

Title: Toward pluripotent materials through tempering of dynamic covalent polymer networks

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Abstract:

Pluripotency, defined as a system not fixed as to its developmental potentialities, is typically associated with stem cells. Reported herein is an approach to achieve pluripotent polymers by designing dynamic covalent networks that can be tempered, akin to the process utilized in metallurgy. The materials developed in this work can be differentiated into a range of mechanical properties, from hard and brittle to soft and extensible, by altering the tempering temperature of a single pluripotent feedstock. Key to this behavior is the use of labile, dynamic thia-Michael bonds, whose extent of bonding can be thermally modulated and retained. These pluripotent materials can be (re)processed via tempering, exhibit adaptive shape memory behavior, and be thermally patterned to spatially control mechanical properties.

One-Sentence Summary:

Pluripotent polymers have been designed that can be differentiated through tempering into a range of materials with distinct mechanical properties.

Main Text:

Limited access to materials in resource-scarce areas (at sea, in space, on the battlefield) presents challenges, as operational tasks often require a wide range of materials with distinct mechanical properties for specific applications. Techniques, such as additive manufacturing, have attempted to satisfy these constraints by providing an incredible enhancement in form factors that allow for a single material to be used in various applications or locations (1–4). Still, the bulk material properties remain primarily limited by the resin itself. Developing a single feedstock that can be differentiated into materials with distinct, interconvertible mechanical properties would prove transformative for applications in these challenging environments.

Nature has developed her answer to this challenge with pluripotent stem cells that readily differentiate into a host of different cell types with varying properties and purposes in the human

body (5–7). Envisioning pluripotency in a synthetic material, which would have the ability to (inter)convert its properties with environmental or processing cues, could be attained by a system with the following criteria: 1) reconfigurable bonds that allow for bulk property manipulation, 2) sensitivity to tunable stimuli conditions to access a range of material properties, and 3) an ability to ‘lock in’ properties within the operational temperature window of interest (**Figure 1a**). Metallurgists use tempering of metals, defined as the isothermal heating of a material below a critical point (e.g., melting point) before rapidly quenching (8,9), to expand the application of steel from knives to structural beams. Applying the thermal processing technique of tempering to polymeric materials would open the door to achieving pluripotent, synthetic materials and provide a new paradigm for polymeric material design.

Following criterion 1, a wide range of reconfigurable/dynamic bonds are accessible, which include supramolecular interactions (10,11) and dynamic covalent bonds (12-16). Within polymeric materials, a wide array of dynamic chemistries have been exploited to alter the adaptive properties of the material and allow them to be used in a breadth of applications (17-32). The reversibility of dynamic bonds under different environmental stimuli satisfies the first of the above criteria to access pluripotency. Still, particular emphasis must be placed on materials that can be reconfigured at low processing temperatures ($< 100\text{ }^{\circ}\text{C}$) to achieve practicality in resource-scarce areas. Previous work has shown that polymer networks that contain room temperature dynamic thia-Michael (tM) bonds, obtained by the catalyst-free reaction of a ditopic benzalcyanoacetate (BCA), end-capped triethylene glycol Michael acceptor with a tetrathiol crosslinker (**Figure 1b**), can be used to access films with robust mechanical properties on account of a dynamic reaction-induced phase separation (DRIPS) process (30,31). The equilibrium constant (K_{eq}) of the thiol addition to the BCA Michael acceptor can be controlled by the electronic nature of the BCA acceptor, which ranges from 10 M^{-1} for electron-donating substituents (-OMe) to ca. 500 M^{-1} for electron-withdrawing substituents (-NO₂). Given the temperature sensitivity of these relatively weak dynamic bonds, it was hypothesized that such networks may have the potential to be designed into temperable materials that would exhibit pluripotency.

