Supplemental Information

Tough and Circular Glass Fiber Composites via Tailored Dynamic Boronic Ester Interface

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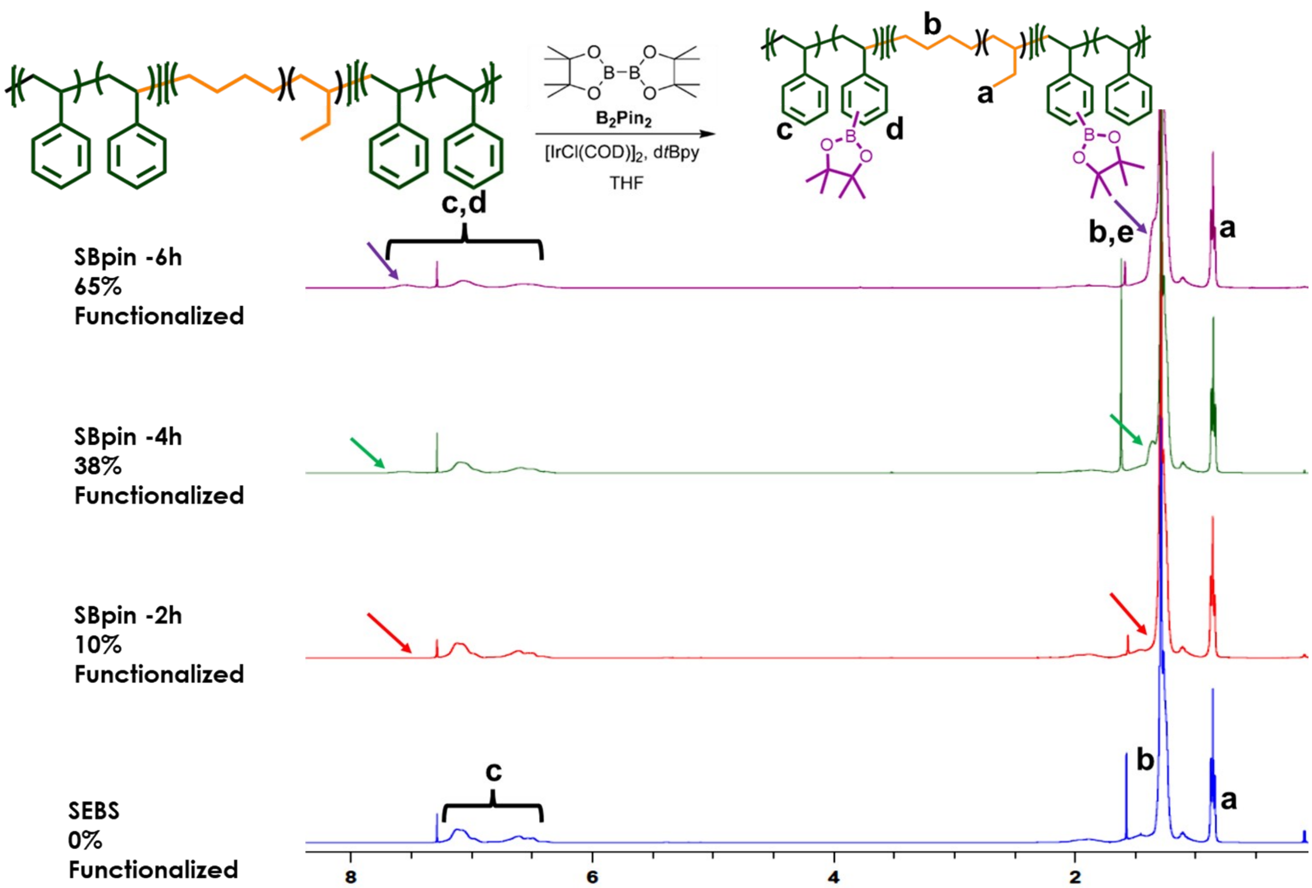

Figure S1. ^{1}H NMR analysis of tailored S-Bpin Polymers. The degree of functionalization was calculated based on the relative intensity of the methyl group in 1,2-butylene unit of the polymer chain (peak a) with respect to the increased integral ratio of the overlapping SEBS-methylene and S-Bpin methyl resonance (peaks b and e at 0.9−1.5 ppm). The right-side set of arrows shows the relative increase of the methyl group peak upon functionalization where the left arrow set shows the immerging of the new aromatic peak due to boronic ester grafting. It was observed that 65%, 38%, and 10% of original styrene (30 mol%) was borylated with 6,4, and 2h reaction time. The concentration for both SEBS and S-Bpin (20 mg/0.6 mL) in $CDCl_3$ was kept constant during the measurement.

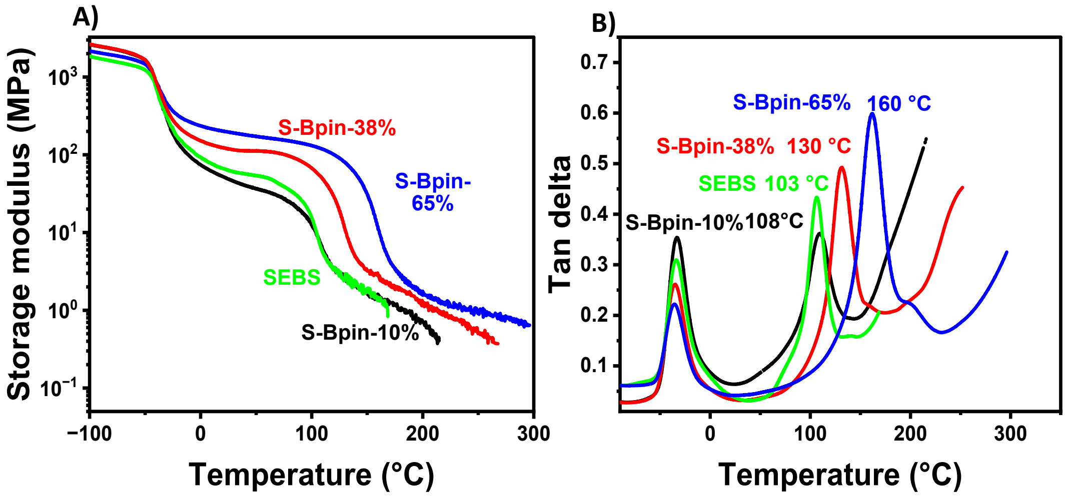

Figure S2. The thermomechanical analysis of different S-Bpin by Dynamic mechanical analysis (DMA); **A)** DMA curves of storage modulus as a function of temperature; **B)** tan delta versus temperature

