

ing energies with Li_2S , the researchers selected poly(vinylpyrrolidone) (PVP) as a promising choice due to the high affinity between the $>\text{C}=\text{O}$ groups in PVP and Li_2S and Li-S^\cdot species. Subsequent electrochemical testing with coin cells assembled with Li_2S cathodes using PVP binders confirmed their hypothesis.

The high binding energies of PVP with Li_2S and Li_2S_n allowed for uniform dispersion of the active material and carbon within the electrode and minimized loss of polysulfides into the electrolytes during cycling. These batteries also exhibited an initial specific capacity of 760 mA h g^{-1} and 94% capacity retention in the first 100

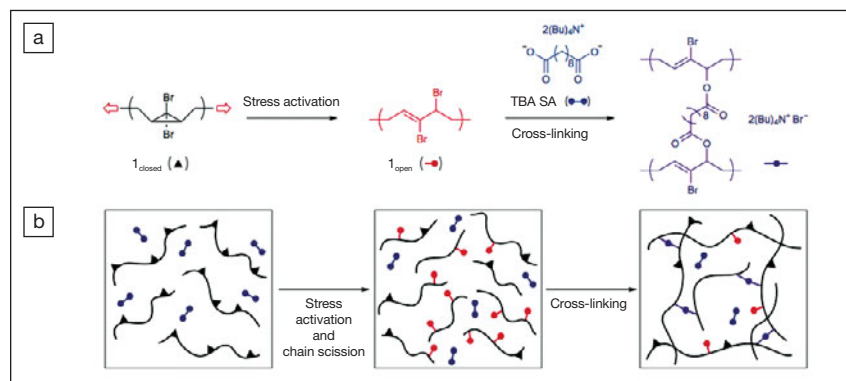
cycles. Even after 500 cycles, the PVP cells retained up to 69% of their initial capacity. The researchers envisage that their simple strategy of rational binder selection could be extended to the identification of new binders for other promising high-capacity electrode materials.

Dominica H.C. Wong

Shear forces self-strengthen mechanochemically active polymers

When polymeric materials are subjected to shear stresses, they typically suffer from bond breaking, which reduces their molecular weight and viscosity. Indeed, this is a problem that plagues oils and lubricants in high-performance engines. Now A.L. Black Ramirez of Duke University, J.A. Orlicki of the Army Research Laboratory, M. Champhekar of North Carolina State University, and their colleagues have developed synthetic polymers where the same forces responsible for the destructive processes of bond scission and chain disentanglement are channeled into constructive, bond-forming reactions.

As reported in the September issue of *Nature Chemistry* (DOI: 10.1038/nchem.1720; p. 757), the researchers inserted “mechanophores” (i.e., functional groups that respond to mechanical perturbation in a controlled manner) in the covalent backbone of the polymers, which enable remodeling and self-repair under mechanical stress. This concept of “activated remodeling through mechanochemistry” (ARM) is illustrated in the figure. Gem-dibromocyclopropanes (gDBCs, 1_{closed}) were embedded within a poly(butadiene) (PB) backbone as mechanophores, which provide the basis for self-repair through covalent cross-linking. When mechanically triggered, these undergo a ring opening polymerization, which leads to a 2,3-dibromoalkene product (1_{open}) open to nucleophilic substitution. The parent gDBC mechanophore is inert toward nucleophilic substitutions, so the repair reaction only occurs when sufficient forces are



Mechanochemistry self-strengthening concept: (a) A gDBC mechanophore within a polymer chain under tension undergoes a ring-opening reaction from 1_{closed} to 1_{open} . This increases the contour length and provides an allylic bromide that is capable of self-strengthening through nucleophilic displacement reactions. (b) System-wide force causes chain scission, but also activates the mechanophore (black triangle to red dot), which subsequently reacts with a cross-linker (blue) to form an active cross-link (purple) that overcomes the damage. Reproduced with permission from *Nature Chem.* 5 (2013), DOI: 10.1038/nchem.1720. © 2013 Macmillan Publishers Ltd.

experienced within the polymer to cause the non-scissile, ring-opening reactions.

The researchers tested the ARM response to shear forces of twin-screw extrusion, a common technique for processing bulk polymers. The conditions employed were destructive on a molecular level, where extrusion of 1_{closed} for 30 minutes at 40°C and 50 revolutions per minute generated bulk shear stresses of $0.15\text{--}0.25 \text{ MPa}$. While these caused bond scission and a reduction in molecular weight from 780 kDa to 560 kDa, these shear forces also mechanically activated the conversion of 7% of the gDBCs into their open form. In contrast, when the extrusion is repeated in the presence of suitable dicarboxylic acid molecules, which can cross-link the polymer chains via nucleophilic reaction with 1_{open} , the mechanically triggered cross-linking outcompetes the destructive shear forces during the extrusion process. The result is that the polymer becomes significantly stronger,

with an increase in the elastic modulus of nearly two orders of magnitude.

In the current system, remodeling takes place throughout the material and occurs in conjunction with irreversible deformation. However, the results raise the intriguing possibility of localized mechanochemical self-strengthening of “at-risk” regions within a load-bearing material. This research therefore has implications in various solid-state applications, where stress-induced bond scission or chain slippage triggers formation of microcracks, which propagate and eventually lead to material failure. Underlying the response is a new principle for materials design, in which chemomechanical coupling is used to capture otherwise destructive mechanical energy and funnel it into constructive processes. As in biological systems, it may be possible to actively remodel a synthetic material in response to its physical environment.

Jean Njoroge

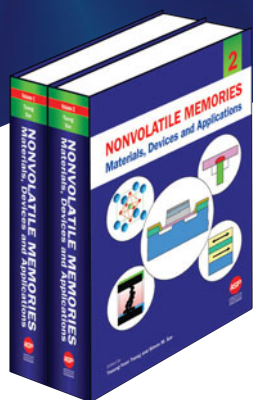


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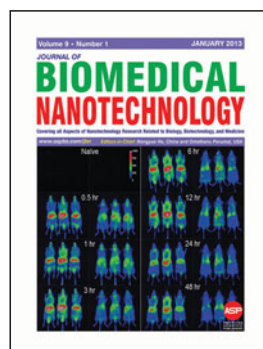
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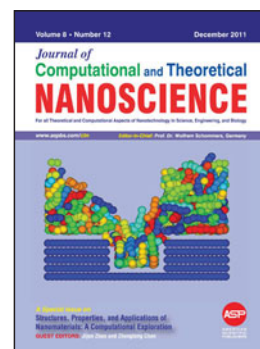
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