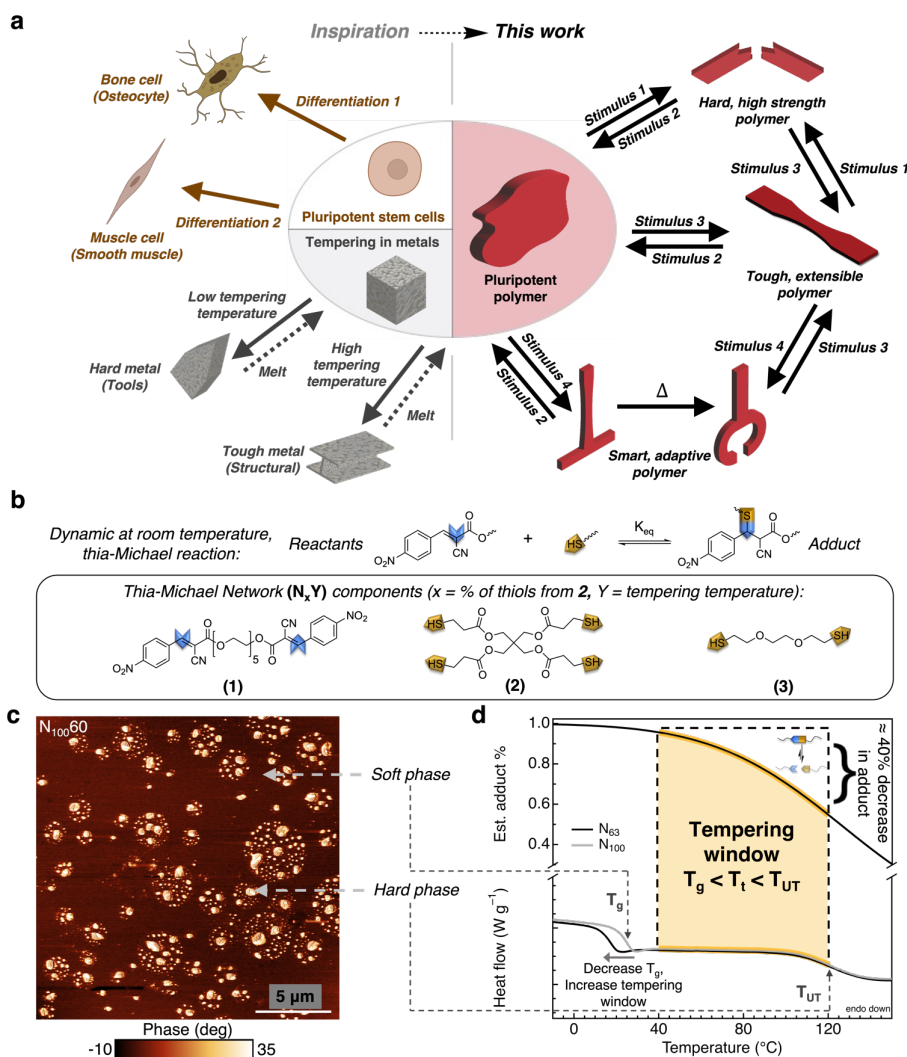


Fig. 1 | Pluripotent polymer design. **a**, Inspiration for the design of a pluripotent polymer comes from stem cells and the tempering of metals. **b**, The dynamic thia-Michael bond consisting of a benzalcyanoacetate-based Michael acceptor and thiol component, with the chemical structures of the thia-Michael network (N) components: a ditopic nitro-bearing Michael acceptor (1) (Figure S1 for NMR characterization), tetrathiol crosslinker (2), and dithiol chain extender (3) with the nomenclature of N_xY used herein, with x being the percentage of thiols from 2 and Y being the tempering temperature. **c**, Representative AFM phase micrograph of N₁₀₀60 showing a dynamic reaction induced phase separation (DRIPS) microstructure with globular hard domains (light) interspersed in a soft matrix (dark). **d**, Defining the tempering temperature window (T_t) for these dynamic networks. Top half shows the estimated adduct percentage as calculated from solution-state ¹H NMR experiments (Figure S3–4) highlighting a ca. 40% decrease in adduct percentage from 45 to 120 °C (the tempering window). Bottom half is a DSC thermogram of N₁₀₀ (grey curve) that shows the presence of two thermal transitions, a lower glass transition temperature (T_g) and an upper transition temperature (T_{UT}) corresponding to the hard phase of the DRIPS

microstructure. The tempering window is defined as the temperature range between T_g and T_{UT} . The tempering window can be expanded by decreasing the T_g through introduction of the dithiol **3** during the network synthesis, e.g., as shown for the N_{63} film (black curve).

Designing a pluripotent polymer

A ditopic Michael acceptor (**1**) with a pentaethylene glycol core was targeted to enhance the flexibility of the network, while the higher equilibrium constant *p*-nitro-BCA Michael acceptor was chosen to aid in maintaining mechanical robustness at elevated temperatures. Crosslinking **1** with pentaerythritol tetrakis(3-mercaptopropionate) (**2**) yielded the tM network (N_xY , where x is the percentage of thiols from **2** and Y is the tempering temperature) films. **Figure 1c** shows a representative atomic force microscopy (AFM) image of an $N_{100}60$ film (where 60 is the temperature (°C) at which the film was isothermally heated (tempered) for at least one day before quenching with liquid nitrogen) in which the characteristic DRIPS microstructure of a continuous soft matrix with interspersed, globular hard domains can be observed. Differential scanning calorimetry (DSC) of N_{100} shows two thermal transitions (**Figure 1d**, grey line), a low glass transition temperature (T_g), and an upper thermal transition (T_{UT}). With the goal of accessing pluripotent materials, it is vital to have a tempering window that allows for tuning the number of dynamic bonds or crosslinks formed in the film so that the mechanical properties can be influenced. For the BCA tM bond, estimations from solution-state 1H nuclear magnetic resonance (NMR) spectroscopy experiments suggest a $\approx 40\%$ decrease in the tM adducts (**Figure 1b**) formed upon raising the temperature between the T_g and T_{UT} of this thia-Michael network (**Figure 1d**), with significantly lower activation energy (E_a) for the bond formation reaction (45 kJ mol^{-1}) than the bond cleavage reaction (122 kJ mol^{-1}) (**Figure S3**). It was hypothesized that after quenching to ambient temperatures, unwanted bond formation (bimolecular coupling) would be impeded by the increased viscosity, while unwanted bond breakage would be hindered by its relatively higher E_a , thereby stabilizing the tempered properties. This combination of 1) the presence of reconfigurable bonds, 2) the thermal sensitivity of dynamic bonds within the tempering window, and 3) the stabilization of the tempered properties at room temperature presents these thia-Michael networks as a prototypical system to explore pluripotency in a synthetic material.

