Electronic Supplementary Material (ESI) for Polymer Chemistry.

A Facile Approach to Thermomechanically Enhanced Fatty Acid-Containing Bioplastics Using Metal-Ligand Coordination

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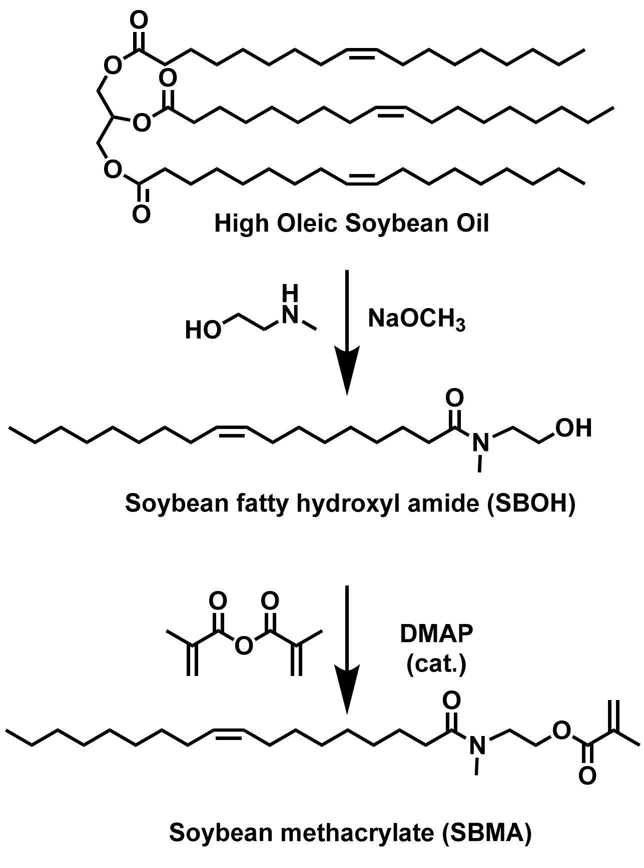
SUPPLEMENTARY INFORMATION



Scheme S1. Synthesis of SBMA monomer starting from soybean oil.

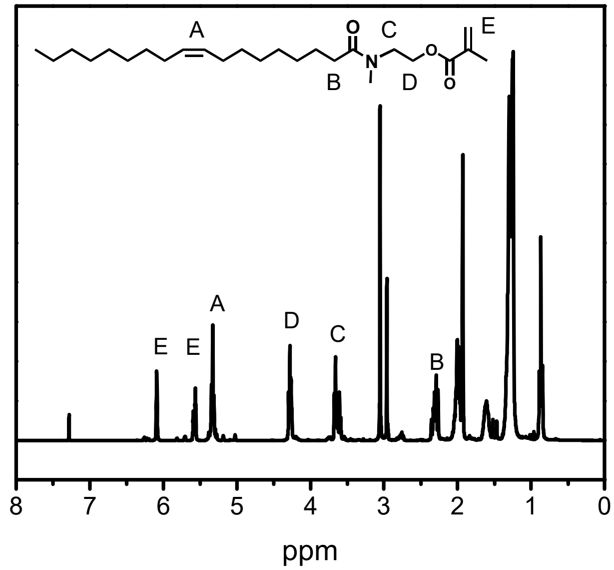


Figure S1. ^{1}H NMR spectrum of SBMA monomer.

