A Stretchable Polysiloxane Elastomer with Self-Healing Capacity at Room Temperature and Solvatochromic Properties

Experimental section

Chemicals

3-Amlnopropyl(diethoxy)methylsllane and 2-aldehydepyridine were purchased from Aladdin (China). Dimethylsiloxane cyclic tetramer were purchased from Datian chemical additives Institute (China). Cobalt chloride was purchased from Tianjin Heowns Biochem LLC (China). The remaining chemicals and solvents were purchased from Chengdu Kelong Chemical Co. Ltd (China). All of the chemicals were used as received without further purification.

Materials and general measurements

¹H NMR spectra were recorded on a Bruker 600 NMR spectrometer in deuterated solvents at room temperature. Gel permeation chromatography (GPC) experiments were performed on an Agilent 1200SERIES chromatographic instrument (USA). Infrared Spectrum were recorded on a Thermo Fisher IS-5 infrared spectrometer. Differential Scanning Calorimetry (DSC) experiments were performed using a DSC 200 F3 from NETZSCH Instruments (GER) at -150~100 °C and heating and cooling speed of 10 °C/min under N₂ atmosphere. Thermogravimetric analyses (TG) were performed on a simultaneous SDT Q600 thermal analyzerfrom TA Instruments (USA). The temperature range is room temperature to 700 °C with a heating rate of 10 °C/min under N_2 atmosphere. Strain-stress curves and Dynamic mechanical analyse (DMA) curves were obtained by using Q800 (TA Instruments, USA). Strain-stress curves were recorded by using tensile mode at room temperature with loading rate of 6 mm min⁻¹. The original length for DMA measurements were set as 10 mm. DMA curves were recorded in the temperature range of -150~50 °C at temperature elevating rate of 5 °C/min. The strip dimensions were $5 \times 0.5 \times 0.1$ cm³. UV-vis spectroscopy of silicon elastomer films, which were fabricated by casting of Py-PDMS/CoCl₂ solution in THF, was conducted on SHIMADZU Solidspec-3700 (JAP). Stretching experiment

was conducted as following procedure. First, the sample was stretched by predetermined stress. Afterwards, the stress was removed. The recovery rate is calculated by $(l_1-l_0)/(l_2-l_0)$, where l_1 is elongated length, l_2 is recovered length and l_0 is initial length.

Syntheses of AP-PDMS

3-Aminopropyl(diethoxy)methylsilane (1.5 g, 1 mmol), Dimethylsiloxane cyclic tetramer (5 g, 2.5 mmol), hexamethyldisiloxane (1 μ L), KOH (65 mg) in H₂O (0.14 ml) and SOCl₂ (0.10 ml) were sequentially added into 50 mL flask. The solution was stirred at 90 °C for 3 hours under a nitrogen atmosphere. Afterwards, the mixture was treated under reduced pressure to remove small molecules. Then, the mixture was heated at 90 °C for another 5 hour and added by acetic acid (70 mg). The solution was treated under reduced pressure at 60 °C to yield transparent oil (5.5 g, 84.6 %).

Syntheses of Py-PDMS

To synthesize pyridine-functionalized PDMS, 2-aldehydepyridine (2.5 g) and AP-PDMS (5.5 g) was mixed in THF (20 ml). The reaction was conducted in flux at 70 °C for 24 hour. After reaction, the solvent was removed by rotary evaporation to give orange oil. This oil was dissolved in petroleum ether (15 ml) and washed with water (15 ml) three times. After drying with Na₂SO₄, the solvent was filtered and evaporated to dryness to give the product as a clear, slightly orange oil (5 g, 80 %).

Preparation of Co-coordinated silicon elastomers (Py-PDMS-Co)

Typical experiment to prepare Py-PDMS-Co is as following: a solution of Py-PDMS (0.1 g) in THF (1 mL) was mixed with 1 wt.% Cobalt chloride/ethanol solution according to predetermined Co/pyridine mole ratios. The solution was casted on polytetrafluoroethene (PTFE) mold and dried at room temperature to give strips with a size of $35 \times 8 \times 0.4$ mm³.