To lower the T_g and expand the tempering window, a series of materials with varying network compositions were prepared in which **1** was combined with a mixture of tetrathiol, **2**, and dithiol, 2,2'-(ethylenedioxy)diethanethiol (**3**) (in ratios of 100:0, 75:25, 63:37, 50:50, 25:75, and 0:100, respectively, based on the percentage of thiols from that particular component) at a 1:1 stoichiometric ratio of [double bond] : [thiol]. Thermal gravimetric analysis (TGA) demonstrated that all films were dry and had decomposition temperatures ranging from 215–262 °C (**Figure S5**). DSC and shear rheometry (**Figures S6–S7**) confirmed two characteristic thermal transitions at all network compositions and identified an increase in the width of the tempering window (**Figure 1d**, grey curve) with decreasing average functionality of the thiol components (f_{thiol} , ranging from f_{thiol} of 4 for N_{100} to f_{thiol} of 2 for N_0). Each N_x was tempered using a tempering temperature (T_t) within the tempering window, whereafter the thermomechanical properties were measured using shear rheometry (**Figure 2a & S6**). Interestingly, for films with at least 50 % of the tetrathiol **2**, both T_g and the plateau storage modulus (G') increased with decreasing T_t , with the difference in G' retained throughout the tempering window. Within a single network composition, an increase of 3.5x in G' and 5 °C in T_g was realized by changing T_t from 95 to 65 °C. By manipulating the formulation with f_{thiol} , a two-order-of-magnitude change in the thermomechanical properties of the N_xY films can be accessed. On account of its room temperature operational window and large

variation in G' , N_{63} was chosen for further investigation of the underlying mechanism of tempering in the N_xY films.

The changes in the thermomechanical properties (such as moduli and T_g) could be governed by the underlying DRIPS microstructure, changes in the amount of dynamic bond formed within the network, or a combination of both. While AFM confirmed a decrease in the hard phase with decreasing f_{thiol} (**Figure S8**), no clear trends were observed when a film (of set f_{thiol}) was tempered at different T_t (see *Methods* for a detailed explanation of tempering for AFM samples), suggesting that the amount of hard phase is not the primary mechanism that drives the significant changes in mechanical properties with tempering conditions. This is in stark contrast to previous results where cooling rate through the T_{UT} resulted in changes to nucleation and growth of the hard phase that led to differences in final mechanical properties (30). Raman spectroscopy can assess the amount of Michael adduct (2257 cm^{-1}) and unreacted BCA moieties (2227 cm^{-1}) present in the films following the tempering process by monitoring the shift in the nitrile stretch upon bonding. **Figure 2b** shows part of the room temperature Raman spectra of a N_{63} film after it was tempered at three different T_t . The data clearly shows that the amount of adduct in the film is highly dependent on the T_t . Higher T_t results in reduced adduct and, therefore, a network with lower crosslinking density (**Figure S9**). Such coupling of the T_t and crosslinking density correlates well with decreasing stiffness in the dynamic network films tempered at high T_t . To further support this hypothesis, rheo-Raman experiments (33) were carried out to probe the time-temperature dependence of the film's thia-Michael equilibrium and mechanical properties during the tempering process. **Figure 2c** overlays the storage moduli and fraction of adduct as a function of time and T_t for a representative material N_{75} , (see **Figure S10** for N_{63} and H_{100} data). As illustrated, the growth in G' of N_{75} at $95\text{ }^\circ\text{C}$ (**Figure 2c**, red curve) was concomitant with an increase in the amount of thia-Michael adducts formed. When the T_t was then lowered to $75\text{ }^\circ\text{C}$, there was a distinct rise in both G' and the amount of thia-Michael adduct in the film. If a film is initially heated at $75\text{ }^\circ\text{C}$, the equilibrium modulus and tM adduct fraction is the same as the film tempered at $95\text{ }^\circ\text{C}$ followed by $75\text{ }^\circ\text{C}$ (**Figure 2c**, blue curve). Heating the $75\text{ }^\circ\text{C}$ tempered film to $95\text{ }^\circ\text{C}$ results in a gradual decrease in both modulus and tM adduct formed. As clearly shown by the rheo-Raman data, the combination of tempering with these tM dynamic bonds leads to materials with reconfigurable crosslinking density and controllable mechanical properties, independent of the tempering pathway. Time-resolved shear rheometry was used to monitor the evolution of G' with time for the films with varying network compositions (**Figure S11**), further confirming that tempering at lower T_t resulted in a higher, equilibrated G' plateau, driven by an increase in adduct formation. After normalization of G' , the time required to reach the halfway point ($t_{0.5}$, **Figure S12**) in the growth curves could be collapsed into a simple Arrhenius relationship (**Figure 2d**), suggesting that growth of G' followed a similar evolution process, independent of network composition, with an activation energy of $84 \pm 7\text{ kJ mol}^{-1}$ for the N_xY films. As changes in adduct fraction and segmental mobility primarily drive the changes in mechanical properties, it is unsurprising that all materials follow a similar G' evolution process. However, it is worth pointing out that this measured activation energy does not correlate precisely to that of the dynamic bond itself in N_xY , either in bond formation (45 kJ mol^{-1}) or bond cleavage (ca. 122 kJ mol^{-1}) (**Figure S3**). The tempering process was further generalized to thia-Michael networks containing hydrogen-substituted Michael acceptors (H_xY) (**Figure S13**), to ensure that tempering was not substituent-dependent and yielded a similar E_a of $89 \pm 2\text{ kJ mol}^{-1}$.

