

## Revealing the metal-ligand coordination effects on the structure modifications for balanced tensile modulus and self-healing of polyurethane films

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**Polyurethane** (PU) films are potential candidate substrate for next-generation stretchable electron devices that attract much attention. Both tensile modulus and selfhealing of PU films are anticipated yet seemingly mutual excluded properties. Here, metal-ligand coordination is proposed to modify the crystalline and nanostructural features of PU films for concomitantly improved tensile modulus and self-healing. PU films of bpyPTD are prepared from reaction of bpy with PTD of different polarity of solvents such as THF or DMF/THF. Metal precursors solutions of Zn, Ni, were selectively mixed into the bpyPTD solution for cast of the final product films of MbpyPTD, with M = Ni, Zn. X-ray absorption is used to elucidate how the critical metalligand coordination between Zinc (II)/Nickel (II) and bipyridine (bpy) could effectively crosslink the segmented PTD into network structures of Zn-bpyPTD and Ni-bpyPTD. The network structures of the M-bpyPTD are tested under film stretching at low temperatures and high humidity, using concomitant small- and wide-angle X-ray scattering (SAXS and WAXS). Density (or mean spacing) of the metal-ligand coordination cites could be enriched with the higher polarity solvent for improved tensile modulus. By contrast, improvement of the self-healing capability is reached with enhanced metal-ligand coordination strength of bpy with Ni. Accordingly, Ni-bpyPTD could be fabricated into pyramidal pressure sensors, showing characteristic pressure response with good cyclability for promising applications as electronic skins.

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