groups of PB. This results in non-centrosymmetric films that exhibit significant second harmonic generation (SHG) intensities where the square root of the SHG intensity scaled linearly with bilayer number. Values of the second order nonlinear susceptibility from the SHG data as large as 31x10*f*{9 esu and the electro-optic coefficient r33 = 14 pm/Volt were found. By comparison, films of LiNbO3, an inorganic material used in EO modulators, exhibit values of second order susceptibility of 200x10*f*{9 esu and r33 = 30 pm/Volt. The pH and ionic strength were critical for achieving a high degree of ordering of PB. The effects of solution concentration, ionic strength, and convection on the deposition rates of the PB dye and the polycation were explored using a quartz crystal microbalance to measure the intrinsic reaction rate of the PB dye with the polycation.