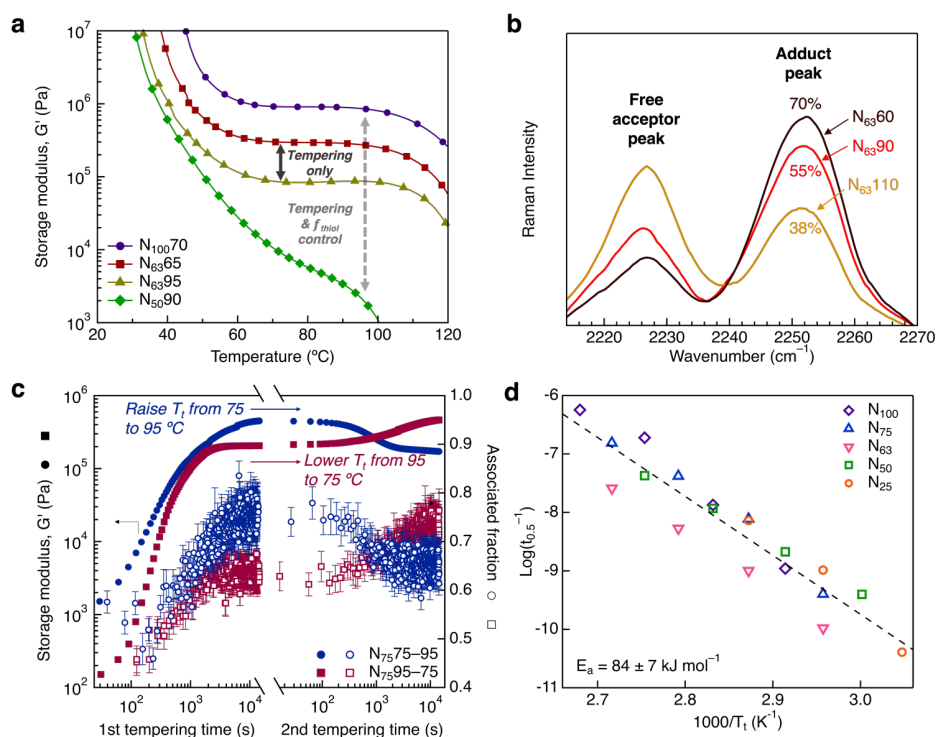


Fig. 2 | Tempering process in N_xY . **a**, Temperature-dependent storage modulus, G' for $N_{100}70$, $N_{63}65$, $N_{63}95$ and $N_{50}90$ showing that both f_{thiol} and T_t impact G' . For the $N_{63}Y$ film there is a 3.5-fold increase in G' (at $80^{\circ}C$) from $N_{63}95$ (84 kPa) to $N_{63}65$ (295 kPa) with the lower T_t . There is also a minor shift in T_g , with $N_{63}95$ ($35^{\circ}C$) having a $4^{\circ}C$ lower T_g than $N_{63}65$ ($39^{\circ}C$). **b**, Room temperature partial Raman spectra of N_{63} films tempered at various T_t with the nitrile stretch of the free acceptor peak at 2227 cm^{-1} and the nitrile stretch of the adduct peak at 2253 cm^{-1} . **c**, Two-step rheo-Raman experiment where films of $N_{75}95-75$ (red squares) and $N_{63}75-95$ (blue circles) were first isothermally held at $145^{\circ}C$ for 15 min to erase history before rapid cooling to the specified temperature. Upon reaching T_t , the evolution of G' and rise in associated fraction was monitored, demonstrating that increasing associated fraction of thia-Michael bonds (determined by Raman data) leads to increased G' , and that the process is reversible. Error bars in the associated fraction represent the standard error from the uncertainty in fitting of the two nitrile stretch peaks, see **Figure S10** for a more detailed discussion. **d**, Arrhenius plot of $t_{0.5}$ values from normalized growth curves against T_t for all N_xY films with an activation energy, E_a , of $84 \pm 7\text{ kJ mol}^{-1}$, which is attributed to a combination of dynamic bond exchange and segmental mobility of the network.