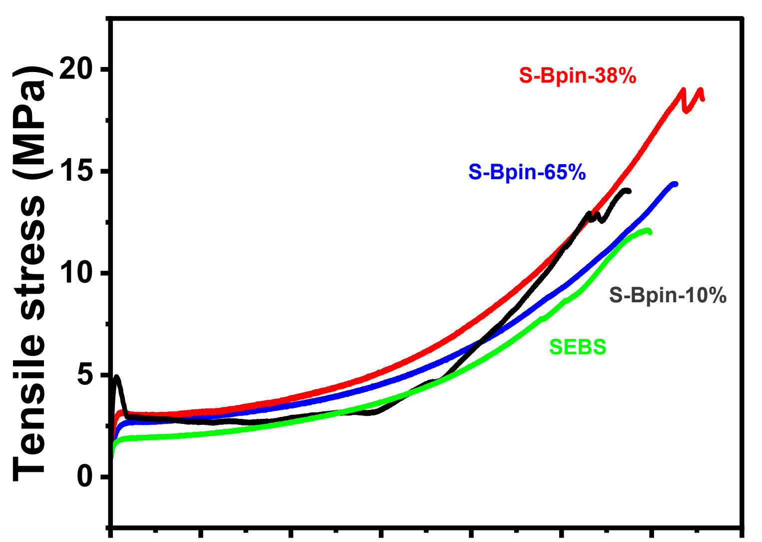

Figure S3. Tensile test analysis for SEBS and functionalized S-Bpin resin, (10,38, and 65%).

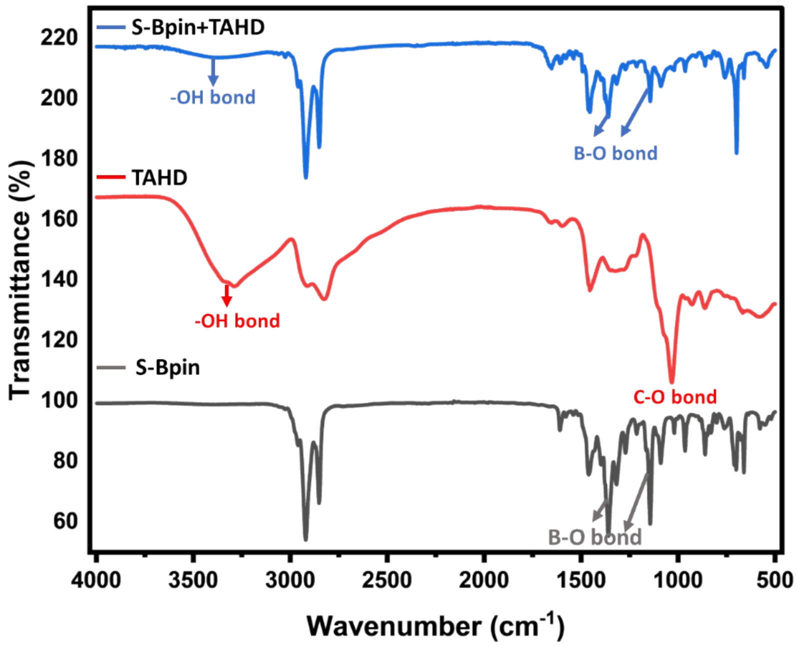

Figure S4. Comparison of FTIR spectra of S-Bpin, TAHD, and S-Bpin+TAHD resins. The FTIR spectrum of S-Bpin+TAHD shows sharp peaks at 1355 cm^{-1} and 1140 cm^{-1} corresponding to the B-O bond and broad -OH group disappearance of the crosslinker at 3500 cm^{-1}. In addition, the C-O stretching of the primary alcohol at 1070 cm^{-1}, which is prominent in TAHD spectrum, disappeared in S-Bpin+TAHD spectrum due to crosslinking with boronic ester.

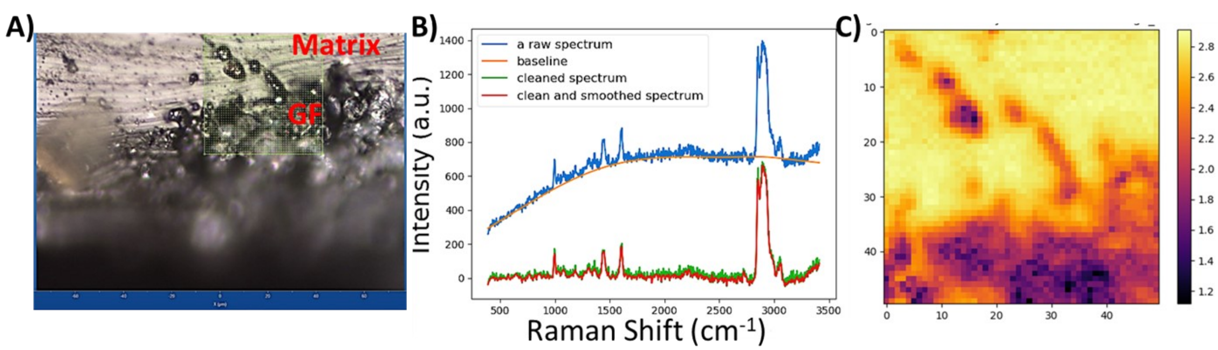

Figure S5. Optical microscopic and Raman image of the unsized GF_S-Bpin sample. A) Optical microscopic image of the unsized GF-S-Bpin sample with a green box indicating the scanned area. **B)** Single Raman spectrum with baseline correction (Prominent single peak at 2902 cm^{-1} for C-H stretching of polymer backbone) **C**) Raman imaging based on the intensity of the single peak (carbon G band centered at 2902 cm^{-1}).

