Design of Functional Polymer Hybrid Hydrogels Based on Imogolite and Halloysite Clay Nanotubes

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イモゴライトとハロイサイトナノチューブを用いた機能性ポリマーハイブリッドハイドロゲルの設計

区　　分　：甲

論　文　内　容　の　要　旨

Recently, clay nanotubes have found an important role in industrial areas because of its multifarious applications. Their unique tubular structure and chemical properties make them useful in several industrial applications such as catalysis, waste adsorption, gas storage, polymer nanocomposite, drug loading and delivery, and anti-corrosion loading and release. Clay nanotubes have various advantages such as large specific surface area, high surface activity and mechanical strength. Moreover, they are low cost and environmentally friendly materials which can be alternatives for expensive nanotubes with time-consuming preparation and small amount production. Above all, imogolite and halloysite nanotubes have been actively researched over the past decades because of their easy modification process resulting in high functional materials in nanocomposites with high mechanical strength.

One-dimensional clay nanotubes have been actively exploited as biological scaffolds component due to their high specific surface area, rigidity, and biocompatibility for boosting mechanical properties of scaffolds and cellular interaction. Thus, one-dimensional clay nanotubes provide promising approaches in biomedical areas as the scaffold materials with the superior mechanical properties and good biocompatibility. Halloysite and Imogolite clay nanotubes allow the design of smart composite materials with the controlled and sustained release of chemically active agents due to their unique surface chemistry and tubular structure. Their inner lumen can store and release molecules in a controllable manner making these nanocarrier attractive for applications in drug delivery and regenerative medicine, self-healing composites, and antimicrobial materials.

In chapter 1, background of clay nanotubes technology and their structure and advantage, and the motivation of clay nanotube research, and scope of this study were described. In chapter 2, hydrophilic clay nanotubes as a structural component in hyaluronic acid polymer network were studied. Imogolite network provided the mechanical strength and efficient energy dissipation and stress relaxation to the crosslinked polymer network when the deformation was given. In chapter 3, effective interaction with halloysite and hyaluronic acid was investigated. Negative charge of both materials hinders the interaction between halloysite and hyaluronic acid polymer network resulting in weak mechanical strength of hydrogels. To overcome this repulsive interaction, acrylate group was functionalized to the surface of halloysite which plays a role in crosslinking point to the polymer chain of acrylate group modified hyaluronic acid via photocrosslinking. In chapter 4, hydrophobic curcumin was loaded into the halloysite lumen. Halloysite lumen provided the safety spot to the curcumin molecule for prohibiting early degradation and releasing. Whereas, halloysite lumen was selectively loaded with Schiff base reacted curcumin-dopamine compound. The catechol group of dopamine is strongly adsorbed to the halloysite lumen, consequently they caused the sustained curcumin release at neutral pH. But when the pH went down, curcumin nanocarrier showed the pH responsive fast release. Such a design of long term and stimulus-responsive released architecture can provide a great opportunity to advance the drug delivery system for cancer treatment and prevention. Furthermore, when HNTs containing dopamine-curcumin were loaded into the hyaluronic acid hydrogels, released curcumin amount was abruptly diminished showing controlled curcumin release due to the tortuous path caused by dual barrier of tubular structure and polymer chain length where curcumin must pass through. In chapter 5, concluding remarks about clay nanotubes were given. In light of the result of the research, clay nanotubes can be a good candidate as a scaffold material and a nanocarrier for biomedical applications.

〔作成要領〕

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