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**Advancing Reversible Shape Memory by Tuning Network Architecture** QIAOXI LI, JING ZHOU, MOHAMMAD VATANKHAH VARNOSFADERANI, University of North Carolina at Chapel Hill, DMYTRO NYKYPANCHUK, OLEG GANG, Brookhaven National Lab-CFN, SERGEI SHEIKO, University of North Carolina at Chapel Hill, UNIVERSITY OF NORTH CAROLINA AT CHAPEL HILL COLLABORATION, BROOKHAVEN NATIONAL LAB-CFN COLLABORATION — Recently, reversible shape memory (RSM) has been realized in conventional semi-crystalline elastomers without applying any external force and synthetic programming. The mechanism is ascribed to counteraction between thermodynamically driven relaxation of a strained polymer network and kinetically preferred self-seeding recrystallization of constrained network strands. In order to maximize RSM's performance in terms of (i) range of reversible strain, (ii) rate of strain recovery, and (iii) relaxation time of reversibility, we have designed a systematic series of networks with different topologies and crosslinking densities, including purposely introduced dangling chains and irregular meshes. Within a broad range of crosslink density ca. 50-1000 mol/m$^3$, we have demonstrated that the RSM's properties improve significantly with increasing crosslink density, regardless of network topology. Actually, one of the most irregular networks with densest crosslinking allowed achieving up to 80% of the programmed strain being fully reversible, fast recovery rate up to 0.05 K$^{-1}$, and less than 15% decrease of reversibility after hours of annealing at partial melt state. With this understanding and optimization of RSM, we pursue an idea of shape control through self-assembly of shape-memory particles. For this purpose, 3D printing has been employed to prepare large assemblies of particles possessing specific shapes and morphologies.

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