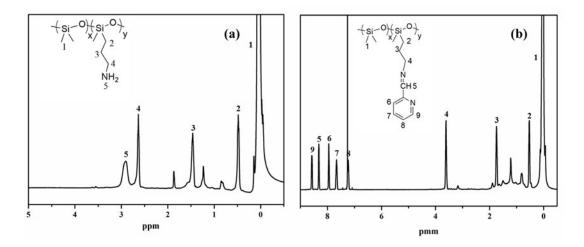


Figure S1. ¹H NMR spectra of a) aminopropyl-polydimethylsiloxane (AP-PDMS) and b) pyridine-polydimethylsiloxane (Py-PDMS).

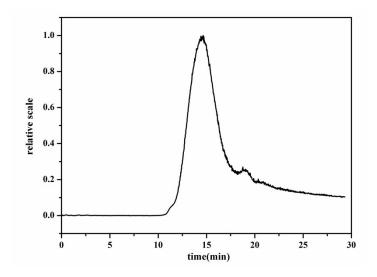


Figure S2. GPC curve of Py-PDMS.

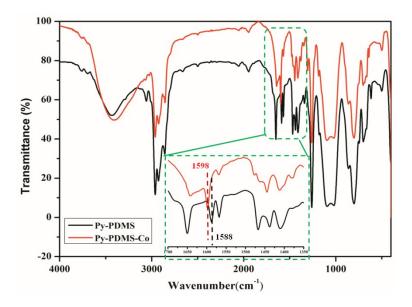


Figure S3. FTIR spectra of Py-PDMS and derived Co-incorporated Py-PDMS (Py-PDMS-Co)

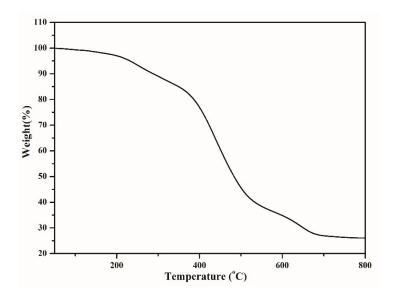


Figure S4. TGA spectrum of Co-incorporated Py-PDMS (Py-PDMS-Co-0.5)

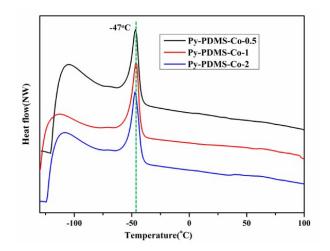


Figure S5. DSC curves of Py-PDMS-Co-0.5, Py-PDMS-Co-1 and Py-PDMS-Co-2.

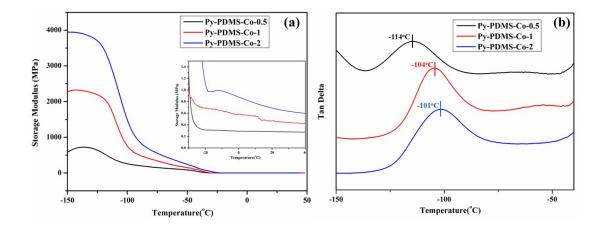


Figure S6. Storage moduli vs temperature (a) and Tan(delta) vs temperature (b) curves of Py-PDMS-Co elastomers.

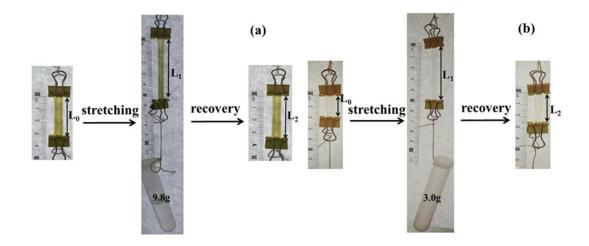


Figure S7. Stretching experiment pictures of Py-PDMS-Co-0.5 (a) and Py-PDMS-Zn-0.5 (b).

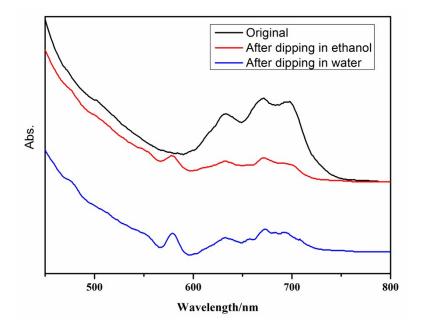


Figure S8. UV-vis spectra of original Py-PDMS-Co-0.5 and that dipped in ethanol and water.