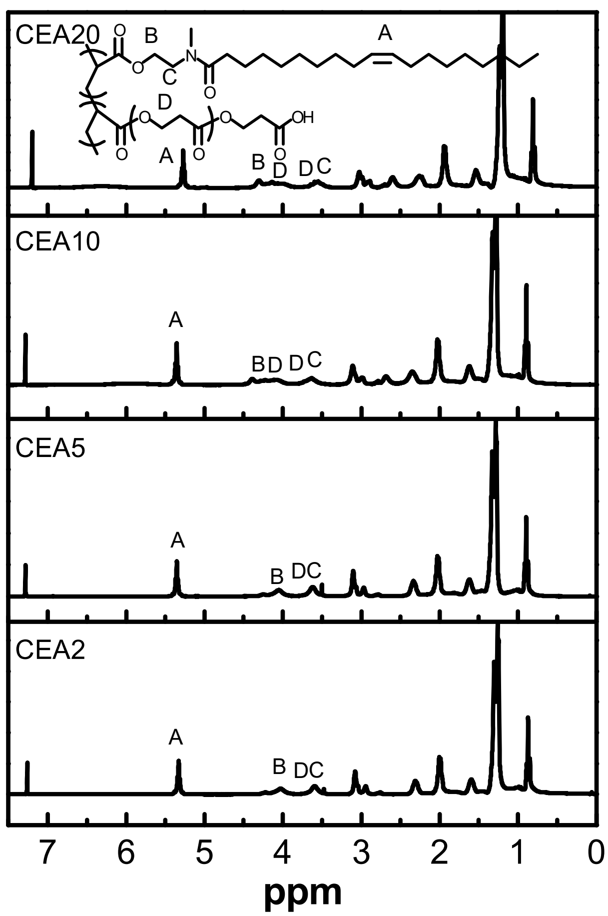


Figure S2. ^{1}H NMR spectra of soybean oil based copolymers.

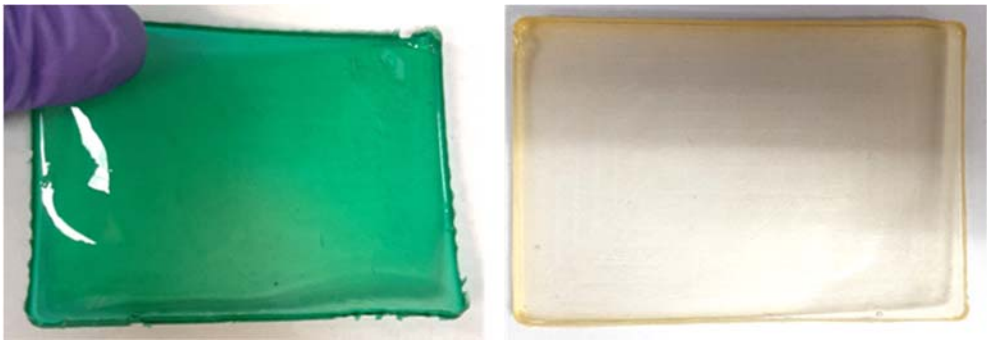


Figure S3. Photographs of (left) **CEA5Cu** and (right) **CEA5Zn**.

Table S1. Glass transition temperature (T_g) of copolymers and metal-coordinated polymers.

Sample code	Copolymer T_g (°C)	Copper Coordination T_g (°C)	Zinc Coordination T_g (°C)
CEA5	-5	39	40
CEA10	-3	52	56
CEA20	-1	66	69

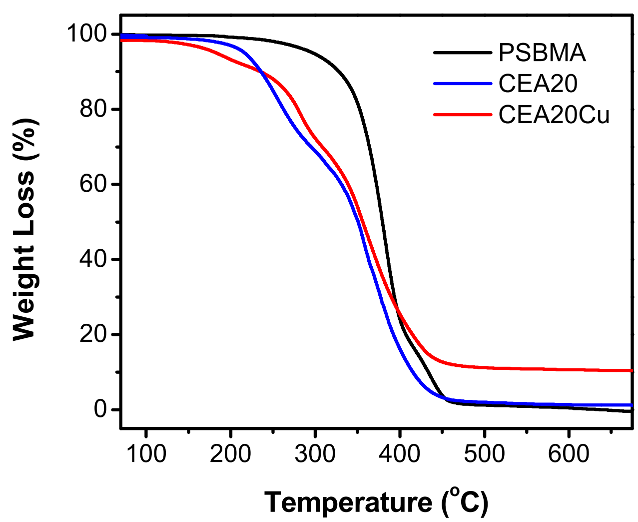


Figure S4. TGA curves of copolymers and metal-coordinated polymers.

