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Fabrication of topologically anisotropic micro particles and their surface modification with pH responsive polymer brush

Ifra
Indian Institute of Technology Delhi, India

Abstract

The performance of particles is highly influenced by particle size, shape, surface chemistry, elasticity and permeability1. Electrohydrodynamic jetting technique has proven to be a versatile technique to fabricate particles with different shapes and sizes. In this work, we have fabricated topologically anisotropic cup shaped made from polylactide (PLA)/ poly[methylmethacrylate-co-2-(2-bromopropionyloxy) ethyl methacrylate] (75/25) of ~6 µm size using electrojetting technique. Solution and processing parameters were changed to understand the mechanism of cup shape formation and to control particle's shape from cups to discoids. Surface initiated atom transfer radical polymerization (ATRP) of stimuli responsive DMAEMA (2-dimethylamino ethyl methacrylate) was subsequently carried out for 1 h onto the surface of cup shaped particles to observe pH responsiveness of the modified anisotropic particles. An interesting change in the morphology of cup shaped particles was observed which changed to elongated cup and showed significant swelling under acidic pH (swelling ratio:~1.6), also enhanced dye adsorption at specific pH was observed by optical microscope and confocal laser scanning microscope implying that DMAEMA polymerization happened onto the surface of the composite microparticles. The Raman microscopy and FTIR spectra obtained from the particles after polymerization further confirmed the immobilization of pH responsive poly(DMAEMA) brushes onto the cup shaped particles which may potentially function as triggered/targeted drug delivery vehicles. Moreover, the brush modified cup shaped particles were found to be two times more efficient in adsorbing dye compared to disc shaped one indicating a clear advantage of using cup shaped particles over other shapes for immobilizing/adsorbing charged species e.g. sensitive biomolecules.

Responsive polymer brushes are a category of polymer brushes that are capable of conformational and chemical changes in response to external stimuli. They offer unique opportunities for the control of bio−nano interactions due to the precise control of chemical and structural parameters such as the brush thickness, density, chemistry, and architecture. The design of responsive brushes at the surface of nanomaterials for theranostic applications has developed rapidly. These coatings can be generated from a very broad range of nanomaterials, without compromising their physical, photophysical, and imaging properties. Although the use of responsive brushes for nanotheranostic remains in its early stages, in this review, the aim is to present how the systems developed to date can be combined to control sensing, imaging, and controlled delivery of therapeutics. The recent developments for such design and associated methods for the synthesis of responsive brushes are discussed. The responsive behaviors of homo polymer brushes and brushes with more complex architectures are briefly reviewed, before the applications of responsive brushes as smart delivery systems are discussed. Finally, the recent work is summarized on the use of responsive polymer brushes as novel biosensors and diagnostic tools for the detection of analytes and biomarkers.systems closely resembles that of CO2 transport through an aqueous solution. We have interpreted this to mean that the hydrate and the matrix mineral surfaces are separated by liquid-containing channels. These channels will serve as escape routes for released natural gas, as well as distribution channels for injected CO2.

Nanotheranostics integrate diagnostic and therapeutic functions in one system and have received significant attention in the past few decades for the improvement of diagnosis, and the prevention and treatment of diseases.[1, 2] Advances in nanotheranostics benefit greatly from deeper understanding of the interactions between nanomaterials and biological systems, the refinement of multifunctional nanohybrids for simultaneous diagnosis and therapy, and the ability to harness the unique physicochemical properties of nanomaterials for specific and selective detection and treatment of diseases.[3] Although having shown promising results in many in vitro and in vivo studies, the concept of nanotheranostics remains a new paradigm and its clinical use is still in its infancy. In addition to challenges in commercialization, one of the key issues related to the translation of nanotheranostics remains the control and understanding of nano-bio interactions.[2] Upon interaction with biological systems, the physiological properties of nanoparticles determine their stability, pharmacokinetics, biodistribution, and toxicity profiles.[4] Those are crucial parameters for assessing their biocompatibility and avoid any adverse immunoreaction or inflammation,[5] but also improve efficacy as diagnostic and therapeutic tools. Hence, different techniques and modification strategies of nanomaterials have

been developed and characterized to overcome these limitations. Historically, the decoration of nanomaterials with polymers has been particularly successful to tailor and design the properties of these systems.[6] This includes self-assembly of monolayers,[7] the stabilization of nanoparticles via ligand exchange methods,[8] and the coating with polyelectrolyte assemblies.[9] These strategies have enabled the improved dispersity, prolonged circulation time via PEGylation,[10] enhanced cellular uptake and loading efficiencies of the therapeutics,[11] and increased availability to link with other biomolecules for targeting[12] or other purposes.

In particular, polymer brushes, defined as thin polymer coatings in which individual polymer chains are tethered by one chain end to a solid interface, are considered among the most powerful tools to control interface properties. Polymer brushes generated via the “grafting from” approach, in which initiating moieties are coupled to surface and allow the growth of polymer chains, are extremely attractive for the precise design of biomaterials and control over bio-nano interactions. A number of controlled/“living” polymerization techniques, in particular those based on radical chemistry have been applied to generate such coatings on various types of substrates.[13] It enables the grafting density, the thickness, and the chemistry of the coating to be manipulated very readily without altering the bulk mechanical properties of biomaterials.