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Intensification of photooxygenation reactions by combining recyclable photoactive polymer colloids with LED-driven flow chemistry and membrane-based separation

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Abstract

Singlet oxygen is a robust and selective oxidant which is broadly used in synthesis [1], including also at industrial scale (i.e. arteminism [2] or rose oxide [3] syntheses). It is produced by safe photosensitization of oxygen in the visible range at ambient temperature. Soluble Photosensitizer (PS), of organic or inorganic nature, is generally used while the concept of solid-supported PS offers undeniable advantages: their separation from the other products is easier, they can be recycled, it is a strategy for transposing poorly soluble PS into green solvents, and their photostability is enhanced [4]. PS can be covalently bonded to silica, metal or polymer-based materials. However, employing macromolecular engineering to design photoactive colloids offers greater flexibility for tuning size, PS loading and localization while achieving high singlet oxygen production quantum yield [5]. Flow photochemistry has emerged as promising alternatives to batch processing, particularly when combined with LEDs [6, 7].

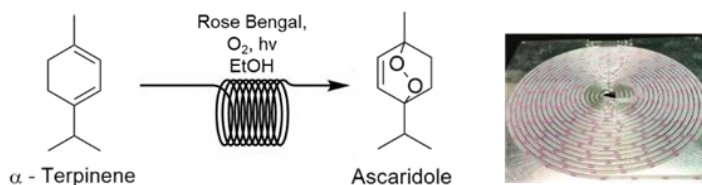


Figure 1. Benchmark photooxygenation and LED-driven spiral-shaped microreactor with “slurry” Taylor flows

In this context, the present work aims at demonstrating how photooxygenation processes could be intensified by combining recyclable photoactive polymer colloids with continuous-flow LED-driven microstructured and membrane-based technologies. For this purpose, the synthesis of ascaridole was considered (Fig. 1). In contrast to prior studies [5, 8], a new generation of polymer colloids was implemented, consisting of core-shell nanoparticles in which Rose Bengal (RB) molecules were covalently bonded into the shell.

The continuous-flow process consisted of a spiral-shaped microreactor irradiated by a custom-built visible LED panel (which photon flux could be tuned), followed by an ultrafiltration membrane fiber to separate the colloids from the reaction medium. Slurry “Taylor” flows were generated, allowing efficient contact between reactive species and colloid transport in the reaction medium. Full conversions with short residence times and high selectivity could be obtained whatever the sets of polymer colloids. Interestingly, the photobleaching levels were very limited when compared to free RB. The membrane-based recovery efficiency was found to be strongly dependent on the latex size, concentration and structure.

At last, the findings open promising perspectives for designing a whole continuous-flow process, including photooxygenation, colloid separation and recycling steps.

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