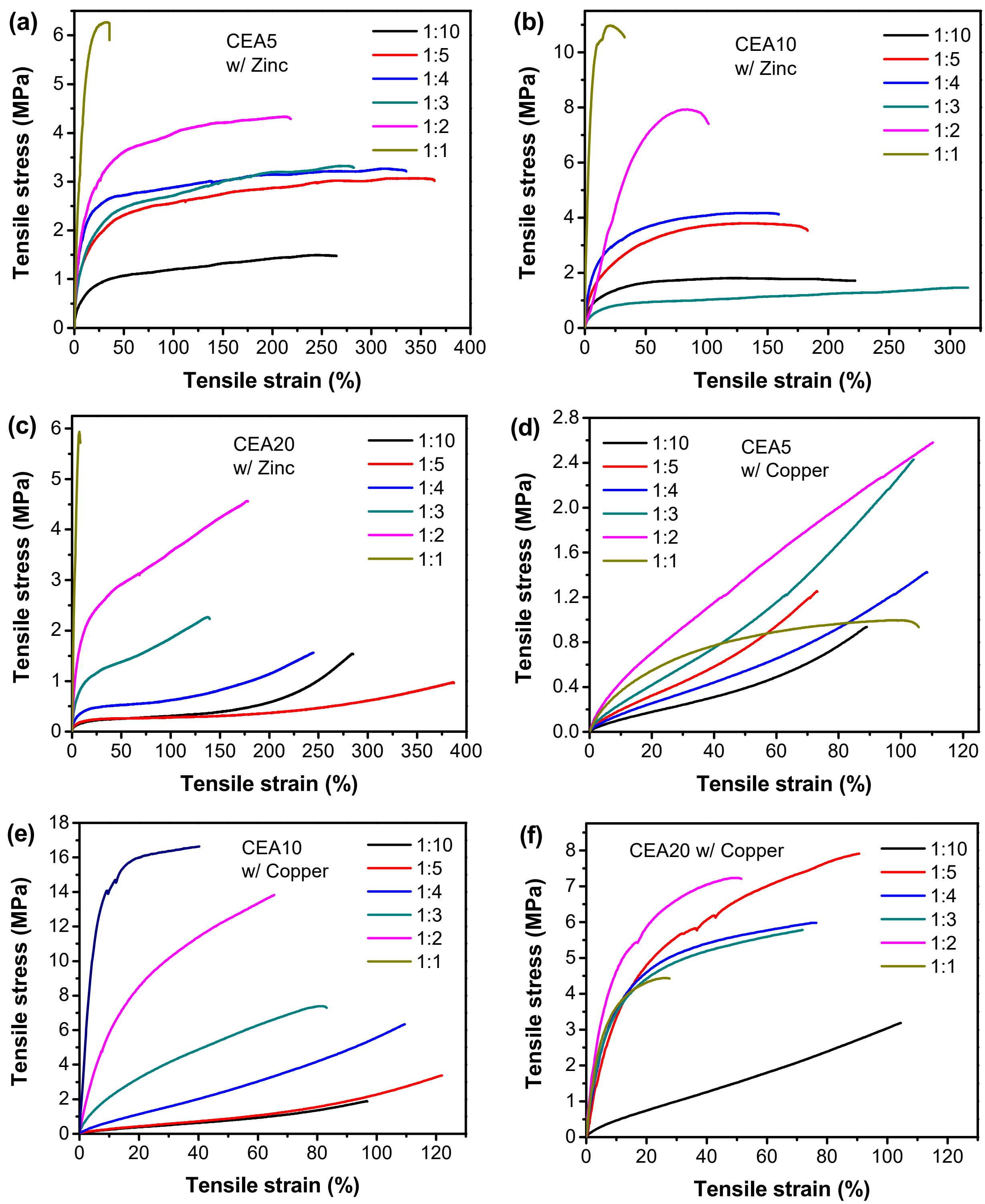


Figure S5. Tensile curves of all metal-coordinated copolymers at different metal-to-acid ratios. Using zinc salt for coordination with (a) **CEA5**, (b) **CEA10**, (c) **CEA20**. And using copper salt for coordination with (d) **CEA5**, (e) **CEA10**, (f) **CEA20**.

