Polyelectrolyte Coated Clay Nanotubes With pH Controlled Release

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Statement of Purpose: Halloysite nanotubes (HNTs) exist abundantly throughout the natural world and have versatile hollow tubular structures composed of two-layered aluminosilicates.[1][2] The geometrical structure and surface charges of HNTs allow it to be loaded and nanocoated with a variety of materials, such as drugs and bioactive macromolecules and polymers, for sustained and extended releases. HNTs exhibit high levels of biocompatibility and very low cytotoxicity, making it an ideal candidate for new drug delivery systems.[3] The incorporation of nanocoatings on HNTs offer more possibilities for target and trigger-responsive drug delivery platforms.

Methods: PVP/PAA multilayers were organized on halloysite nanotubes at pH3 through the Layer-by-Layer technique. Vials with of HNTs were filled with water at pH 3, shaken, centrifuged, and decanted. After washing, the vials were filled with polyvinylpyrrolidone solutions (PVP 1mg/mL at pH3). Tubes were mixed for five minutes and centrifuged. The PVP solution was decanted and the vials were filled with water, which were adjusted to pH3. The tubes were mixed for three minutes, centrifuged, and water was decanted. This wash step was repeated. Next, the vials were filled with poly-acrylic acid solutions (PAA 3mg/100mL at pH3). Tubes were mixed, centrifuged, and decanted. The cycling of PVP and PAA coatings were repeated until the desired polyelectrolyte architectures were created. Alizarin red (AZR) and methotrexate (MTX) were infused within layers four and five, and the drug releases were examined under pH 3, 7.4 and 9.4. Releases from polyelectrolyte coatings were measured by UV-Visible spectroscopy (AZR- λ 259 nm and MTX- λ 307 nm). Nanocoated HNTs surface characteristics were examined with a scanning electron microscope.

Results: This study showed controlled pH-dependent releases of a model drug, alizarin red (AZR), and methotrexate (MTX) from polyelectrolyte multilayers, which included polyvinylpyrrolidone (PVP) and poly-acrylic acid (PAA). UV-visible spectroscopy results showed that the stability of the nanocoatings and the release of drugs were greatly influenced by the pH of immersing solution and stock solutions.

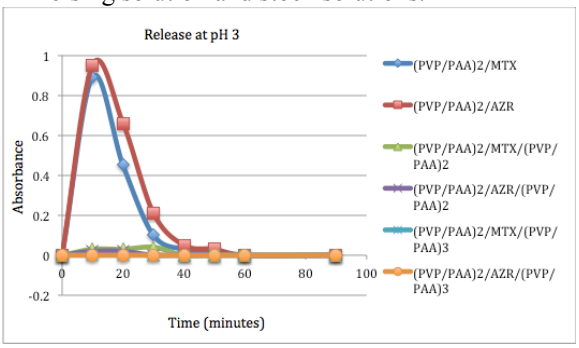

Figure 1. MTX and AZR releases from coatings at pH 3

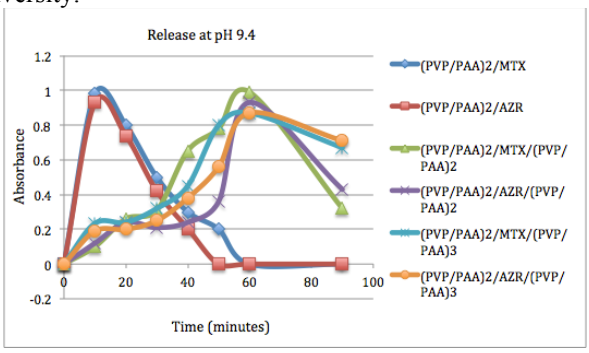

Figure 2. Increase of MTX and AZR releases from coatings at pH 9.4

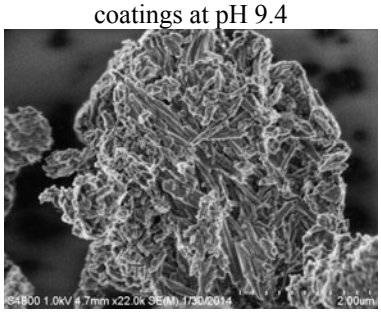

Figure 3. Scanning electron microscopic image of the polyelectrolyte coated HNTs.

Conclusions: In this study, we took advantage of the outside polyelectrolyte multilayer's surface area and porosity, rather than the inside luminal space of halloysite nanotubes. This approach has more advantages for complex delivery systems that could include multiple drugs stored within polyelectrolyte layers and halloysite nanotube lumens. It is suggested that these molecular architectures may have potential applications in nanoscale trigger-responsive drug delivery systems.

References:

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