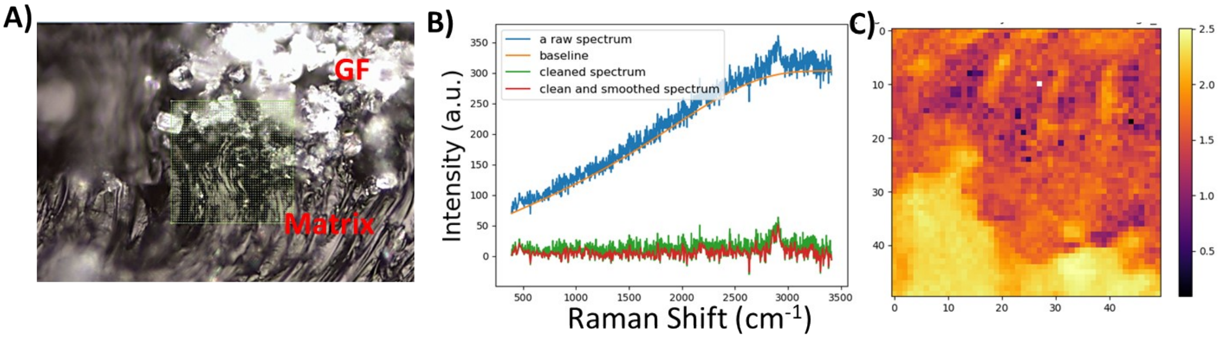

Figure S6. Optical microscopic and Raman image of the unsized GF_SEBS sample. A) Optical microscopic image of the unsized GF-SEBS sample with green box indicating the scanned area. **B)** Single Raman spectrum with baseline correction (broader single peak at 2902 cm^{-1} for C-H stretching of polymer backbone) **C**) Raman imaging based on the intensity of the single peak (carbon G band centered at 2902 cm^{-1})

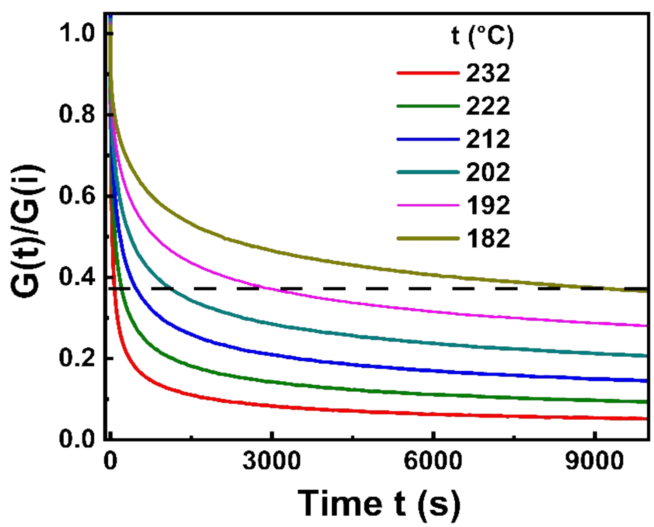

Figure S7. Normalized stress relaxation curves from Figure 4A re-plotted on linear time scale. The horizontal dashed line corresponds to a 1/e decay (0.37) of the normalized curves.

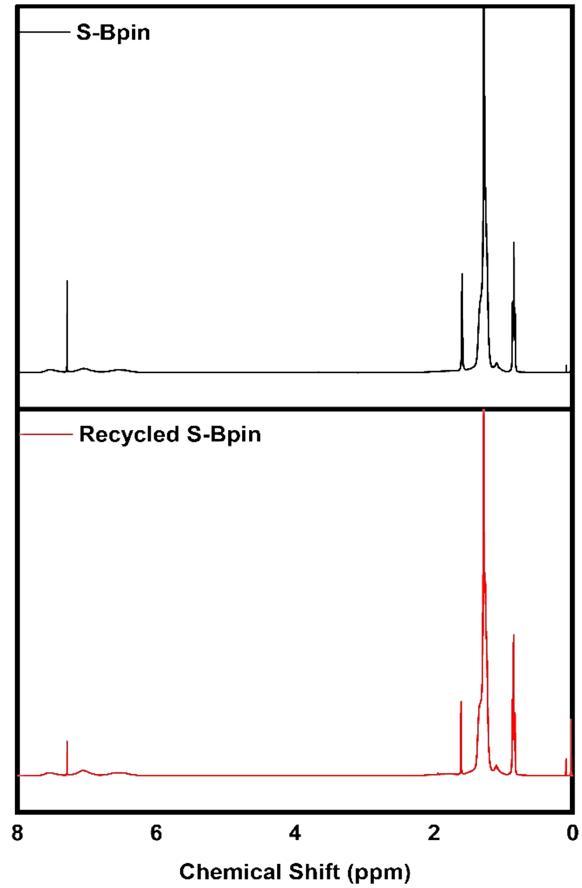

Figure S8. ^{1}H NMR spectrum of S-Bpin (black) and recycled S-Bpin (red) in $CDCl_3$. The S-Bpin polymer was collected without any change to its original chemical structure. The recycled S-Bpin ^{1}H NMR spectrum is the same as the original S-Bpin's ^{1}H NMR spectrum.