Re-forging plastic

One of the key advantages of dynamic polymer networks over traditional covalent networks is their ability to be reprocessed and recover their original mechanical properties (34). While reprocessing does allow recovery of a single mechanical performance, tempering has the added benefit of being able to (re)define the material properties across a range of common polymer mechanical properties through control of Young's modulus, E , and strain at break, ϵ_b . (Re)programming dogbone specimens of N_{63} at various T_t illustrates the tailorability and reliability of this method to achieve different, repeatable mechanical performance from a single material after tempering for less than 24 h with a simple quenching step on a cold, metal block (**Figure 3a**, **Figure S14**, **Table 1**, see *Methods*). While all samples were tempered under an inert atmosphere

to prevent unwanted side reactions, there was no evidence of disulfide formation when tempering under open, ambient conditions for nine days, suggesting that tempering could be achieved without inert conditions in future studies (**Figure S15**). Programming with T_t 's of 70 °C, 90 °C, 100 °C, 110 °C, and 120 °C resulted in a spectrum of room temperature properties, ranging from brittle thermoset ($E = 1520 \pm 27$ MPa, $\epsilon_b = 2 \pm 0.4$ %) (**Figure 3b**, red curve with squares), to tough thermoplastic ($E = 670 \pm 84$ MPa, $\epsilon_b = 130 \pm 30$ %) (**Figure 3b**, blue curve with rhombi), and soft, extensible polymer ($E = 210 \pm 20$ MPa, $\epsilon_b = 250 \pm 14$ %) (**Figure 3b**, green curve with pentagons) respectively. Further investigation also demonstrated that tempering could control E in lower T_g (≈ 0 °C) materials containing a mixture of N/H_xY (**Figure S16**), albeit to a lower extent. Beyond programming across this range of properties at room temperature, **Figure 3c** further illustrates the ability to (re)cycle between **N₆₃60** ($E = 1470 \pm 70$ MPa on first cycle to $E = 1660 \pm 50$ MPa after six reprocessing cycles) and **N₆₃110** ($E = 220 \pm 20$ MPa on first cycle to $E = 390 \pm 64$ MPa after four reprocessing cycles), with the Young's Modulus being generally within error during the processing cycles (**Figure S17**). Furthermore, mechanical aging studies (see *Methods*) confirmed that the moduli could be effectively retained for over a month in both **N₆₃60** (approximate loss of 1.1 MPa day⁻¹) and **N₆₃110** (approximate gain of 2.2 MPa day⁻¹) (**Figure S18**). In brief, these experiments highlight the ability to temper-in mechanical properties at a specific operating temperature (room temperature for this study), easily switch between these properties without any chemical modification, and retain those tempered-in properties for over a month.

To expand the type of behavior achievable in a material, metallurgists have commonly used differential tempering to spatially control mechanical properties (e.g., in swords with sharp edges and toughened spines). The ability to spatially control a film's mechanical properties has been an emergent area of study in polymer design and synthesis (35,36). Within additive manufacturing (1,37,38) this technique is used to access unique multimaterial designs previously limited to biological materials (3). While advanced strategies have addressed early concerns about poor interfaces between materials (2), limitations still exist in recycling or reprogramming the materials after they are formed. The tempering of dynamic covalent networks has the potential to enable access to interesting multimaterial or mechanically graded designs in recyclable N_xY materials. As a first demonstration, a **N₆₃60** film was prepared to create a hard, brittle plastic with high strength and low toughness. Next, the film was placed onto a heated block composed of half aluminum and half polyimide fiber insulation (**Figure 3d**, see *Methods*) to locally temper only the film exposed to the aluminum substrate, as confirmed by the IR camera (**Figure S19**). After tempering, the film showed preferential deformation in the "softer" region processed at a higher T_t (**Figure 3e**, **Movie S1**). Additionally, the differentially tempered bar was loaded into a mechanical load frame at room temperature and strained to failure. On application of strain, the extensible, high T_t network preferentially deformed under the low applied stresses, while the other half had no observable deformation occur (**Figure 3f**, **Movie S2**). For increased complexity, an ABA system was prepared via a low-high-low spatial tempering process (**Figure 3g**, see *Methods*), as confirmed by the IR camera image (**Figure S20**), imparting a hinge-like response under bending (**Movie S3**) in contrast to the brittle failure of the un-tempered material (**Movie S4**). The preferential deformation was further illustrated on the hinge-like sample using a mechanical load frame and nominal strain rate of 25 % min⁻¹. As expected, the material deformed in the region tempered at higher T_t , leading to a local ductile failure away from the interface (**Figure 3h**, **Movie S5**). A more complex 'S' shape was also differentially tempered in an **N₆₃** film and could be visualized upon strain (**Figure S21**). From these demonstrations, differential tempering of a single film can be used to access spatially controlled mechanical properties without the need for complex welding or adhesion of disparate materials, and could be further expanded through more complex

patterning techniques in future studies (39). Interestingly, the shape of the AB system could be recovered through light heating (rubbing with fingertips), which motivated further investigation through cyclic loading/unloading experiments (see *Methods*). Through these experiments, the yielding behavior could be recovered through both time and temperature, suggesting an ability to minimize material performance degradation due to plastic deformation (**Figure S22**).

