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Supplementary Data for

Autonomous Self-Healing and Highly Stretchable Polymer Maltose Polyborosiloxane for Improving Soft Electronics and Soft Robots

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Supplementary Table

Table S1. Comparison of performances of self-healing PM PBS in relation to previously reported self-healing composites.

Materials/ Mechanism	Self-healing time	Healing efficiency	Healing condition	Maximum strain at break	Availability for soft robot	Reference
PDMS/clay hydrogel	24 h	98%	RT	1500%	-	[1]
PDMS/MPU/IU	48 h	78%	RT	1200%	-	[2]
PAA/PDMAPS/APS	48 h	100%	RT	>10000%	Yes	[3]
Raw PBS	6 h	100%	RT	180%	Yes	[4]
Raw PBS/PDMS	24 h	20%	RT	2100%	-	[5]
Raw PBS/PDMS	72 h	86%	RT	340%	-	[6]
Raw PBS/Ecoflex	1 h	99%	RT	504%	-	[7]
Block PBS	30 s	100%	RT	1500%	-	[8]
PM PBS	**10 s**	**100%**	**RT**	**>3000%**	**Yes**	**This work**
Elastomer/Hydrogen bonds	3 h	90%	50 ˚C	950%	-	[9]
Elastomer/Boronic ester	16 h	95%	50 ˚C	446%	-	[10]
Elastomer/Ion-dipole interaction	24 h 24 h	25% 93%	RT 50 ˚C	2000%	-	[11]
Elastomer/Disulfides	2 h	90%	RT	923%	-	[12]
Hydrogel/Hydrogen bonding	30 min	-	90 ˚C	1400%	-	[13]
Hydrogel/Hydrogen bonding	30 s	100%	RT	>10000%	-	[14]
Hydrogel/Double network	2 h	97%	RT	1500%	-	[15]
Hydrogel/Imine bond	5 min	100%	RT	-	-	[16]
Hydrogel/dynamic ionic interaction	2 min	100%	RT	1500%	-	[17]
DA cross-links	4 h	93.4%	RT-80 ˚C-RT	-	Yes	[18]
SHeaLDS	24 h 1 h	77% 84%	RT 80 ˚C	1449% 1154%	Yes (for sensing only)	[19]

Supplementary Figures

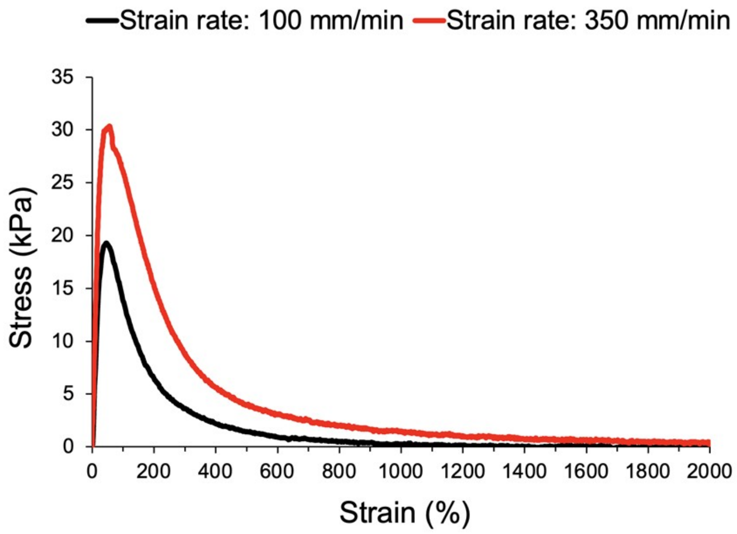


Fig. S1. Tensile stress-strain curves representative of PM PBS samples at strain rates of 100 mm/min and 350 mm/min.