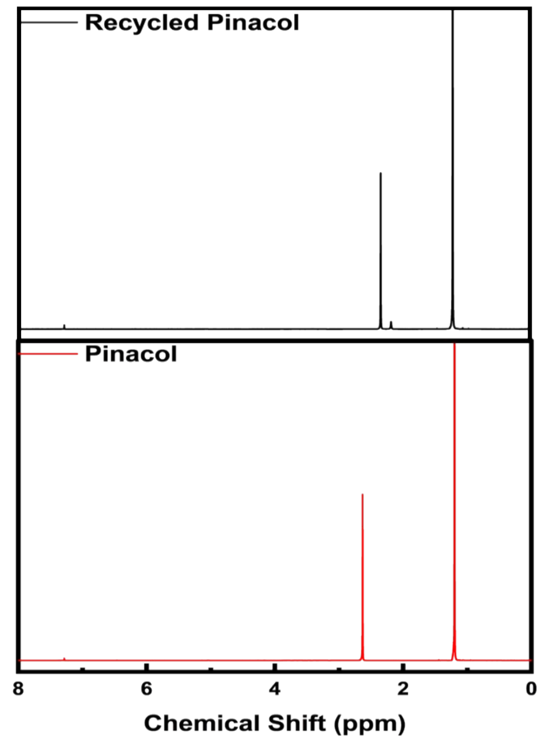

Figure S9. **^{1}H NMR Spectroscopy of pinacol and recycled pinacol in $CDCl_3$.** After the recrystallization pinacol can be obtained in its pure form.

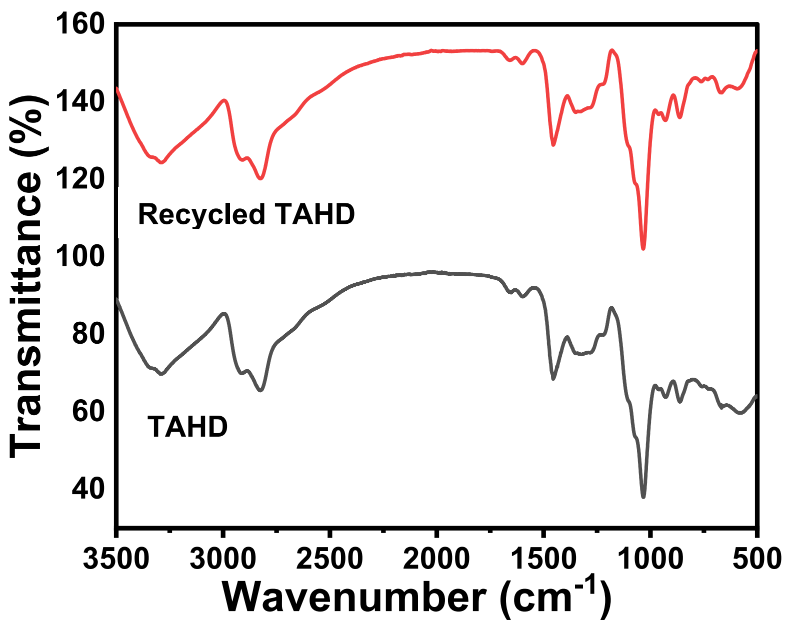

Figure S10. FTIR spectra of TAHD crosslinker and the recycled. Both spectra are identical to each other indicating that the recycled TAHD retains the chemical structure as the pristine.

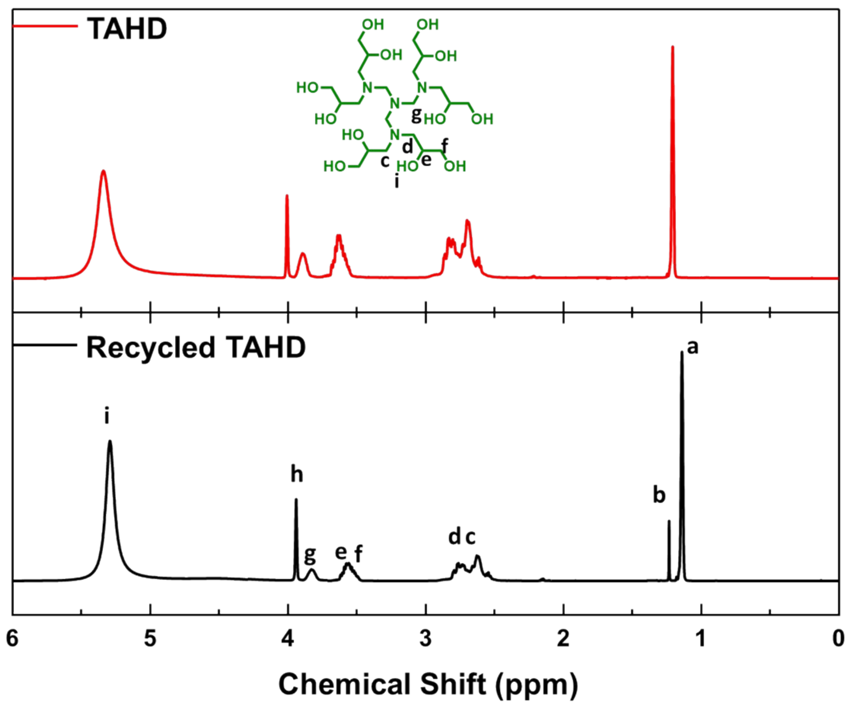

Figure S11. ^{1}H NMR Spectrum of the crosslinker TAHD and the recycled TAHD in deuterated isopropanol. The two spectra resemble each other except the peak b, which can be attributed to a small amount of pinacol residue left in the recrystallized sample. (Peaks a and h are given due to solvent peaks of IPA)

