



Design of core-shell magnetic nanocomposite by using linear and branched polycation as an ad-layer: Influences of the structural and viscoelastic properties

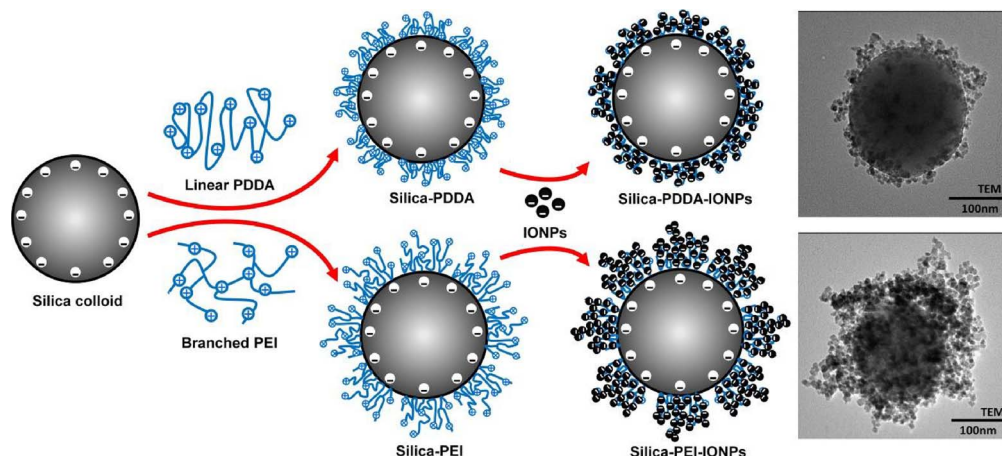
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GRAPHICAL ABSTRACT



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ABSTRACT

We outlined in this study the assembly of polyelectrolyte layers and its impact toward the immobilization of iron oxide nanoparticles (IONPs) onto silica colloid. The deposition kinetic of IONPs was investigated by the dynamic light scattering (DLS) and quartz crystal microbalance with dissipation (QCM-D) techniques. The structural properties of nanocomposite formed was examined by transmission electron microscope (TEM) and atomic force microscopy (AFM) to ascertain the development of core-shell morphology. From our results, polyelectrolyte layer constructed by branched poly(ethyleneimine) (PEI) was in more extended form compared to the linear PEI and poly(diallyldimethylammonium) chloride (PDDA). The layer thickness obtained for adsorbed polyelectrolyte was analyzed by both mean field and scaling approaches. From these analyses, it was found that the IONPs deposited onto the more stratified PEI polymeric network occurred at higher rate compared to PDDA layer. To demonstrate the potential application of this structure, the nanocomposite with different polyelectrolyte architecture was tested on dye removal by taking two different types of dye as the model system, namely cationic

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Methylene Blue (MB) and anionic Methyl Orange (MO). We hypothesized that the nanocomposite with more ramified structure (branched PEI), not high charge density, should have better pollutant removal capability.

