

**Figure S1.** High-resolution XPS spectra of PZ-PAA@Fe<sup>3+</sup> bilayer hydrogel.

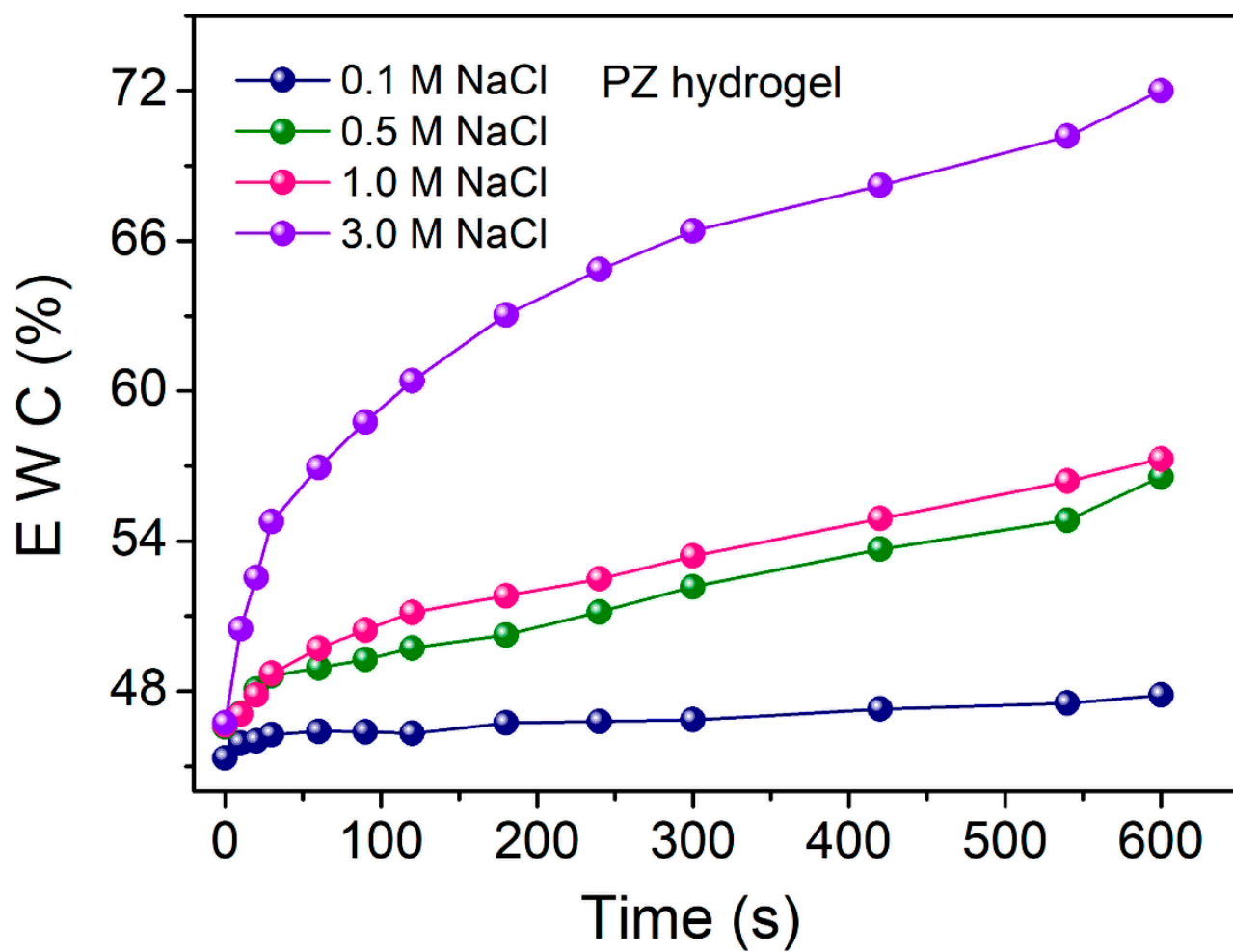
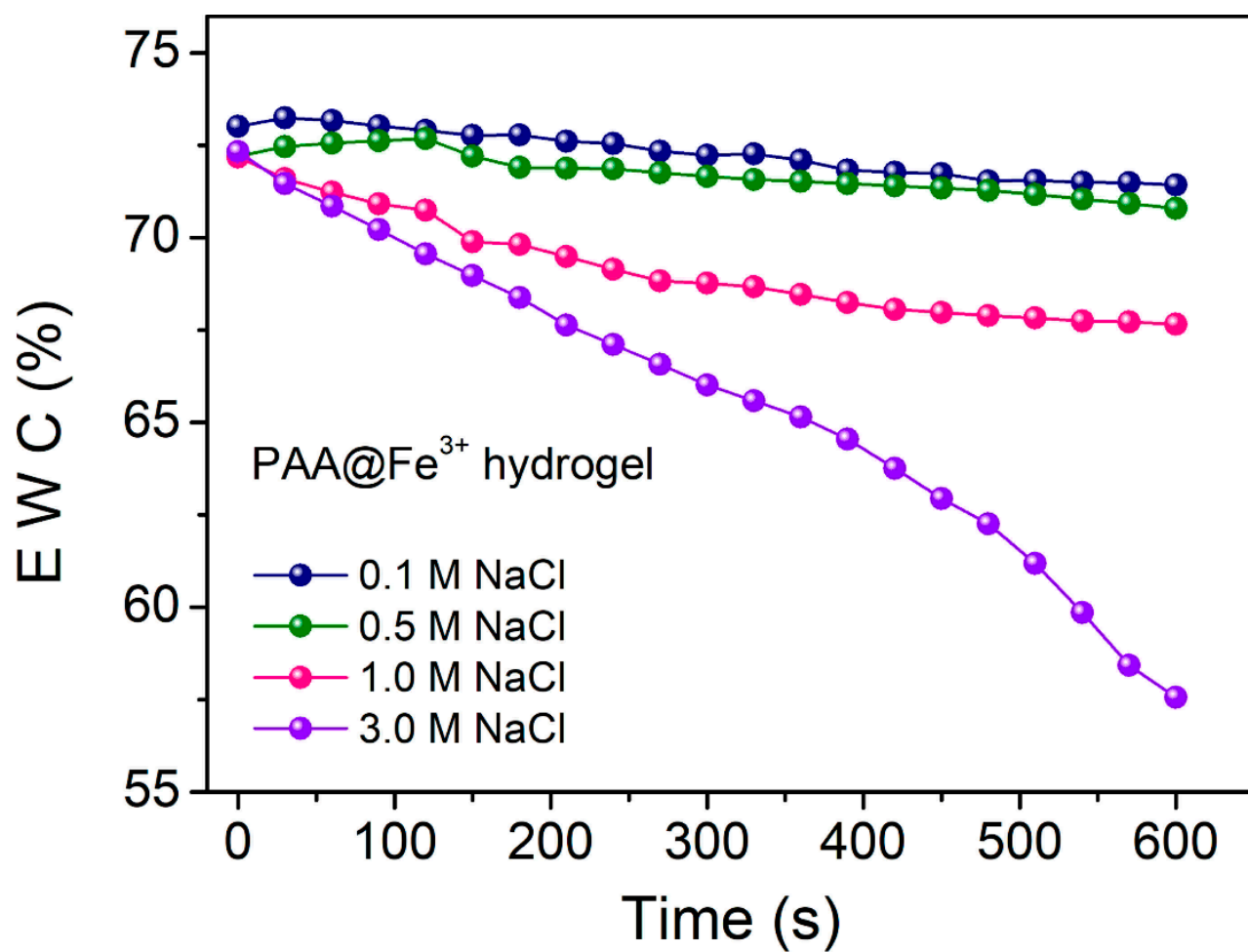