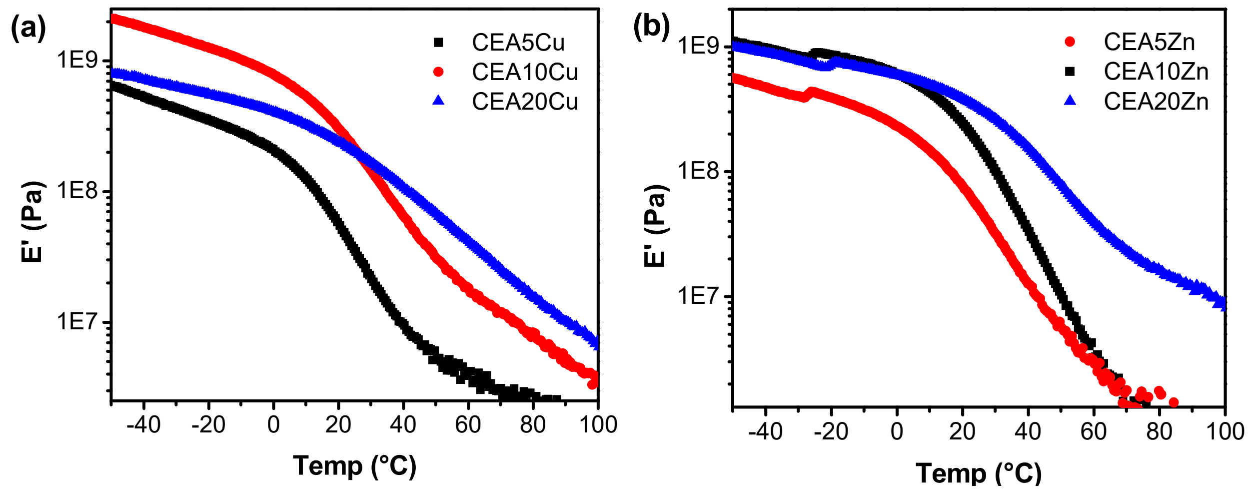


Figure S6. Storage modulus curves of all (a) copper-coordinated copolymers and (b) zinc-coordinated copolymers.

Table S2. Storage modulus data for all copper-coordinated copolymers.

Polymer	Metal	Storage Modulus at 25 °C (Pa)	Storage Modulus Plateau Temperature (°C)
P6	Copper	3.49E7	80
	Zinc	5.08E7	---
P7	Copper	2.07E8	100
	Zinc	1.61E8	---
P8	Copper	2.02E8	120
	Zinc	3.21E8	100

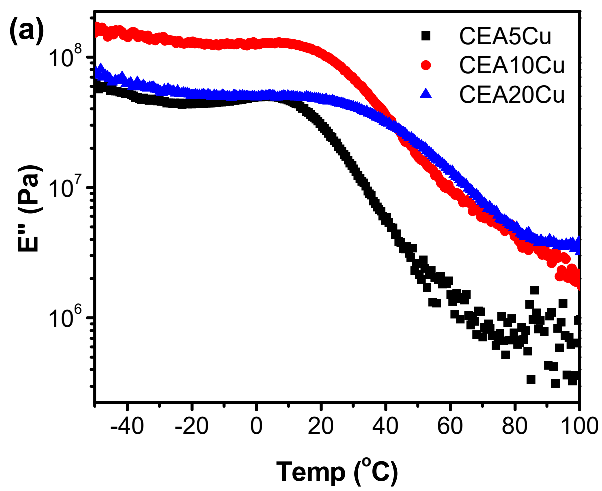


Figure S7. Loss modulus curves of (a) all copper-coordinated copolymers and (b) all zinc-coordinated copolymers.