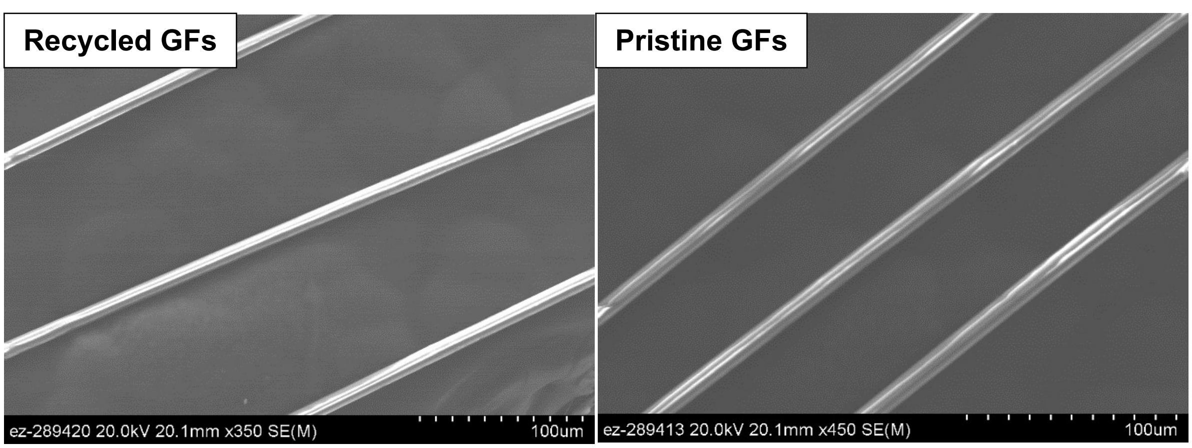

Figure S12. SEM images of the pristine S-glass fibers and recycled glass fibers.

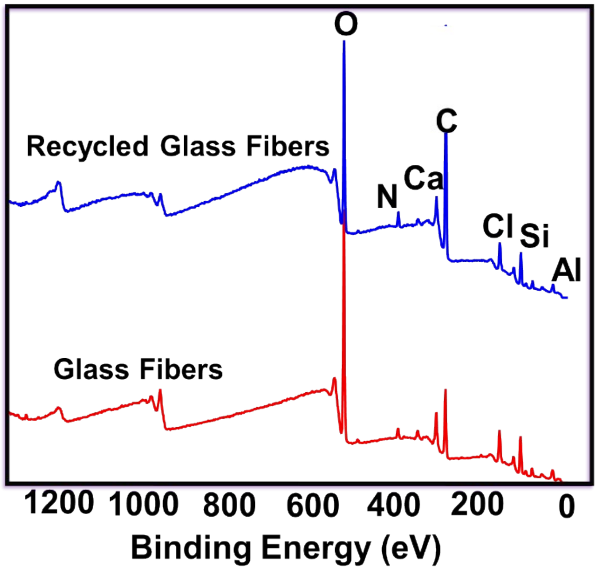

Figure S13.**The full scan of XPS spectra of unsized GF, and recycled GFs.**

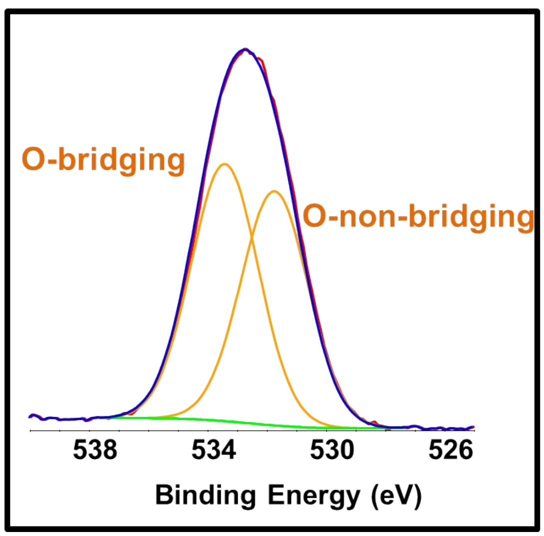

Figure S14. Fitted O1s XPS spectra for pristine GFs, exhibiting bridging and non-bridging oxygen on the fiber surface.

	BE/eV	FWHM	%
O1s_bridge	533.4	2.7	52.3
O1s_non_bridgie	531.8	2.7	47.7

Life Cycle Assessment (LCA)

Goal and scope of the study — The goal of this study is to estimate the environmental impact and energy consumption of S-Bpin composites containing 70% of GF reinforcement material and compare them to conventional GF composites. The inputs for the S-Bpin fabrication process in LCA modeling include commercial SEBS, solvents, and a catalyst. The amount of energy used during polymer synthesis and composite fabrication was also added to the model (Electricity, medium voltage, US). The data from performed preliminary LCA models for S-Bpin polymer synthesis is further used to build an LCA model for S-Bpin GFRP composites synthesized from

