*T2*-weighted magnetic resonance imaging (*T2*-MRI) have been intensively studied as an imaging tool in medical research and clinical diagnosis. [1, 2] It measures the response of water molecules in the presence of magnetic fields, more precisely, the variation of proton densities and transversal relaxation times of testing subjects. [3, 4] Exogenous MRI contrast agents are often required for the enhancement of resolution because of their capabilities of shortening the relaxation time of water proton. [5]

The most commonly used *T2* contrast agents in clinic are dextran coated superparamagnetic iron oxide nanoparticles (SPIONs) and gadolinium (Gd3+) chelates.[6] Gd3+ based contrast agents have been known to induce nephrogenic systemic fibrosis (NSF) in patients, which could impair kidney function.[7,8] SPIONs-based *T2* contrast agents (e.g., Feridex) have been showing much better biocompatibility and negligible toxicity, however their efficacies as the contrast agents still need further improve to achieve high resolution.[9-11]

The efficacy of *T2* contrast agents is evaluated by its proton relaxivity, *r2*, which is defined as the paramagnetic transverse relaxation rate (1/*T2*) enhancement of the water protons at unity concentrations.[4,5] The nature of core and coating both affect the *T2* properties of contrast agents, i.e., the material composition, core size,[12-15] and aggregation[16] of SPIONs. In earlier study, we have discussed the influence of coating thickness and hydrophilicity on *r2* relaxivity, and we found that *r2* value could be increased by reducing the thickness of polymeric coating, and we concluded that the higher *r2* could be obtained with the accelerated exchange of water molecules between the bulk phase and the adjacent layer surrounding SPIONs. [17] In this contribution, we developed a method to prepare monolayer coated SPIONs with ultrahigh *r2* relaxivity by reducing the thickness of superhydrophilic succinylated heparin (Su-HP) coating to only 2 nm. The dextran (Dex) coated SPIONs were prepared according to the reported procedure for Feridex, and natural heparin (HP) coated SPIONs were also prepared as the comparison, and we found that the *r2* value of Su-HP coated SPIONs (Su-HP-SPIONs, 460 mM-1 s-1)was significantly higher than heparin coated (HP-SPIONs, 299 mM-1 s-1) and dextran coated SPIONs (Dex-SPIONs, 147 mM-1 s-1). To our knowledge, the *r2* value of Su-HP-SPIONs is the highest for the pure iron based SPIONs with the hydrodynamic size less than 20 nm.[18]