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COMMENTARY

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Redefining hydrogel mechanics through reversible supramolecular chains

Boya Song,[1,3,4,5] Jifei Zhang,[2,5] Sanwei Hao,[1,3,4,*] Changyou Shao,[2,*] and Runcang Sun[2]

[1]School of Materials Science and Engineering, Shandong University of Technology, Zibo 255000, China

[2]Liaoning Key Laboratory of Lignocellulose Chemistry and BioMaterials, Liaoning Collaborative Innovation Center for Lignocellulosic Biorefinery, College of Light Industry and Chemical Engineering, Dalian Polytechnic University, Dalian 116034, China

[3]Shandong Key Laboratory of Functional-Structural Integrated Ceramics, Shandong University of Technology, Zibo 255000, China

[4]Discipline and Technology Center for High Temperature Functional Ceramic, Shandong University of Technology, Zibo 255000, China

[5]These authors contributed equally

*Correspondence: shaocy@dlpu.edu.cn (C.S.); haosanwei@sdut.edu.cn (S.H.)

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Hydrogels represent a cornerstone of soft matter science, bridging the gap between biological tissues and synthetic polymers.[1,2] Their high-water content and mechanical compliance render them highly suitable for biomedical and soft robotic applications. Nevertheless, their practical deployment has been consistently hampered by limited extensibility. Conventional hydrogels typically withstand only limited uniaxial stretching and are highly susceptible to fracture under large biaxial deformations, primarily due to network heterogeneity and the finite extensibility of polymer chains.[3] Although strategies such as slide-ring cross-linking and double-network architectures have enhanced their toughness and stretchability, the maximum reversible biaxial strain achieved to date remains below 2500%. This limitation severely hinders their application in pneumatic actuators, artificial muscles, and other advanced soft robotic systems.

In this context, Chen and colleagues[4] report a hyperelastic hydrogel capable of achieving an ultra-large reversible biaxial strain exceeding 10,000%. The exceptional hyperelasticity of this material originates from reversible supramolecular thermodynamic mechanisms (Figure 1A). Under water-deficient conditions, hydrophobic interactions among alkyl side chains trigger local chain collapse into subnanometric clusters, while ionic groups maintain hydration to form stable connecting segments. Upon deformation, mechanical input drives the unfolding of these clusters, thereby releasing concealed segments of the polymer chains and significantly expanding the available conformational space. Once the strain is removed, entropic forces together with hydration-mediated stabilization restore the clusters, returning the chains to a low-energy, bead-like configuration. This reversible process establishes an elastic mechanism fundamentally distinct from covalent bond rupture or irreversible plastic flow. Such an entropy-dominated and energy-efficient mechanism constitutes the underlying basis for the material's ultrafast recovery kinetics.

Mechanical characterization reveals the exceptional performance of the pearl-necklace chain (PNC)-based hydrogels (Figure 1B). Uniaxial stretchability reaches ~3000%, and cyclic loading-unloading tests show recovery ratios exceeding 98% even at 1500% strain. These hydrogels exhibit minimal strain-rate dependence and recover fully within seconds after extreme deformation. In compression, they retain superb resilience over 1000 cycles without significant hysteresis. Most impressively, biaxial stretching tests confirm an areal strain of 10,000%, nearly an order of magnitude higher than previous benchmarks. Finite element simulations of balloon-like actuators further validate the uniform strain distribution, consistent with experimental observations. Collectively, these properties significantly surpass those of conventional rubbers and soft hydrogels, establishing a new mechanical paradigm for water-rich polymer networks.

The authors provide direct experimental evidence for the formation and reversibility of the PNC architecture using atomic force microscopy, cryogenic transmission electron microscopy, and small-angle X-ray scattering (Figure 1C). Subnanometric bead sizes below 2 nm were observed, and SAXS revealed isotropic networks with interbead distances ranging from 1.14 and 2.09 nm. These findings confirm the dense packing of PNC assemblies within the hydrogel matrix. Theoretical analysis further indicates that the maximum extensibility of PNC chains falls between that of freely jointed chains (FJCs, scaling as $\lambda_{max} \sim N^{1/2}$) and coiled chains (CCs, scaling as $\lambda_{max} \sim N^{2/3}$). Given that a constant monomer fraction (~0.8) participates in bead formation, the extensibility λ_{PNC} tends toward the upper limit of $N^{2/3}$, fundamentally breaking the classical $N^{1/2}$ scaling law that governs conventional hydrogel elasticity. This molecular-level design provides the structural basis for achieving ultralarge reversible deformations that surpass the limitations of traditional hydrogel architectures.