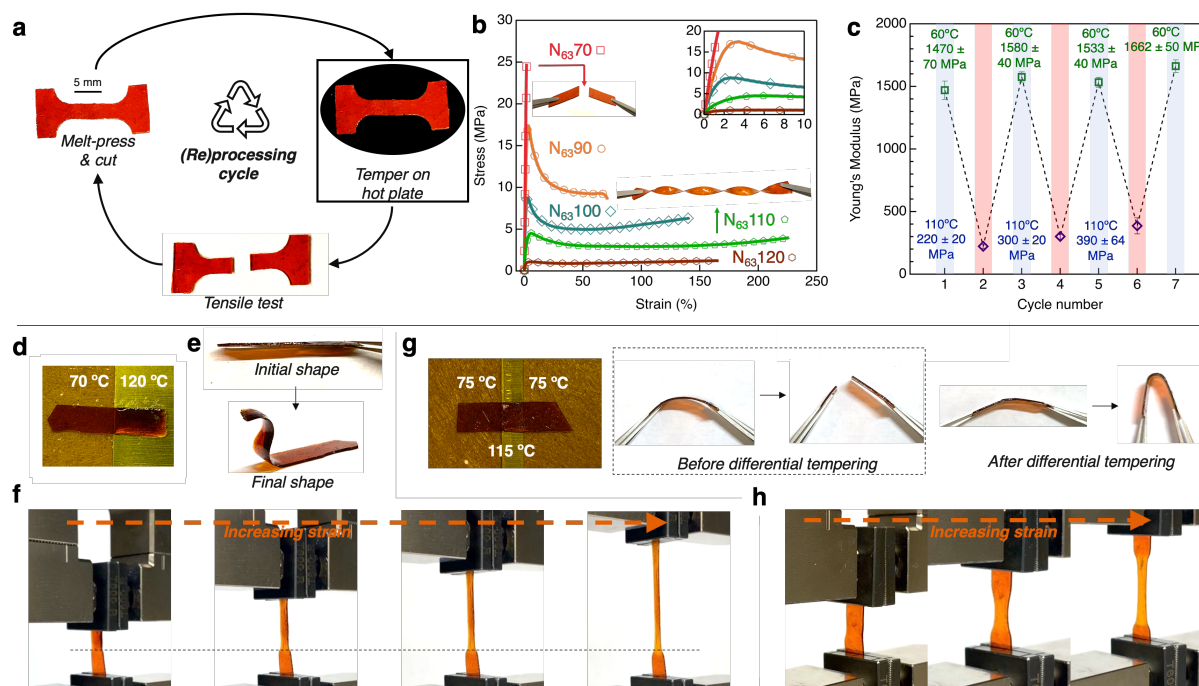


Fig. 3 | Reprocessability of N_xY . **a**, Schematic of the (re)processing cycle for a dogbone specimen of a $N_{63}Y$. **b**, Representative stress-strain curves (at room temperature) of $N_{63}70$ ($N = 3$, red square), $N_{63}90$ ($N = 9$, orange circle), $N_{63}100$ ($N = 5$, blue rhombus), $N_{63}110$ ($N = 4$, green pentagon), and $N_{63}120$ ($N = 6$, brown hexagon) films with a strain rate of $25 \% \text{ min}^{-1}$. Dogbone specimens were tempered for less than 24 h under inert atmosphere and quenched on a cold, metal block before testing (see *Methods*). **c**, Young's modulus versus recycling experiments showing the ability of the film to be reprogrammed between $N_{63}60$ (with $N = 5$ (cycle 1), 6 (cycle 3), 4 (cycle 5), and 5 (cycle 7)) and $N_{63}110$ (with $N = 7$ (cycle 2), 6 (cycle 4), and 3 (cycle 6)) (full stress-strain curves shown in **Figure S17**). **d**, Picture of AB differential tempering stage with N_{63} film. **e**, Image of N_{63} film after differential tempering, before and after applying strain. **f**, Images of AB differentially tempered N_{63} film during tensile testing with strain visible in the soft, extensible section of the film but not in the rigid section. **g**, Picture of ABA differential tempering stage with N_{63} film. **h**, Images of ABA differentially tempered film progressing from bar specimen before application of strain to dogbone-like specimen after the strain was applied.