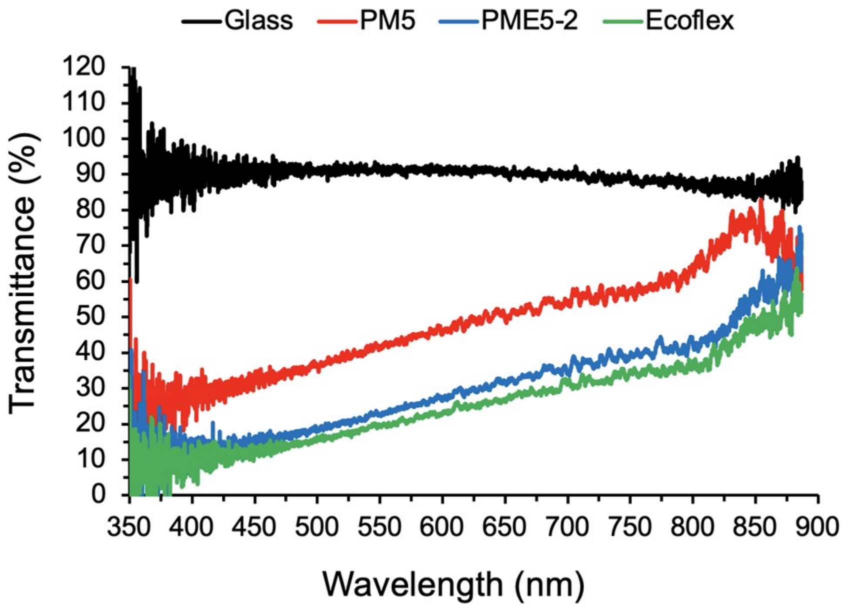


Fig. S2. Transmittance spectra of glass, PM5 (PM PBS), PME5-2, and Ecoflex. The thickness of each elastomer sample is 2 mm.

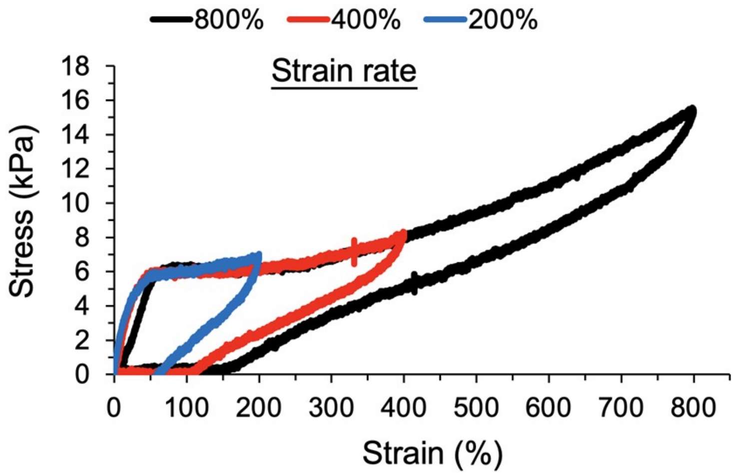


Fig. S3. Hysteresis loops acquired during tests with progressively increasing maximum strain (from 200% to 800%) of PME5-0.65. The strain rate is 10 mm/min.

