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Project title: Sustainable, Low-Cost Route to Cellulose Nanocrystals by Electrochemical Oxidation

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Abstract (approx. 200 words): Cellulose nanocrystals (CNCs) are short, rigid rods of crystalline cellulose derived from plant-based or microbially generated filaments. They have attracted significant research attention as they are sustainable lightweight materials possessing high strength and stiffness as well as biocompatibility. However, current methods for CNC production are costly, labor-intensive, environmentally hazardous, and typically result in low yields, hindering their utilization.

One approach to tackle the issues of yield and sustainability in CNC preparation is to perform the hydrolysis with HCl gas, followed by TEMPO-mediated oxidation. This reduces and simplifies the purification steps for CNCs and introduces carboxylic groups on CNC surfaces to facilitate their dispersion – an approach that has shown improved yields. However, the use of hypochlorite as an oxidant in TEMPO-oxidation is expensive and environmentally problematic.

We outline a method utilizing *electrochemical* TEMPO-oxidation for CNC preparation after pre-hydrolysis with HCl gas. This process occurs entirely under aqueous conditions and only requires the use of HCl (g) and TEMPO catalyst, which have both demonstrated recyclability. Previous electrochemical approaches were only able to obtain cellulose nanofibers, which show inferior mechanical properties to CNCs. By performing our reaction in water without hypochlorite, our approach offers a more sustainable pathway to CNCs with improved yields and simplified purification, paving the way to their wider utilization.

Key words: Cellulose nanocrystals; sustainability; TEMPO-oxidation; electrochemistry;