Pluripotency in the material's stimuli-responsive properties

The ability to control mechanical properties is an important aspect of pluripotency; however, expanding this control to tailoring the material's stimuli-responsive nature gives access to an even broader range of applications. As related DRIPS networks have been shown to exhibit shape memory behavior (30), it was of interest to see if tempering could be used to alter this responsive property. Tempering N_{63} at 110 °C ($N_{63}110$) showed an ideal fixing ratio (maximum strain, ϵ_m , divided by unloading strain, ϵ_u) of 1.0, and excellent shape recovery (94 %) after reaching an ϵ_m

of 100 % upon loading of 0.16 MPa (**Figure 4a**, see *Methods*). In contrast, **N₆₃** tempered at 60 °C (**N₆₃60**) exhibited a 30 % reduction in ϵ_m relative to **N₆₃110**, despite an almost doubling of the force (0.30 MPa) applied during the shape-fixing process (**Figure 4b**). Importantly, at the service temperatures of 30 and 60 °C, both **N₆₃60** (0.02 % h⁻¹ and 0.11 % h⁻¹, respectively) and **N₆₃110** (0.10 % h⁻¹ and 0.52 % h⁻¹, respectively) showed minimal creep (**Figure S23**). To further demonstrate the contrast in actuation between materials tempered at different T_t , **N₆₃110** and **N₆₃60** were extended to 300 % and 220% strain, respectively (see *Methods*). Upon heating, **N₆₃110** could linearly lift a 20 g weight (work = 60 kJ m⁻³, **Figure 4b**, **S24**, **Movie S6**), while the **N₆₃60** was able to linearly lift a 50 g weight over a smaller displacement (work = 110 kJ m⁻³, **Figure 4d**, **Movie S7**).

As a final proof-of-concept of these materials' shape memory and linear actuation capabilities, a 'T' shape was cut from a film of **N₆₃** and folded into a claw-like shape around a metal I-bar before being tempered at 100 °C for 1 h (**Figure S25**). After deformation, the claw was fixed to tensile grips and slowly lowered to a heated metal I-bar, where the 'fingers' of the claw grasped the I-bar (**Figure 4e**, **Movie S8**). Upon heating, the arm linearly actuated and lifted the I-bar from the ground. As the tensile grips were raised, the claw maintained control of the I-bar. Thus, materials can be made to not only have passive properties of interest through control of pluripotency, but can also be rationally designed to create adaptive, multifunctional materials.

In summary, a strategy to design pluripotent materials that can be easily programmed and reprocessed to exhibit a wide range of materials properties. These materials, which exploit a synergistic relationship between dynamic networks and phase separation, show a range of room temperature properties from brittle and glassy to soft and extensible with a simple change in tempering without requiring any external chemical modification or additives. Careful control of material form factor and programming enables further control of material response that could be used in soft robotics applications. Pluripotent materials, especially with further optimization, can be a valuable tool in breaking the standard materials' paradigm of one material for one function and could allow for versatile, closed-loop materials that can be (re)programmed and recycled as required by the user. Expanding the guiding principles for pluripotent materials, presented herein, to different matrices, polymer backbones, composites, or using mechanical energy to 'train' materials (40,41) could provide a rich platform for future materials design in areas of resource scarcity.