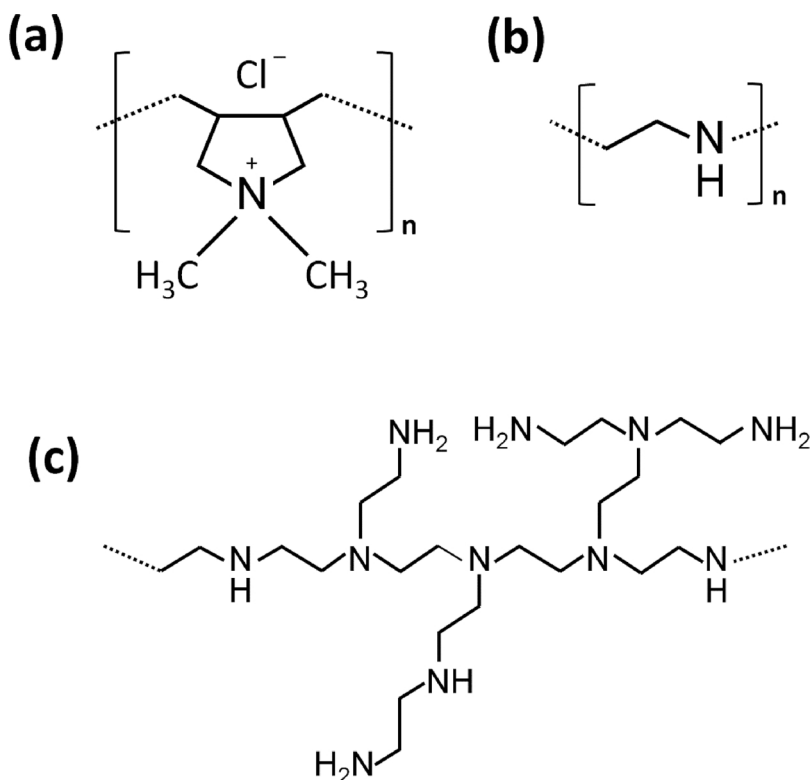
1. Introduction

Synthesis of nanocomposites have been widely studied due to the possibility of combining building-blocks with different materials and functionalities into one single nanostructure [1]. In this regards, core-shell structure has drawn immense attention and is one of the most popular configurations for nanocomposite due to its versatility for wide range of applications [2,3]. Under the core-shell morphology, the organic or inorganic core often plays a role as hosting material to control the size and shape of the composite; whereas the material responsible for targeted application normally residing within the outer shell [4,5]. As a result, the properties of the developed core-shell structures are noticeably different from the individual building block which made up the composite [6]. In fact, magnetic core-shell nanoparticle which is amenable for magnetic separation is in trend to be applied for water treatment purposes and has been proven as a promising remediation agent to remove various pollutants such as heavy metals [7] and organic pollutants [8–11].

In particular, strong flexible polyelectrolyte is an interesting and a predominant building block used to promote the incorporation of charged materials or colloidal objects to form core-shell nanomaterial. In fact, using polymers and nanoparticles mixture to form nanocomposites has been practiced for decades in industry [12–16]. Under this condition, the use of polyelectrolyte is extremely advantageous due to its abundant ionic bonds which could serve as a bridge to link the underlying substrates and film with any charged objects as outer shell [1]. On top of that, optical examination has also proven that there are no defects found in the fuzzy layered of this polyelectrolyte film [17]. Thereby, this structure has widely been used for many applications, such as biomedical [18], biosensor [19], and biotechnology [20]. Hence, the apparent contribution originated from the ‘softness’ related

to the viscoelasticity of the polyelectrolyte layers in developing multi-layer structure should be investigated and is the focus of this study.

The adsorption of polyelectrolyte onto the oppositely charged particles or surfaces has been investigated using various techniques [21]. Under this context, the adsorption of linear polyelectrolyte has been extensively exploited. For instance, Schwarz and coworkers investigated the linear PDDA adsorption amount on different charged surface, namely mica, silica and polymer latex by using electrokinetic measurement [22]. The adsorption mechanism of PDDA on the convex surface of silica particles and flat surface of silicon wafers was studied by Kokufuta and Takahashi [23] and Popa et al. [24]. According to their findings, the adsorption of strong and linear polyelectrolyte is mostly driven by electrostatic interactions where a flat configuration is formed under low ionic strength condition. As the ionic strength increased, loop and tails configurations are formed. For the case of weak polyelectrolyte with either linear or branched morphology, such as polyethylenimine (PEI) used in this work (see Scheme 1), the adsorption kinetic and adsorbed layer structure can become much more complex [25,26]. For instance, Mészáros and coworkers reported the adsorption of PEI on charged surface by using reflectometry [26–28]. The authors studied the influence of pH and ionic strength on the adsorption mechanism and adsorbed amount by describing the basic adsorption characteristic of PEI. In their works, the adsorbed amount of PEI was found to be increased with pH or ionic strength. In general, the overall charge of the adsorbed polyelectrolyte neutralizes and overcompensates the surface charge, giving rise to a charge reversal effect. These protruding polyelectrolyte parts, which extended into the bulk solution, serve as anchoring points for oppositely charged organic or inorganic species while the other ends of chains remained positioned at the solid surface [29]. The different conformation and nature of the adsorbed polyelectrolyte, is believed to play an important role for the



Scheme 1. Schematic structure of (a) linear PDDA, (b) linear PEI and, (c) branched polyethylenimine.

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