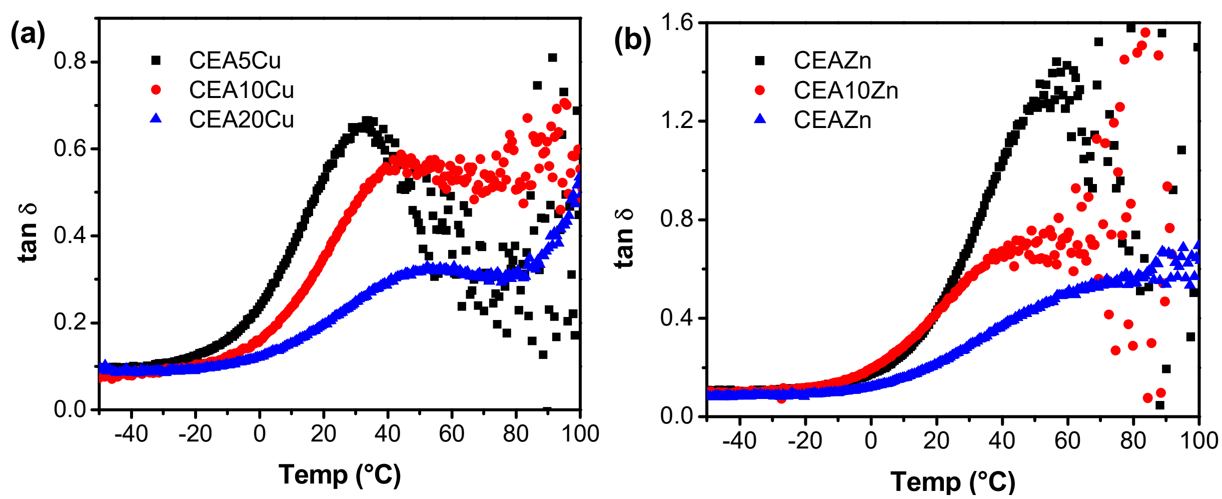


Figure S8. Tangent delta curves of (a) all copper- and zinc-coordinated copolymers.

Table S3. Dissolution studies using various solvents for **CEA20Cu** and **CEA20Zn** films.

Solvent	CEA20Cu	CEA20Zn
THF	Yes	Yes
Toluene	Yes (heated)	Yes
DMF	Yes (heated)	Yes (heated)
Water	No	No
Chloroform	No	Yes
Dichloromethane	No	Yes