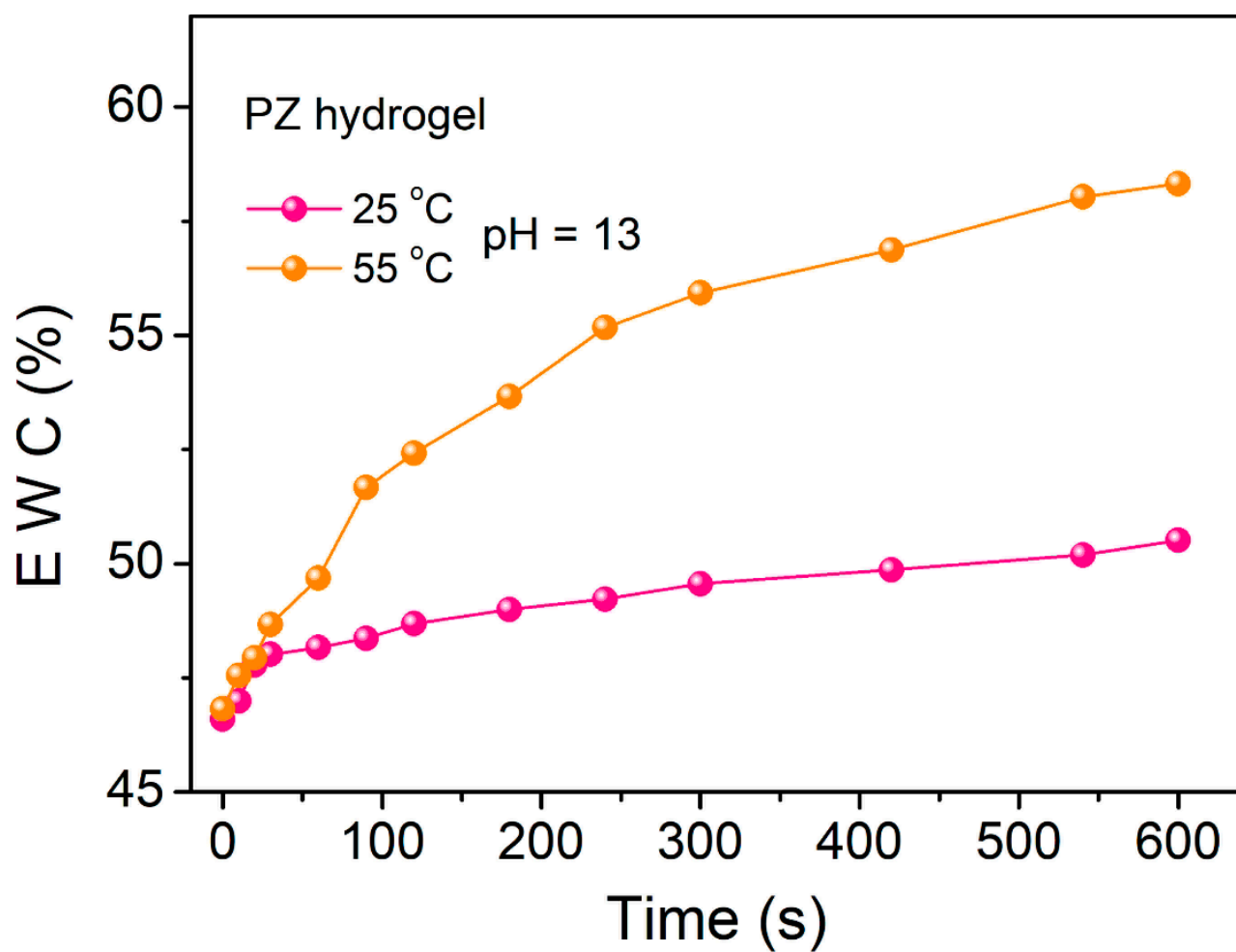


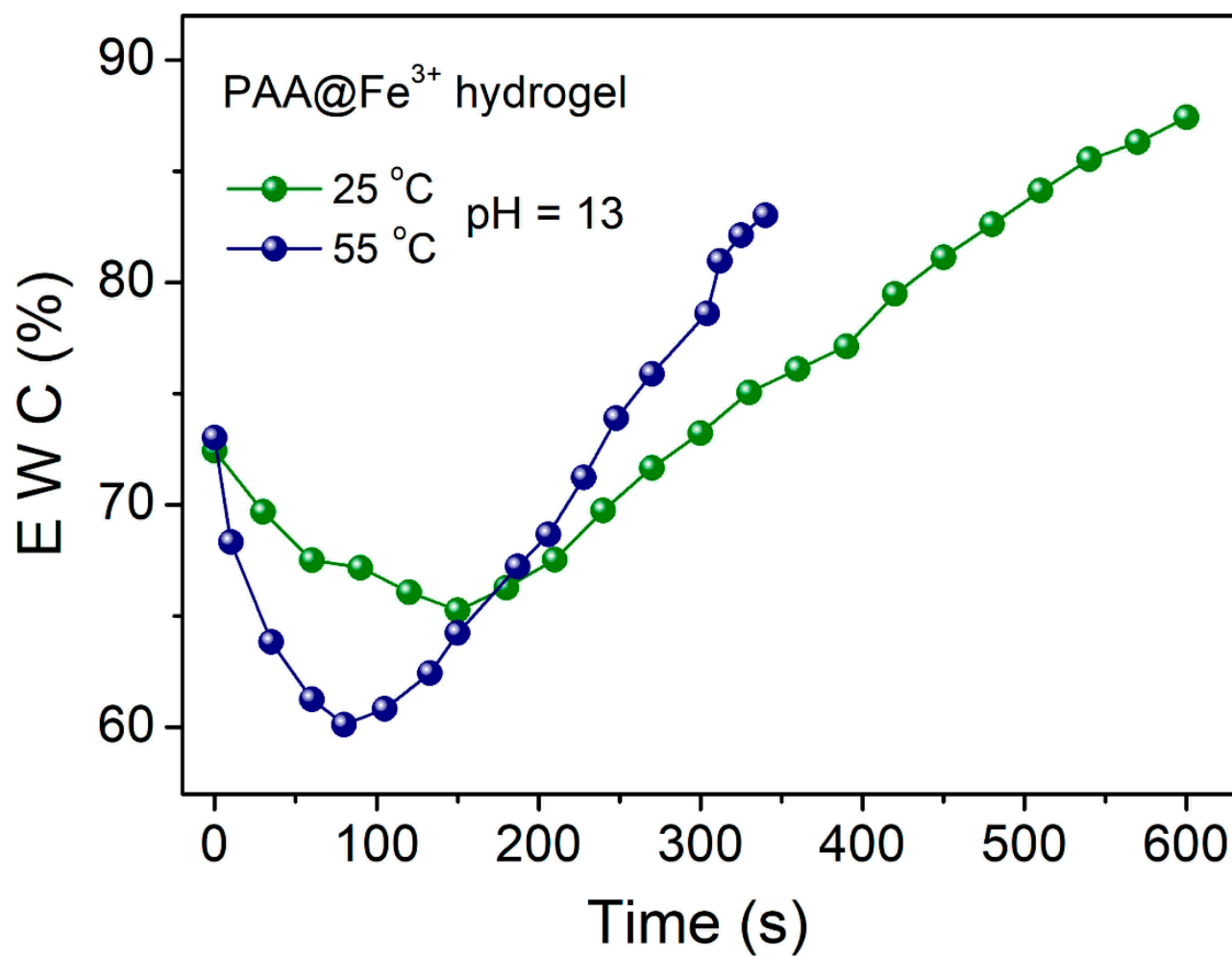
Figure S2. Equilibrium water contents of PZ hydrogel in response to NaCl concentrations.



