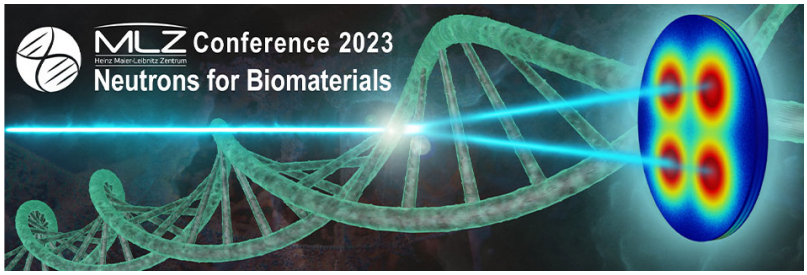

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Multi-responsive nanostructured materials for the targeted delivery of anticancer agents

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Abstract. Novel, multifunctional nanoparticles and hydrogels that exhibits a unique set of properties for the effective treatment of cancer are presented. The materials are comprised of polypeptidic and polyethylene oxide polymers that are a non-cytotoxic polymer. The amphiphilic hybrid materials assemble in aqueous media to form micelles or vesicles, comprised of an outer hydrophilic corona of PEO chains, and a pH- and redox- responsive hydrophobic layer based on poly(L-histidine) (PHis) and poly(L-cystein) (PCys). Due to the presence of the thiol groups of PCys, a crosslinking process was achieved further stabilizing the nanoparticles (NPs) formed. Dynamic Light Scattering, Static Light Scattering and Transmission Electron Microscopy were utilized to obtain the structure of the NPs. Moreover, the pH- and redox-responsiveness in the presence of the reductive tripeptide of glutathione (GSH) was investigated at the empty as well as the loaded NPs. The ability of the synthesized polymers to mimic natural proteins was examined by Circular Dichroism, while the study of zeta potential revealed the "stealth"properties of NPs. The anticancer drug doxorubicin (DOX) was efficiently encapsulated in the hydrophobic core of the nanostructures and released under pH- and redox-conditions that simulate the healthy and cancer tissue environment. It was found that the topology of PCys significantly altered the structure as well as the release profile of the NPs. Finally, in vitro cytotoxicity assay of the DOX-loaded NPs against three different breast cancer cell lines showed that the nanocarriers exhibited similar or slightly better activity as compared to the free drug, rendering these novel NPs very promising materials for drug delivery applications.
Hybrid-polypeptidic materials formed injectable in situ forming quickly self-healing hydrogels, responsive to alteration of pH and increase of temperature. The connection between the alteration of secondary structure of the polypeptides with the viscoelastic behavior was revealed by means of Rheology and Circular Dichroism. Small-Angle Neutron Scattering and Scanning Electron Microscopy were employed to shed light to the structure of the polymers and how it affects their rheological properties. The results suggest that these biomaterials have the potential to be used in a number of bioapplications like drug delivery.

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