In addition to their hyperelasticity, these hydrogels exhibit remarkable damage tolerance and rapid self-healing properties. When punctured or cut, the material rapidly self-heals within seconds due to mobile water molecules and dynamic electrostatic interactions, allowing reinflation of pneumatic devices without leakage. Such behavior directly addresses a critical barrier in pneumatic actuators. Furthermore, the hydrogels retain an ionic conductivity of 4.9 mS/cm even after damage and autonomous healing, ensuring concurrent mechanical robustness and electrical functionality. This integrated feature enables intrinsic self-sensing of deformation, as demonstrated by consistent resistance-time signals captured during repeated grasp-release cycles.

The synergy between hyperelasticity, self-healing, and self-sensing is vividly demonstrated in pneumatic grippers (Figure 1D). These grippers withstand volume expansions of nearly 1000 times their original size, and are capable of manipulating objects ranging from fragile strawberries to rigid items orders of magnitude larger. Remarkably, they remain operational after puncture, immediately sealing and reinflating to resume gripping tasks. These demonstrations underscore the synergistic integration of hyperelasticity, autonomous healing, and self-sensing, establishing a new paradigm for the design of soft robotic actuators that are resilient, adaptive, and multifunctional.

The broader implications of this work are far-reaching, spanning multiple disciplines from soft robotics to materials physics. First, it redefines the theoretical ceiling of hydrogel elasticity, proving that extensibility limits long considered fundamental can be overcome by supramolecular design. This insight is poised to influence not only hydrogel science but also fields such as elastomer physics, supramolecular polymer chemistry, and network theory. Second, it exemplifies how molecular conformations can be harnessed as design variables in materials science. The pearl-necklace state, once a theoretical construct in polyelectrolyte physics, is here translated into a macroscopic property with direct technological utility, highlighting the power of integrating statistical mechanics with practical material engineering. Third, the multifunctional of these hydrogels combining hyperelasticity, autonomous self-healing, and ionic conductivity, directly meets the stringent requirements of next-generation soft devices. In the field of soft robotics, where actuators demand resilience, environmental adaptability, and embedded intelligence, these materials offer a unified platform that seamlessly integrates structural integrity with diverse functional capabilities. Similarly, for bioelectronics and implantable systems, their inherent compliance, rapid elastic recovery, and bio-ion-based conduction mechanism open avenues for the development of highly biocompatible, durable, and self-monitoring devices.

Despite these advances, several challenges must be addressed before practical deployment. The hydrogel's dependence on hydration raises questions about long-term stability under harsh environmental conditions such as dehydration, high salinity, or fluctuating humidity. Although the demonstrated ambient stability over one month is promising, yet a systematic evaluation of long-term performance under continuous or harsh conditions represents a critical next step. Additionally, although the strain capacity is extraordinary, the actual stress values remain moderate compared to those of engineering elastomers, which may restrict use in load-bearing contexts. Scaling the controlled heterogeneous hydration process for manufacturing could also present challenges, necessitating further optimization to ensure uniformity and reproducibility.[5] Finally, integration into complex biomedical or robotic systems will require comprehensive assessment of biocompatibility, cyclic durability, and interfaces with other functional materials.

Looking forward, the design principle of pearl-necklace chains offers a versatile strategy for constructing polymer networks that transcend traditional mechanical boundaries. The combination of ultrahigh stretchability, fast recovery, puncture tolerance, and ionic conductivity establishes a new paradigm in hydrogel mechanics. Near-term applications hold significant potential in soft pneumatic robotics and wearable sensors. Looking further ahead, this approach may pave the way for adaptive biomaterials, deployable medical devices, and responsive architectural elements requiring both high