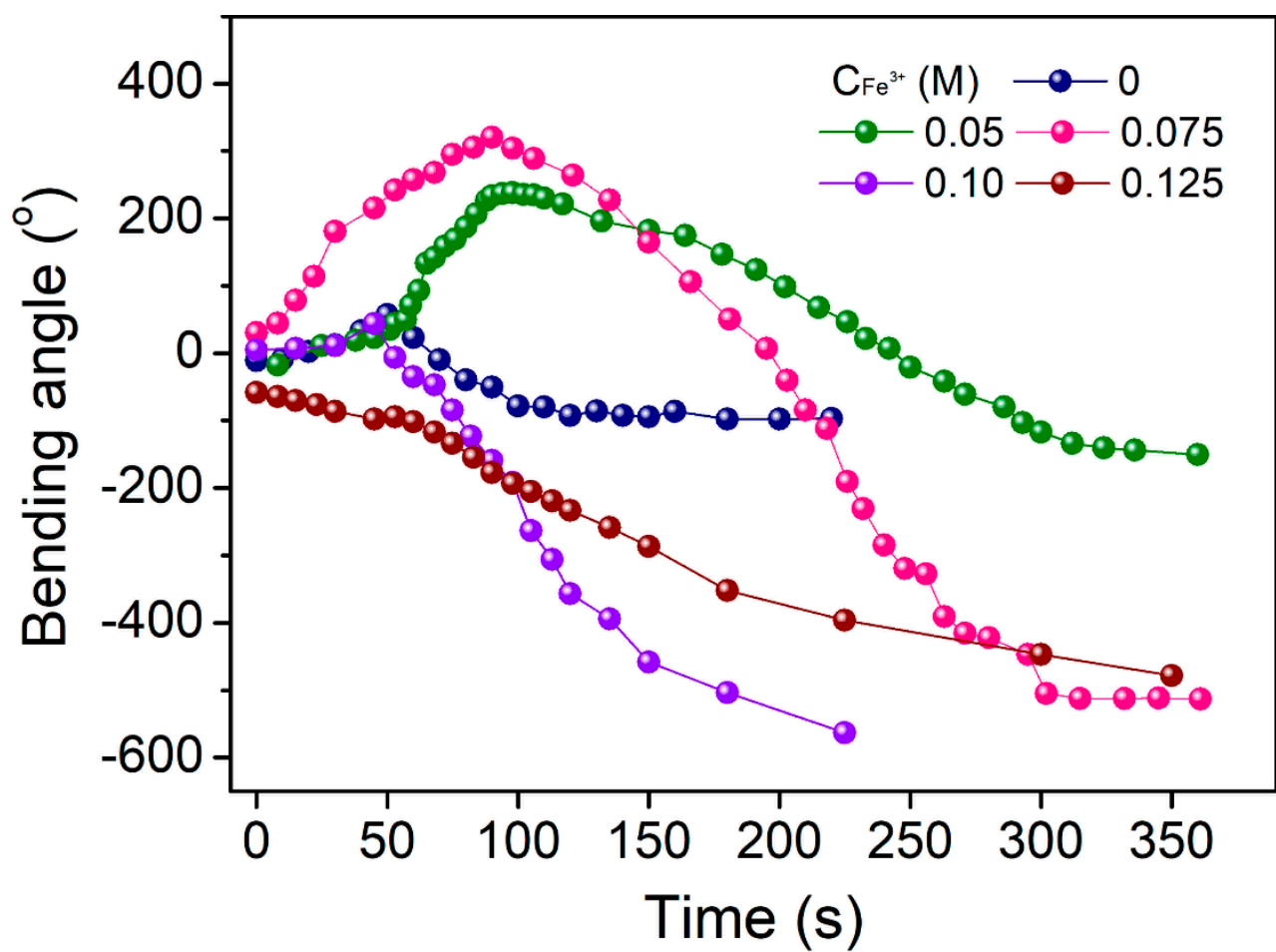
**Figure S3.** Equilibrium water contents of PAA@Fe<sup>3+</sup> hydrogel in response to NaCl concentrations.



**Figure S4.** Equilibrium water contents of PZ hydrogel in response to the environmental changes from cool alkali liquors at 25 °C to hot alkali liquors at 55 °C.



**Figure S5.** Equilibrium water contents of PAA@Fe<sup>3+</sup> hydrogel in response to the environmental changes from cool alkali liquors at 25 °C to hot alkali liquors at 55 °C.



**Figure S6.** Comparison of bending behavior of the bilayer hydrogel strips coordinating with different  $\text{Fe}^{3+}$  concentrations.