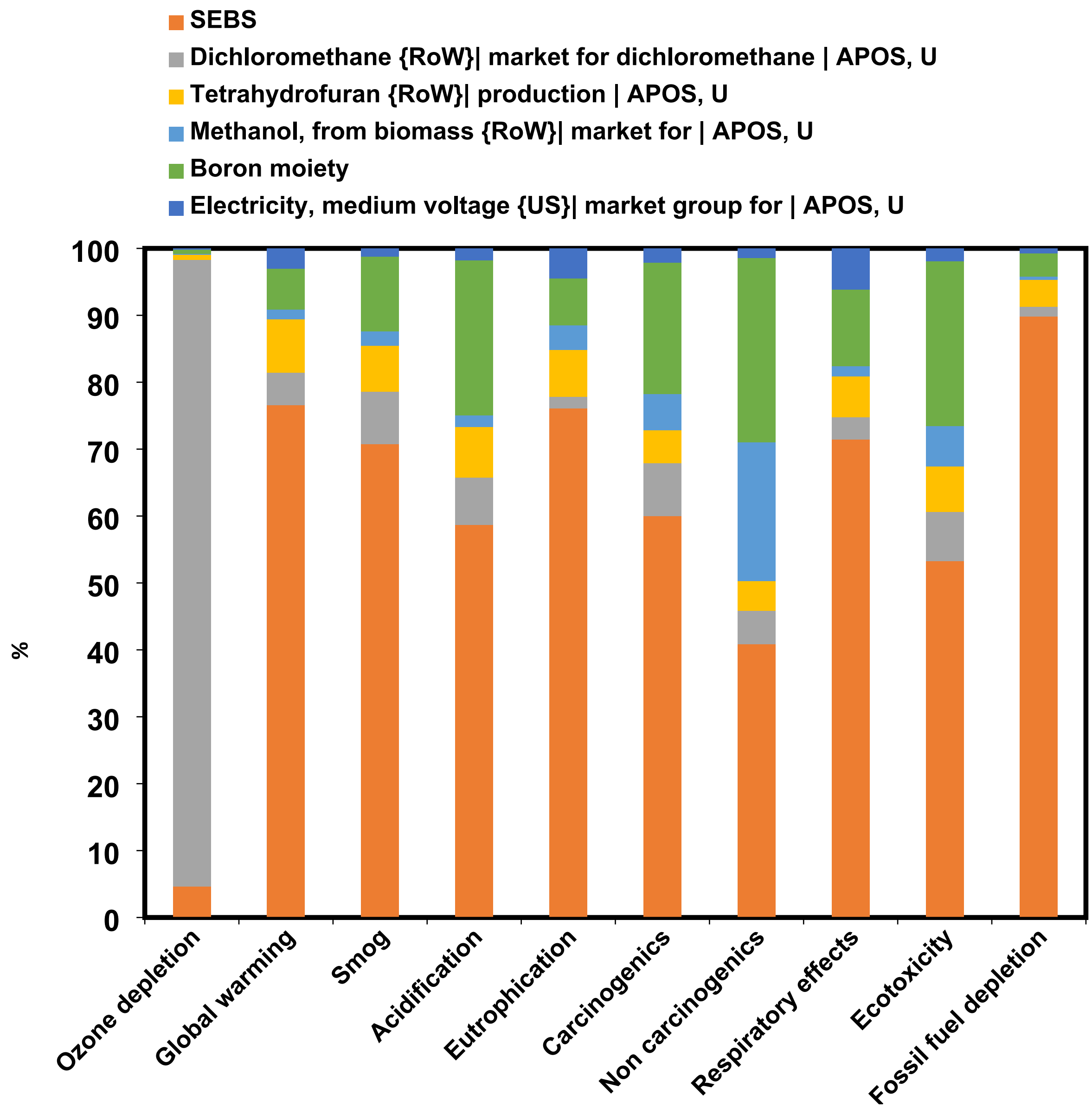

Fig S15. The impact assessment of SBpin polymer. Analyzing 1 kg 'SBpin; TRACI 2.1 V1.05 / US 2008 / Characterization / Excluding infrastructure processes / Excluding long-term emissions

70% of glass fiber and 30% of S-Bpin vitrimer. The compared LCA models for conventional GF composites were built based on the data provided by Ecoinvent V.3 database. The system boundary for this LCA study includes (1) S-Bpin polymer synthesis and (2) S-Bpin glass fiber-reinforced composite. The selected functional unit for this study is 1 kg of material (polymer/composite).

Life cycle inventory and Impact Assessment — The design of GFRP composites starts from the synthesis of S-Bpin, vitrimer fabrication, and finally incorporating GF into the composite. Data for the building of the life cycle inventory (LCI) model consist of the material and energy inputs to (1) synthesis of S-Bpin, (2) synthesis of S-Bpin vitrimer with the crosslinker, and (3) fabrication of glass fiber-reinforced composite. The inputs for composite fabrication modeling were based on lab-scale results. The information for the synthesis of S-Bpin and its vitrimer was taken from stoichiometric calculations of materials needed for polymer synthesis. In our model, we assume that the excess catalyst, solvents, and other functional materials used for chemical recycling are recycled and returned to the process.

The tool for reduction and assessment of chemical and other environmental impacts (TRACI) 2 method (including ten environmental impact categories, e.g., ozone depletion, and global warming) was used to study the environmental impact of the polymer synthesis and composite fabrication process. The energy consumption of glass fiber-reinforced polymer composite was expressed in MJ/kg $_{eq.}$ using values of the non-renewable, fossil fuels category.