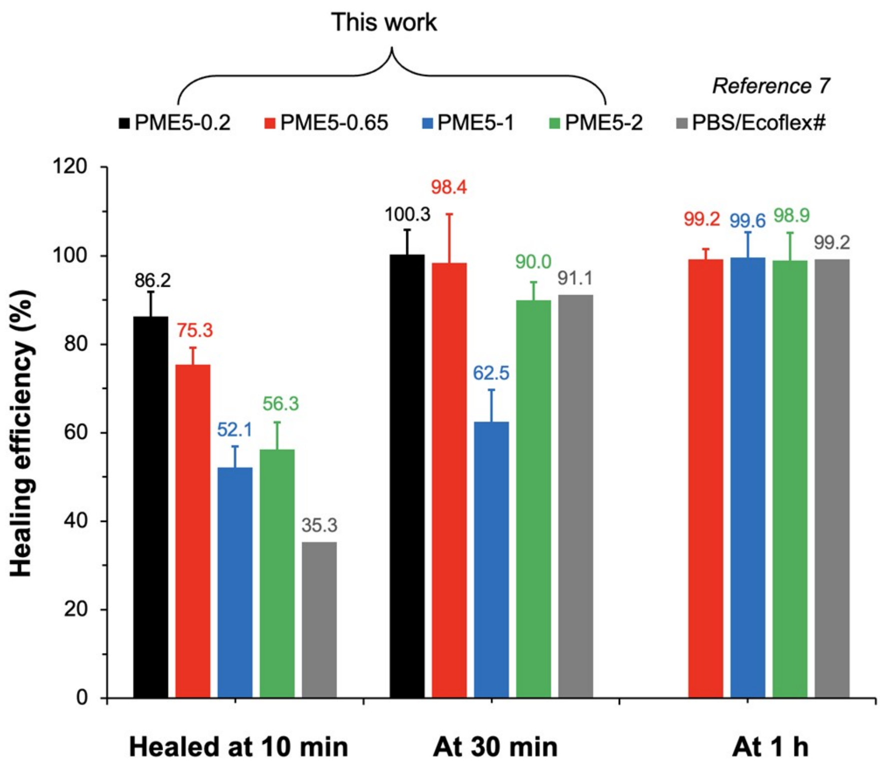


Fig. S4. Comparing healing efficiency for PMEs and previously reported PBS/Ecoflex composite [7] at different healing times (10, 30, and 60 min). The strain rate is 10 mm/min. Each dataset of PME represents three individual repetitions, as displayed in mean ± SD.

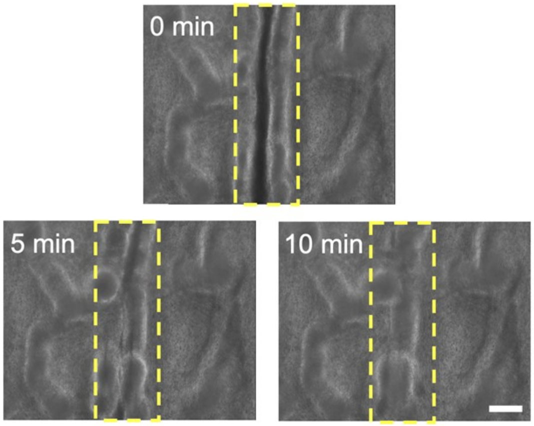


Fig. S5. Optical images of a cut PME5-0.65 sample before and after self-healing (10 min at room temperature). Scale bar, 200 μm.

Fig. S6. Demonstration of PME gripper dealing with different irregular objects.

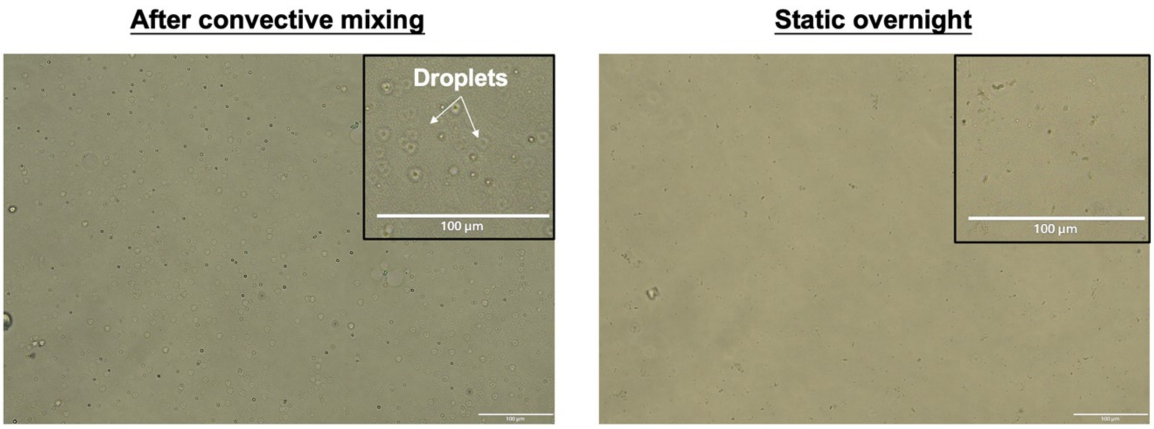
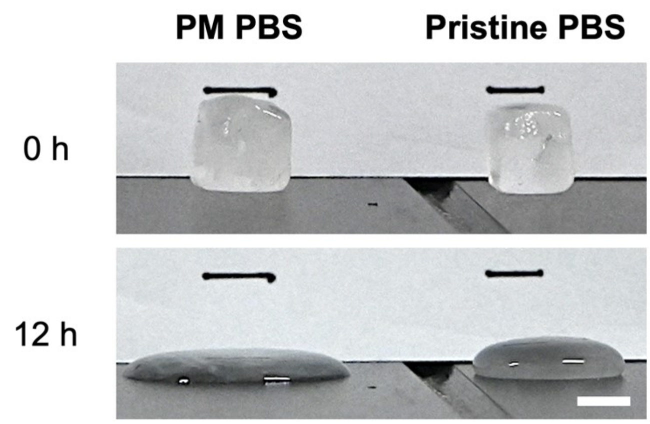