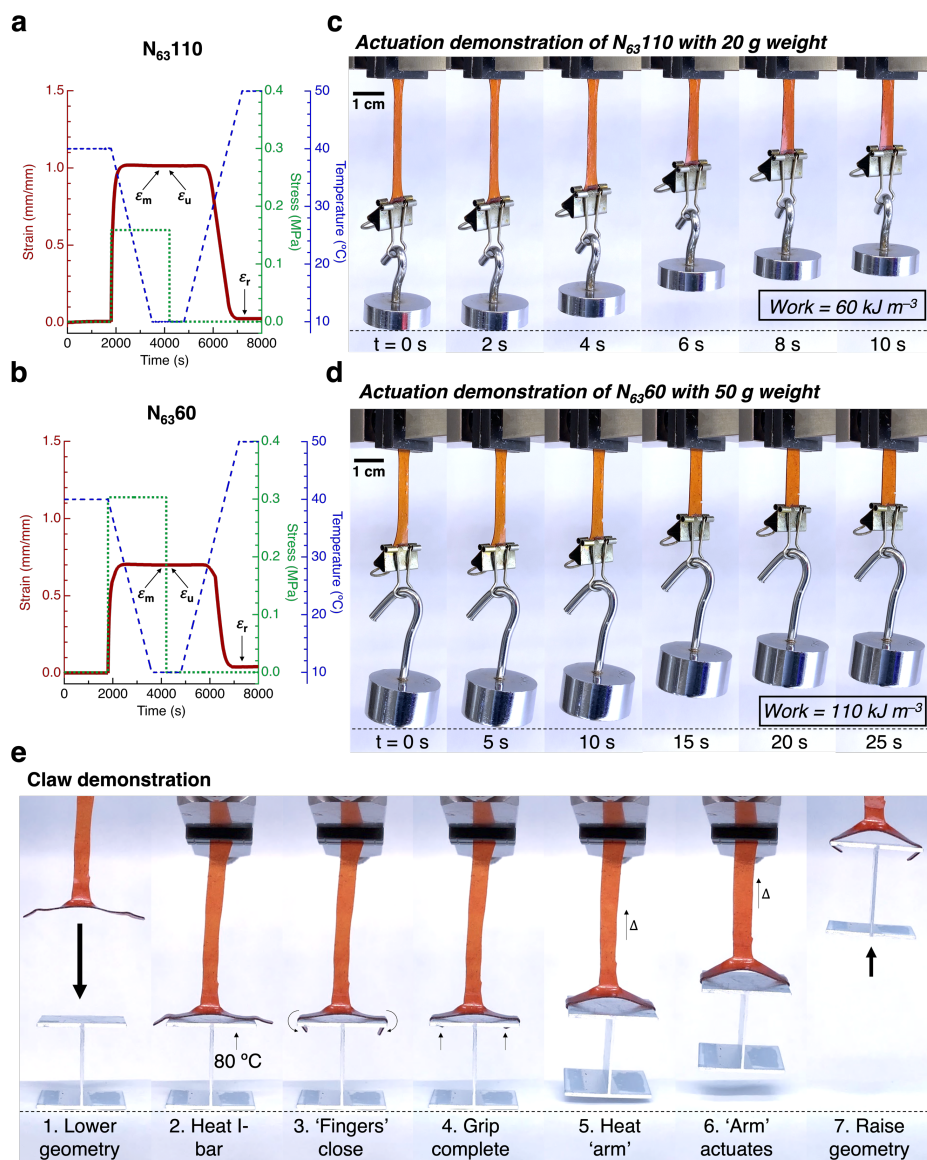


Fig. 4 | Adaptive, pluripotent materials. Thermomechanical shape memory curves for **a**, N_{63110} , and **b**, N_{6360} obtained using Dynamic Mechanical Analysis (DMA) experiments. **c**, Shape memory actuation demonstration for N_{63110} with a 20 g weight attached. **d**, Shape memory actuation demonstration for N_{6360} with a 50 g weight attached. **e**, Images of the shape memory 'claw' demonstration for N_{63100} where the (1) the geometry is lowered until the claw contacts the I-bar, (2) the fingers of the claw then contact the heated I-bar ($\approx 80 \text{ }^\circ\text{C}$), which (3) stimulates the fingers of the claw to spontaneously close around the I-bar. Once the (4) grip of the fingers is complete, (5) the 'arm' of the claw is heated ($\approx 60 \text{ }^\circ\text{C}$) to (6) initiate actuation of the arm, which removes the I-bar from contacting the ground. After this, (7) the geometry is raised to demonstrate successful grip of the I-bar by the claw (**Figure S25**, **Movie S8**).

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Acknowledgments:

25 Parts of this work were carried out at the Soft Matter Characterization Facility and the Materials Research Science and Engineering Center (MRSEC NSF DMR1420709) at the University of Chicago. The authors thank Alex Crolais for high-resolution mass spectrometry (HRMS) of **1**. The authors thank Dr. Philip Griffin, Dr. Justin Jureller, and Dr. Tyler Jorgenson for insightful advice on instrumentation. The authors thank Samir Hossainy for his help designing **Figure 1** using BioRender.com. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the official policies, either expressed or implied, of the Army Research Laboratory, or the U.S. Government. The U.S. Government is authorized to reproduce and distribute reprints for Government purposes notwithstanding any copyright notation herein. Certain commercial equipment, instruments, or materials are identified in this paper in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.

Funding:

This work was supported by National Science Foundation under award number DMR-2104694 and by the University of Chicago Materials Research Science and Engineering Center, which is funded by the National Science Foundation under award number DMR-2011854. N.R.B was supported by a NASA Space Technology Graduate Research Opportunity at the NASA Glenn Research Center. J.M.D. was sponsored by the Army Research Laboratory under Cooperative Agreement Number W911NF-20-2-0044 and NDD was supported in part by the NIST Center for Hierarchical Materials Design, 60NANB15D077.

National Science Foundation Grant, DMR-2104694 (SJR)

NASA Space Technology Graduate Research Opportunity, 80NSSC21K1296 (NRB)

Army Research Laboratory Cooperative Agreement, W911NF-20-2-0044 (JMD, JLL)

NIST Center for Hierarchical Materials Design, 60NANB15D077 (NDD)

National Science Foundation Grant, DMR-2011854 (CAL)

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Competing interests:

Authors declare they have no competing interests.

Data and materials availability: All data are available in the main text or the supplementary materials.

Supplementary Materials

Materials and Methods

Supplementary Text

Figs. S1 to S25

Table 1

References (42–45)

Movies S1 to S9