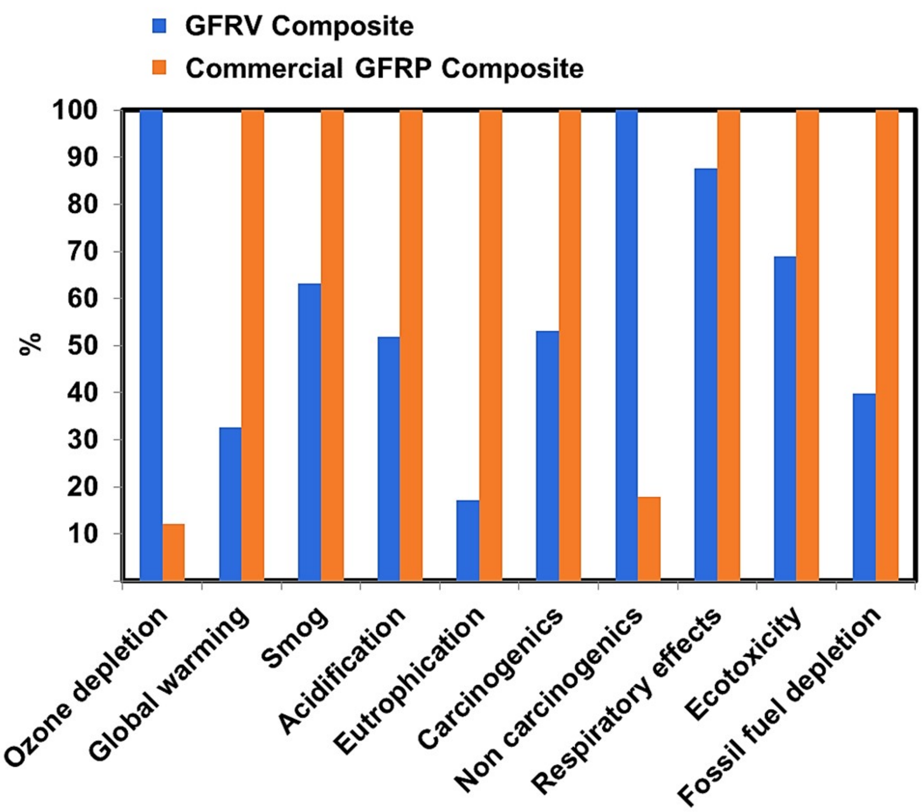

Figure S16. **The comparison of environmental factors between GFRV and commercial GFRP composites.** Comparing 1 kg GFRV composite with 1 kg injection molded, polyamide based GFRP composite (Method: TRACI 2.1 V1.05 / US 2008 / Characterization / Excluding infrastructure processes / Excluding long-term)

The LCA model for S-Bpin was developed to assess its environmental impact across various impact categories. Several assumptions were made, including a 90% solvent recycling rate and catalyst recycling, among others. Using this LCA model, the environmental impact of the GFRV composite was evaluated based on a polymer-to-filler ratio of 30:70. Data for the glass fiber was sourced directly from the Ecoinvent V.3 database. Carbon emissions and energy demand were calculated through LCA modeling (using SimaPro software), as the sum of the contributions from each component involved in the process.

The GHG emission per 1 kg of S-Bpin polymer was first calculated (Table S1). These emission value was then incorporated into the final CO_2 emission value for 1 kg of GFRV composite, which was determined based on the fiber content of 70% (Table S2).

Table S1. Major Components Contributing to GHG Emissions per 1kg of S-Bpin

Impact category	Unit	SBPin	SEBS	DCM	THF	CH3OH	B_2Pin_2	Electricity
Global warming	kg CO_2 eq	**3.52**	2.69	0.17	0.28	0.05	0.22	0.11

Table S2. Major Components Contributing to GHG Emissions per 1kg of GFRV Composite

Impact category	Unit	GFRV Composite	SBPin	Glass fiber	Electricity
Global warming	kg CO_2 eq	**2.84**	1.05	1.68	0.11

Similar to the GHG emission calculation, the non-renewable energy consumption was first calculated for 1 kg of S-Bpin polymer. Based on the fiber-to-polymer ratio, these values were then incorporated into the non-renewable energy consumption calculation for 1 kg of GFRV.

Table S3. Major Components Contributing to Non-Renewable Energy Consumption per 1kg of S-Bpin

Impact category	Unit	S-BPin	SEBS	DCM	THF	CH_3OH	B_2Pin_2	Electricity
Non renewable, fossil energy	MJ	**103.9**	91.7	1.5	4.9	0.6	3.9	1.3

Table S4. Major Components Contributing to Non-Renewable Energy Consumption per 1kg of GFRV

Impact category	Unit	GFRV Composite	S-BPin	Glass fiber	Electricity
Non renewable, fossil energy	MJ	**53.78**	31.17	21.32	1.28

