Magnetic resonance imaging (MRI) is the widely used noninvasive clinical diagnosis modality that provides anatomical tissue images of high spatial resolution. The technique is based on $^1$H-NMR signal recording and thus, limited by low-sensitivity. To deal with the issue, imaging processes are often accompanied by prior administration of a paramagnetic substance, which is known as contrast agents (CA). Gd(III)–based stable and water–soluble complexes are most widely studied as CAs because of its seven unpaired electrons, long electronic relaxation time, and fast water exchange rate. While, the $r_1$ relaxivity values of the commercially available Gd(III)–based MRI contrast agents are in the range 4.4–5.2 mM$^{-1}$s$^{-1}$, with comparable thermodynamic and kinetic stability to that of commercially available Gd(III)–based CAs, the water soluble, aquated Gd(III) complexes of ligand H$_4$peada (2A) and H$_4$bedik (3A) provided higher $r_1$ relaxivity values of 6.08 mM$^{-1}$s$^{-1}$ and 7.30 mM$^{-1}$s$^{-1}$ at 1.41 T, 25 °C and pH ~ 7.4, respectively. Moreover, in contrast to the current trend of low $r_1$ relaxivity values of the commercially available Gd(III)–based MRI contrast agents at higher field strength, the complex 3A provided higher $r_1$ relaxivity value even at higher magnetic field strength, holding promise regarding the development of MRI imaging probes that can be even used at higher magnetic fields. In the search of safer MRI contrast agent for patients having severe renal diseases, Mn(II) complexes of ligand H$_4$bedik (4A) and H$_2$pmpa (5B) with one and two inner sphere water molecules showed higher $r_1$ relaxivity values of 3.11 mM$^{-1}$s$^{-1}$ and 5.88 mM$^{-1}$s$^{-1}$ at 1.41 T, 25 °C and pH ~ 7.4 respectively. These higher relaxivity values along with their higher thermodynamic stability make them promising candidates for the development of effective MRI contrast agents for safer and emerging applications in MRI.