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Halloysite Clay Nanotubes as carriers for loading photosensitizer molecules

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Halloysite nanotubes (HNTs) are natural aluminosilicate clay with a hollow tubular structure that enables loading and sustained release of biomacromolecules and drugs. A significant advantage of HNTs is their easiness to obtain and low price in comparison with other nanomaterials, like carbon nanotubes [1].

Among the minerals, halloysite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \cdot n\text{H}_2\text{O}$), a naturally forming two-layered aluminosilicate with a hollow nanotubular structure, consists of one alumina octahedron sheet and one silica tetrahedron sheet in a 1:1 stoichiometric ratio. The external surface is composed of siloxane (Si-O-Si) groups, whereas the internal surface consists of a gibbsite-like array of aluminol (Al-OH) groups. The different chemical constitution determines the negatively charged outer surface and positively charged inner lumen within a certain pH range (Figure 1) [2].

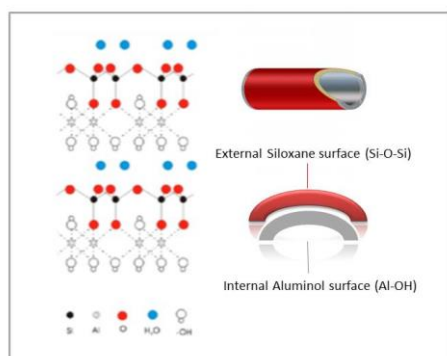


Figure 1. Composition and structure of a halloysite nanotube.

The environmental friendly and biocompatible nature enables halloysite nanotubes (HNTs) to be a promising nanomaterial for developing organic/inorganic composites in the biomedicine field [3].

Halloysite nano tubes have been used for entrapment and subsequent release of different drugs: Nifedipine (antianginal), Furosemide (antihypertensive), and Dexamethasone (corticosteroid) [4].

Halloysite is a biocompatible material but because of the aluminosilicate chemistry should not biodegrade. Therefore, its usage in medicine may be restricted to oral dosing, dental uses and dermal application. Photodynamic therapy (PDT) and related techniques are mainly aimed at topical diseases because the main requirement for efficient treatment is illumination of the treated area. Although light can be delivered within the body via optical fibres nowadays, the use of PDT is still mostly limited to skin diseases. However, delivering PDT in cases of skin diseases is not a straightforward task owing to the high structural

complexity and efficient barrier properties of the skin. Therefore, delivery of drugs and photosensitizers (PSs) into the skin is still a problematic area. The major limitation of topical PDT is the poor penetration of PSs through biological barriers, like the skin. A series of PSs are under study in PDT experiments, however, most of them are relatively hydrophobic and have low capacity of accumulation in target tissue. Recently, studies have been focused on the development of different strategies to overcome these difficulties, including nanocarriers to delivery.

In the present work we present the interaction between some potential photosensitizers for PDT bearing squaraine structure with HNTs (Figure 2). Squaraines, when exposed to light of specific wavelength, are capable of generating singlet oxygen and other cytotoxic reactive oxygen species (ROS) that kill tumor cells [5].

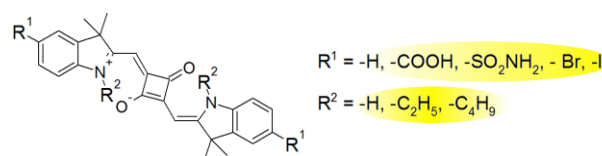


Figure 2. Squaraine structures

The study of the interaction of the organic molecules and HNTs was performed under different experimental conditions: by simple contact or by vacuum/air entrapment. The Entrapment Efficiency (EE%) was calculated. The release rate of squaraines was analysed at physiological conditions. Cytotoxicity and cell penetration on MDCK cell line of the complex squaraine/HNTs were performed.

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