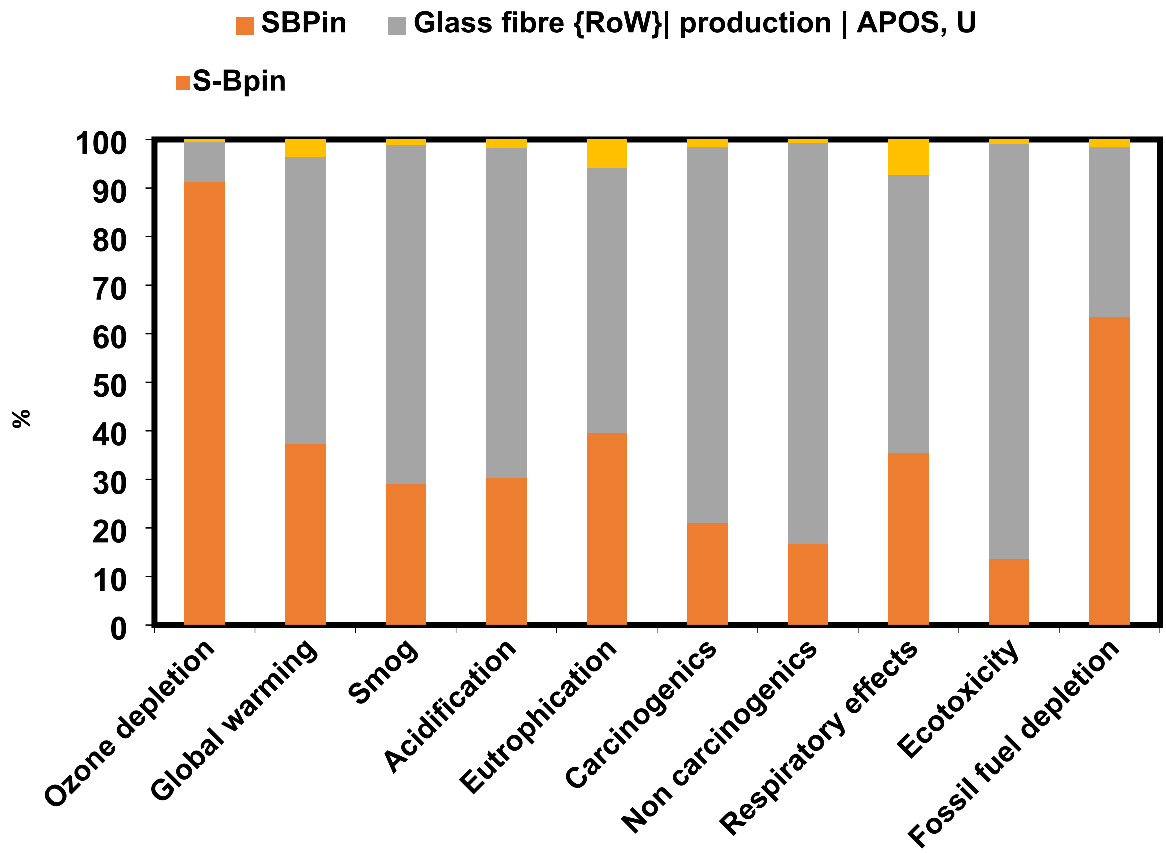

Fig S17. The impact assessment of GFRV composite. Analyzing 1 kg 'S-Bpin Composite 70% fiber. Method: TRACI 2.1 V1.05 / US 2008 / Characterization / Excluding infrastructure processes / Excluding long-term emissions

Table S5. Environmental Impact of our GFRV and commercial GFRP Composites

Impact category	GFRV Composite	Commercial GFRP composite
Ozone depletion kg CFC-11 eq	1.18×10^{-6}	1.43×10^{-7}
Global warming kg CO_2 eq	2.83	8.71
Smog kg O_3 eq	0.23	0.36
Acidification kg SO_2 eq	0.02	0.03
Eutrophication kg N eq	0.001	0.008

Carcinogenic CTUh	3.86×10^{-8}	7.29×10^{-8}
Non carcinogenic CTUh	7.94×10^{-7}	1.42×10^{-7}
Respiratory effects kg PM2.5 eq	0.003	0.004
Ecotoxicity CTUe	3.57	5.19
Fossil fuel depletion MJ surplus	6.17	15.5

Table S6. Energy Demand for GFRV and commercial GFRP composites.

Impact category	GFRV Composite/ MJ	Commercial GFRP composite/. MJ
Non-renewable, fossil	53.8	124.7
Non-renewable, nuclear	4.12	14.76
Non-renewable, biomass	0.0014	0.0009
Renewable, biomass	3.48	2.07
Renewable, wind, solar, geothermal	0.43	0.41
Renewable, water	1.29	1.40

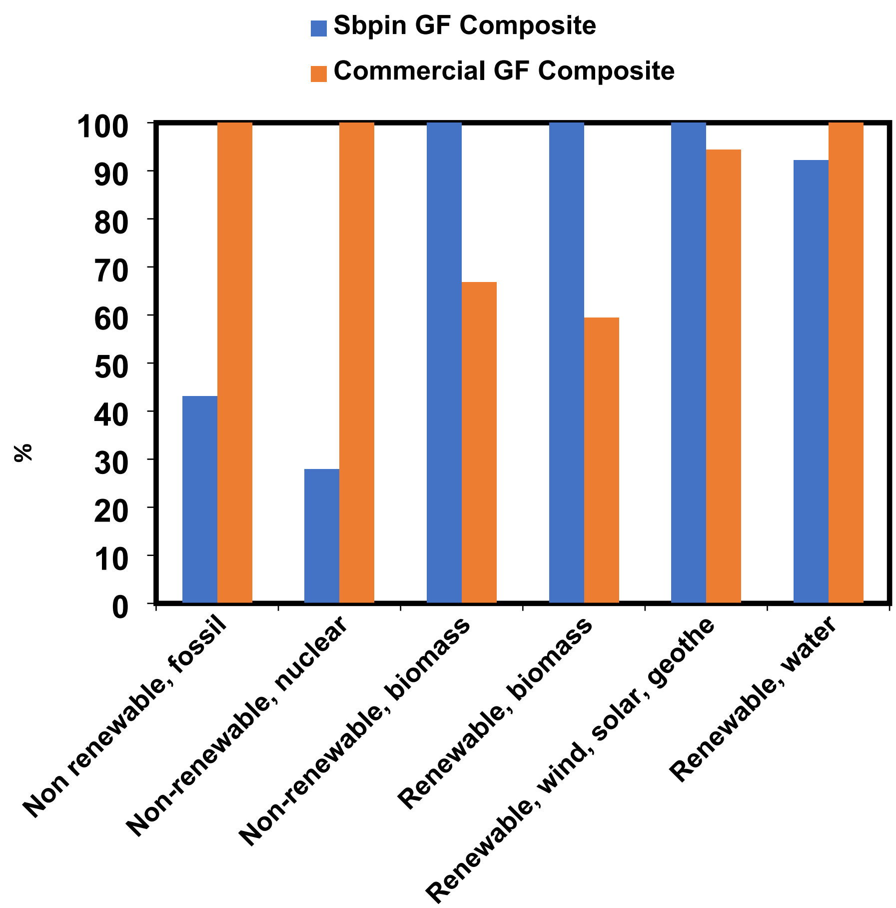

Figure S18. The bar chart for the energy demand calculation of GFRV vitrimer composite and commercial GFRP composites. Comparing 1 kg 'S-Bpin Composite 70% fiber' with 1 kg 'Glass fiber reinforced plastic, polyamide, injection molded {RoW}| production | APOS, U'; Method: Cumulative Energy Demand V1.11 / Cumulative energy demand / Characterization / Excluding infrastructure