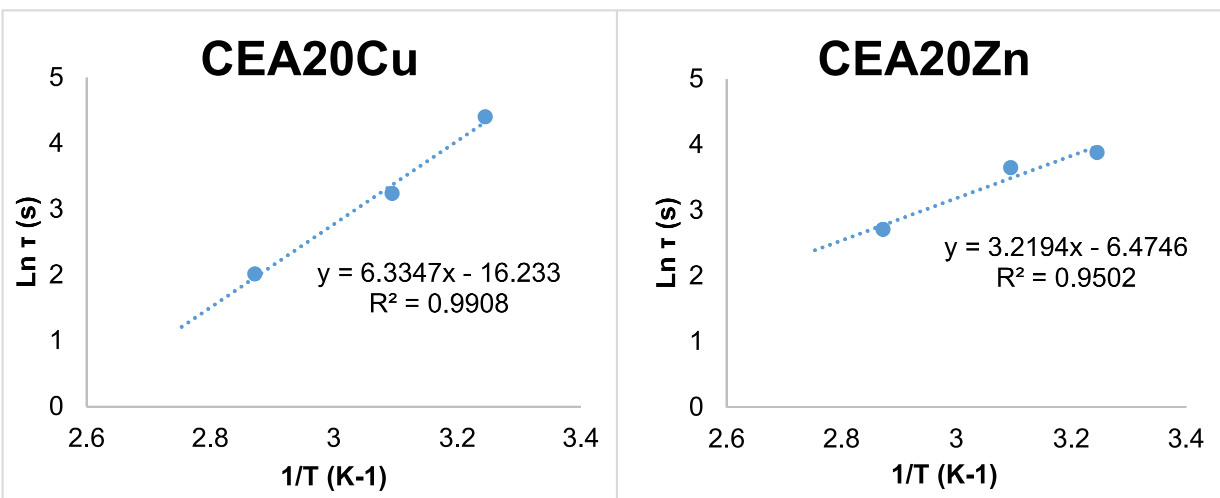


Figure S9. Stress relaxation time (τ) vs. 1/T for (a) **CEA20Cu** and (b) **CEA20Zn**. Follows an Arrhenius law:

$$\text{Arrhenius law:} \quad \frac{1}{\tau} = \frac{1}{\tau_o}\, e^{-\frac{Ea}{RT}}$$

Where τ_o is constant (s), E_a is activation energy (J/mol), R is the ideal gas constant (8.314472 J/mol*K), and T is temperature (K). Activation energy is determined from the slope E_a/R.[1] For copper this is 52.7 kJ/mol, while for zinc this is 26.8 kJ/mol.

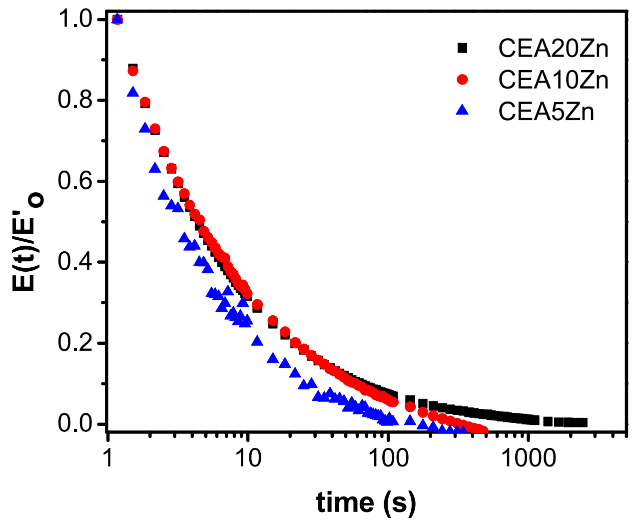


Figure S10. Stress relaxation of zinc-coordinated polymers at 75 ºC.

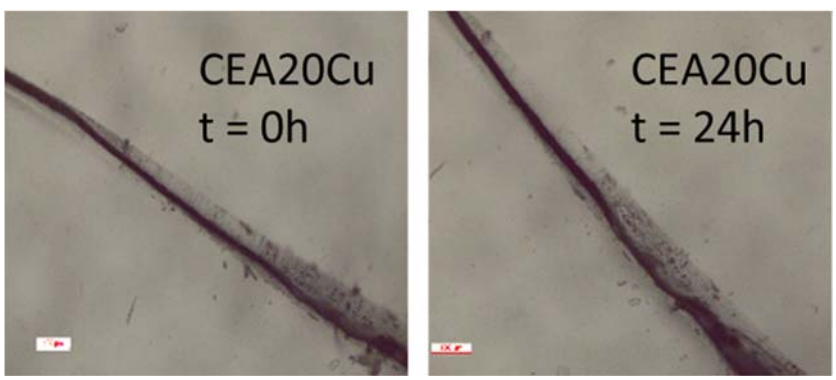


Figure S11. Optical microscopy images of copolymer films of **CEA20Cu**: (left) as-cut film; (right) self-healed film.