Fig. S7. Images of PM PBS after convective mixing and being left static overnight. It is worth noting that numerous droplets are only formed during the process of convective mixing. Scale bars, 100 μm.



Deforming rate	0.47 mm/h	0.33 mm/h

Fig. S8. Comparing deformation rates at ambient conditions: PM PBS versus pristine PBS. Scale bar: 5 mm.

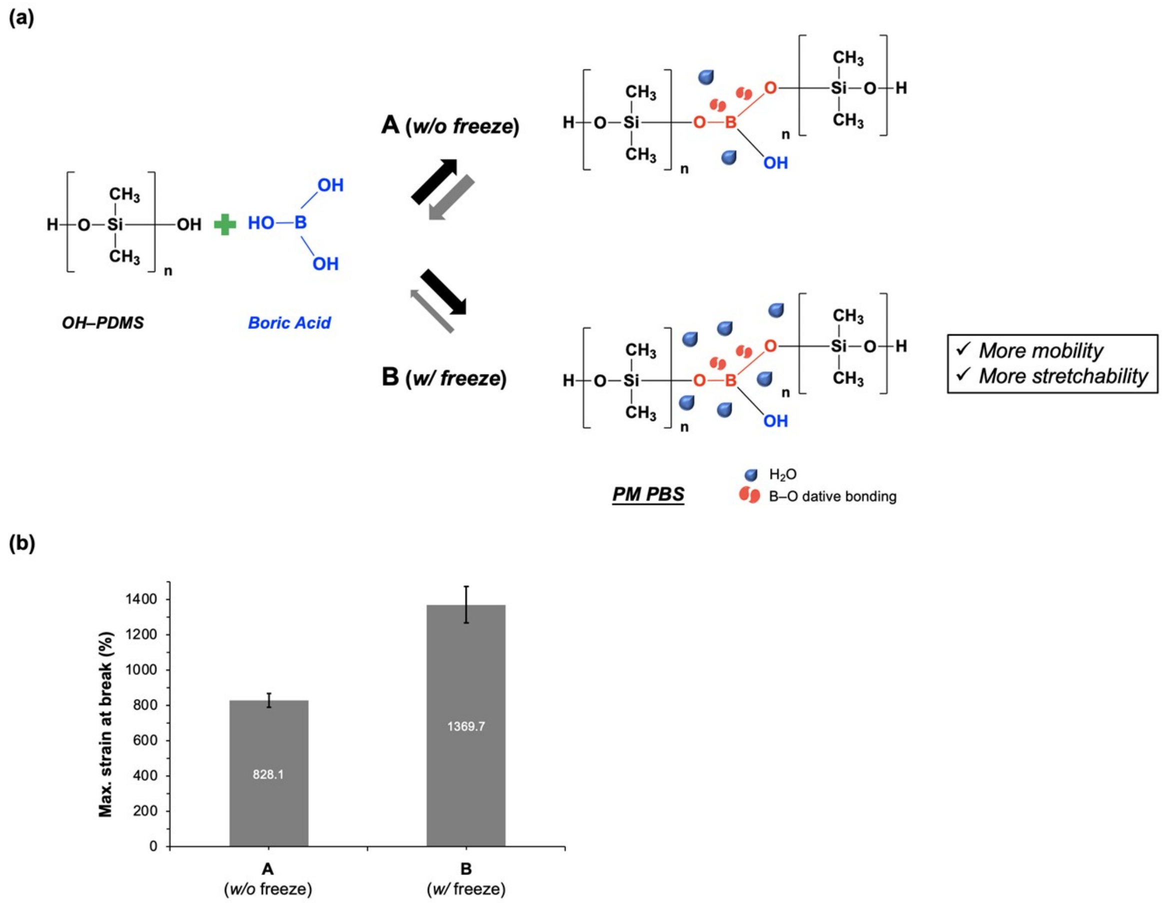


Fig. S9. Comparison of stretchability of PME5-1 under frozen and non-frozen conditions. (a) A schematic representation of the networks of PM PBS under both conditions. Condition **A** indicates PM PBS samples stored at ambient conditions until analysis, while Condition **B** denotes samples stored at -20 ˚C for 30 minutes and then thawed at ambient conditions before analysis. Freezing and thawing result in the generation of more water molecules within the PM PBS networks, enhancing their mobility and consequently stretchability compared to the initial samples. (b) Comparison of the maximum strain at break for PM PBS without freezing (Condition **A**; w/o freeze) and with freezing (Condition **B**; w/ freeze). It reveals an increase in maximum strain from 828.1% to 1369.7% after freezing and thawing, likely due to the increased presence of water molecules within the PM PBS network, thus enhancing its stretchability.

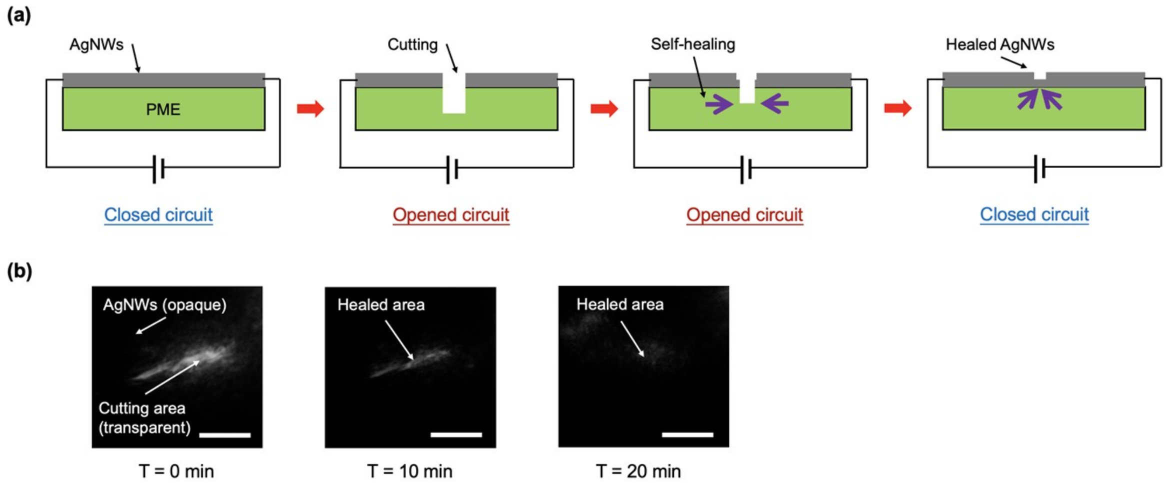