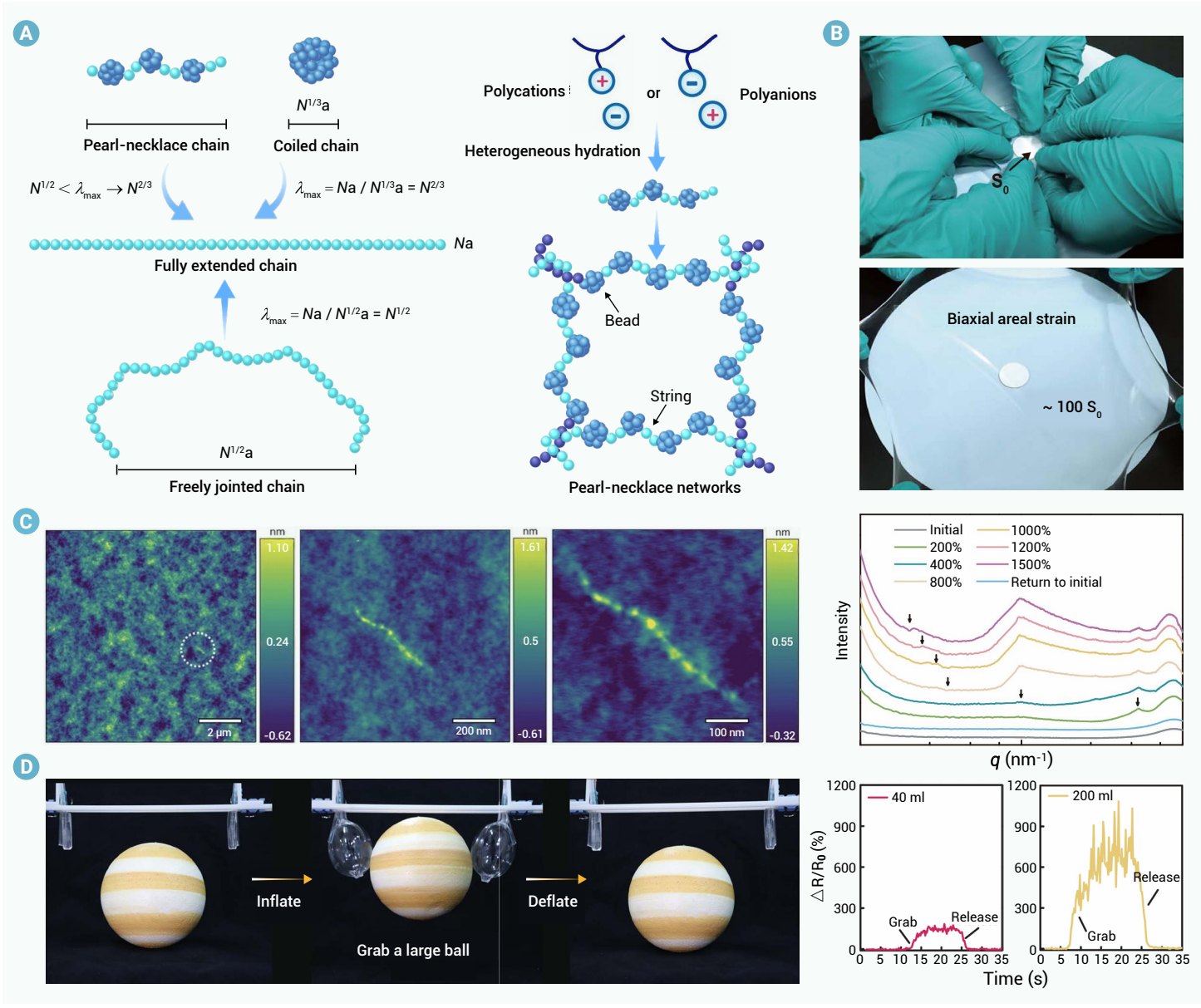

Figure 1. Schematic design and application demonstration of hyperelastic hydrogels (A) Pearl-necklace chain model showing bead–string conformations for enabling ultra extensibility. (B) Hydrogel film exhibiting ultra-large biaxial areal strain up to ~100-fold its initial area. (C) AFM and SAXS evidence of pearl-necklace networks with reversible structural evolution under high strain. (D) Hyperelastic hydrogel pneumatic grippers achieve large-object handling with self-sensing based on strain-regulated ion transport and resistance variations.

elasticity and multifunctionality.

The hyperelastic hydrogel reported by Chen et al. represents a landmark in soft matter design. By leveraging pearl-necklace chain conformations, the authors have unveiled a new regime of elasticity, circumventing the constraints of classical chain statistics and redefining the mechanical possibilities of hydrogels. The result is not only a material with unparalleled deformability but also a conceptual bridge linking molecular theory to macroscopic performance. In a broader perspective, this work exemplifies how supramolecular engineering can unlock new paradigms in material design. Following the paradigm shift brought by supramolecular plastics in polymer sustainability, pearl-necklace hydrogels now establish a new cornerstone for elastic materials. These breakthroughs collectively suggest that intrinsic material boundaries can act as catalytic platforms for innovation, ultimately reimagining the limits of design.

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DECLARATION OF INTERESTS

The authors declare no competing interests.