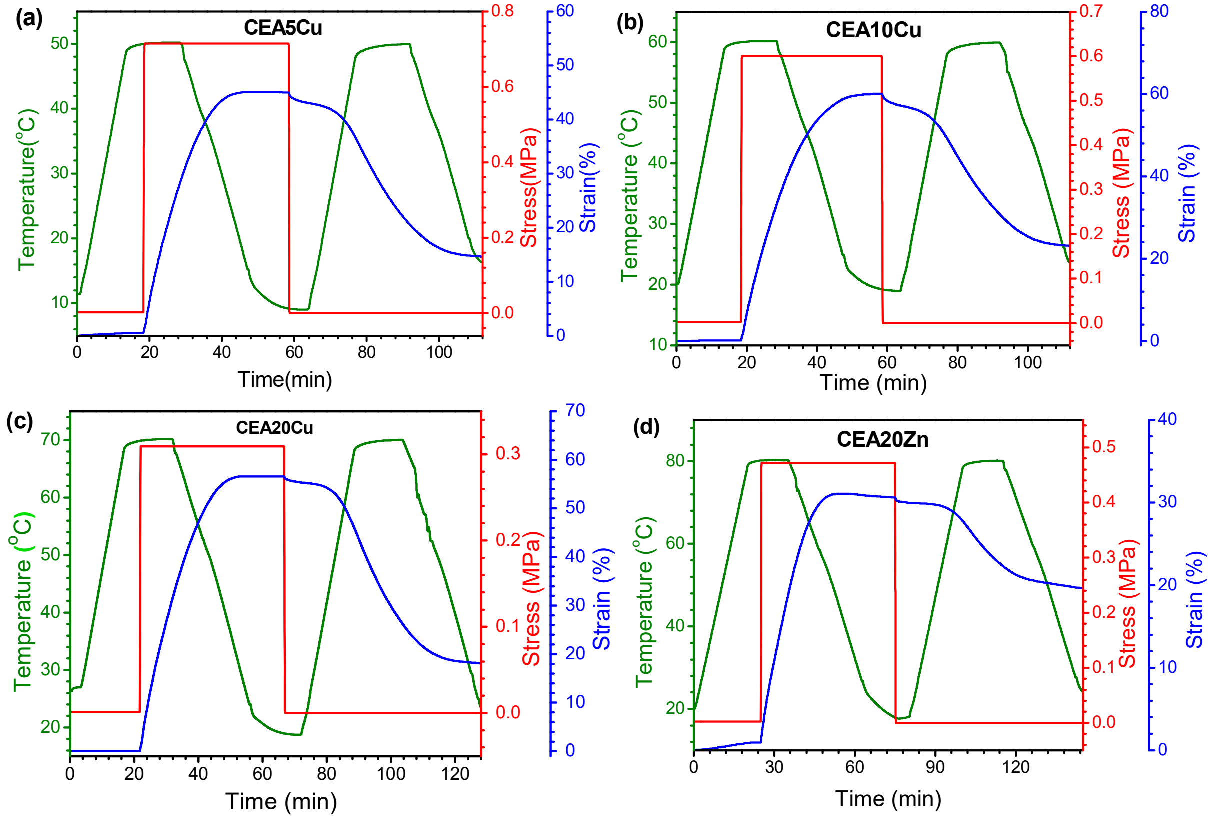


Figure S12. Dual (stress- and temperature-) programmed shape memory testing of (a) **CEA5Cu**, (b) **CEA10Cu**, (c) **CEA20Cu**, and (d) **CEA20Zn**.