Fig. S10. Examination of self-healing AgNWs on PME substrate. (a) Depiction of self-healing mechanism in AgNWs-PME5-0.65 electrode. PME undergoes dynamic B-O dative bonding, enabling inner polymer networks to flow and facilitate self-healing, restoring damaged AgNWs. (b) Time-lapse images show cut AgNWs-PME5-0.65 merging over 20 minutes, observed through optical microscope. Scale bar, 10 μm.

Supplementary Video Captions

Video S1. Displaying the remarkable stretchability of PM PBS through manual stretching, highlighting that PM PBS can be extended up to 50 times its original size.

Video S2. Capturing the time-lapse tensile stress-strain testing of self-healing PM PBS. The PM PBS sample is initially cut into two separate pieces. After aligning the two segments and allowing them to heal for 10 seconds, the testing apparatus is activated to stretch the healed samples and halted once it reaches its travel limit.

Video S3. Video demonstrating the robust adhesion of PM PBS to two glass beakers underwater. It's noteworthy that the weight of PM PBS is 0.8 g, while the glass beaker weighs 75 g.

Video S4. Video demonstrating the capability of PM PBS as a self-healing electronic substrate underwater. It’s noteworthy that the healing time underwater is as quick as 20 s.

Video S5. Demonstration of the PME-0.65 tactile sensor.

Video S6. Demonstration of the self-healing PME5-2 gripper.

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