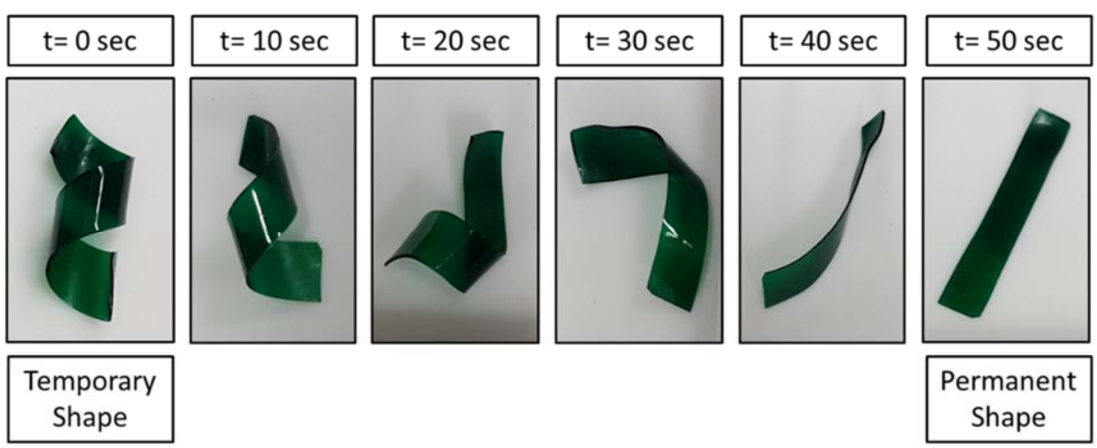


Figure S13. Time-dependent thermo-responsive shape memory testing of **CEA20Cu**.

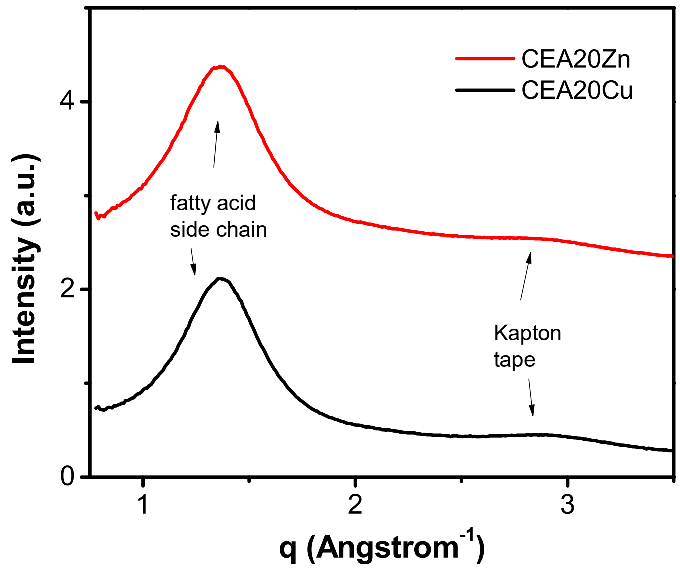


Figure S14. WAXS profiles of metal-ligand coordinated copolymers: **CEA20Cu** and **CEA20Zn** at 25 ºC.

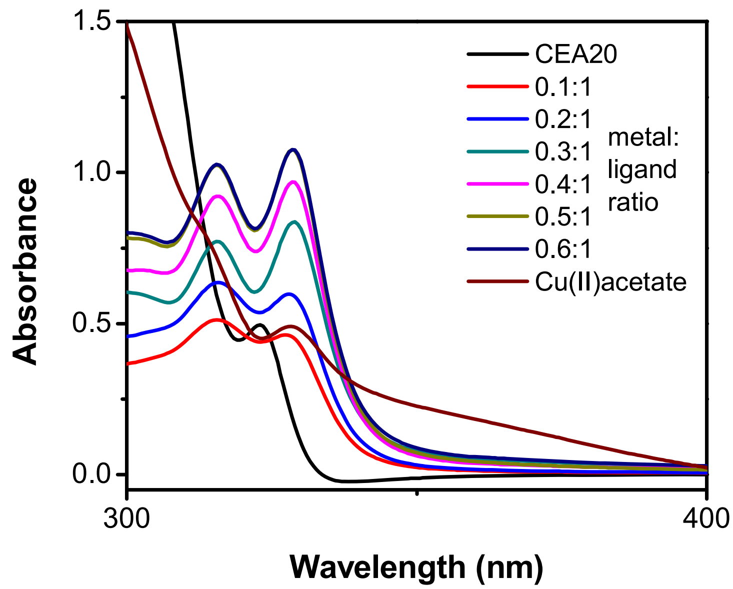


Figure S15. UV-VIS spectra of copolymer **CEA20Cu** being titrated with metal salt.

References

1. Zhao, S.; Abu-Omar, M. M. Recyclable and Malleable Epoxy Thermoset Bearing Aromatic Imine Bonds. *Macromolecules* **2018,** 51, 9816-9824.