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**Electronic Supplementary Information** 

Ultrastrong bonding, on-demand debonding, and easy re-bonding of non-sticking materials pair enabled by the reversibly interlocked macromolecular networks based Janus-like adhesive

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## 1. Materials

Poly(tetramethylene ether glycol) ( $M_n$  = 1000 g mol<sup>-1</sup>, PTMEG1000) was purchased from Aladdin and dehydrated at 110 °C in vacuum for more than 6 h to remove moisture. Dibutyltin dilaurate (DBTDL), isophorone diisocyanate (IPDI), 2,2'-dithiodianiline (DTDA), triethanolamine (TEOA), butyl acrylate (BA), acrylic acid (AA), 2,2'-azobis(2-methylpropionitrile) (AIBN) and glycidyl methacrylate (GMA) were obtained from Aladdin Reagent Ltd. Zirconium (IV) chloride (ZrCl<sub>4</sub>), 1,4-benzenediboronic acid, N,N-dimethylformamide (DMF, with molecular sieves, water < 50 ppm), tetrahydrofuran (THF), and hexane were supplied by Innochem Technology Co., Ltd. All chemicals and solvents were used as received without further purification unless otherwise stated.

Tempered ultra-clear figured glass (80 mm long, 60 mm wide and 3.2 mm thick) was purchased from Shenzhen TeSi Photonics Technology Co., Ltd. The fluorine polymer covered TPT back sheet was purchased from Hangzhou Hongfu Technology Co., Ltd. 304 McMaster-Carr stainless steel wire cloth with 100 × 100 mesh size (170 mm × 50 mm) was supplied by Suzhou Linzhe Hardware Product Co., Ltd.

## 2. Syntheses and preparation

# 2.1 Synthesis of Polyacrylate Single Network (SN-PA, i.e. Crosslinked Polyacrylate Containing Reversible Boronic Ester Bonds)

Firstly, the diol monomer, 2,3-dihydroxypropyl methacrylate (DHMA), was synthesized according to the method described elsewhere.<sup>1</sup> Briefly, GMA (14.20 g, 100 mmol) was added into deionized water (140 mL). Then, the emulsion was vigorously stirred at 80 °C overnight. Lastly, the obtained homogeneous aqueous solution was saturated with sodium chloride and purified by extraction, drying, filtration and concentration, offering the final colorless liquid. The structure of the resultant DHMA monomer was confirmed by proton nuclear magnetic resonance (<sup>1</sup>H NMR) and Fourier transform infrared (FTIR) spectra (Fig. S1). <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>; Me<sub>4</sub>Si)  $\delta$  (ppm): 6.08 (1 H, s), 5.54 (1 H, s), 4.14 (2 H, d), 4.05 (1 H, m), 3.95 (2 H, m), 3.74 (1 H, m), 3.63 (1 H, m), 3.55 (1 H, m), and 1.88 (3 H, s). The peaks on the <sup>1</sup>H NMR spectrogram corresponding to the characteristic peaks of the diol monomer are consistent with literature.

Besides, the FTIR spectrum discloses the characteristic peaks at 3370 cm<sup>-1</sup> and 1640 cm<sup>-1</sup>, respectively. The former is attributed to O-H and the latter to C=C, which confirms the formation of DHMA.



**Fig. S1** Synthesis and characterization of DHMA monomer. (a) <sup>1</sup>H NMR and (b) FTIR spectra of the resultant DHMA.

For obtaining SN-PA, as shown in Fig. S2, butyl acrylate (BA, 8.17 g, 63.4 mmol), acrylic acid (AA, 2.15 g, 31.67 mmol), DHMA monomer (0.80 g, 5 mmol), and AIBN (16.40 mg, 0.1 mmol) were dissolved in THF (30 mL). The mixture was degassed by dry argon gas bubbling for 1 h, heated to 70 °C and polymerized for 12 h. Then, the product was precipitated in 200 mL hexane. The white sticky precipitate was collected and dried at 60 °C overnight. The wight-average molecular weight of the prepolymer was found to be 2.8  $\times$  10<sup>4</sup> g/mol. Afterwards, the product was dissolved in DMF, and 1,4-benzenediboronic acid (0.33 g, 2



Fig. S2 Synthesis of SN-PA.

mmol) was added into the mixture. By pouring the final solution into a polytetrafluoroethylene (PTFE) mold and curing at 80 °C for 48 h, the colorless polymer film of SN-PA was available.

The structure of the resultant SN-PA was confirmed by the attenuated total reflection Fourier transform infrared (ATR-FTIR) and Raman spectra (Fig. S3). The boronic ester bonds of SN-PA were identified by the characteristic peak at 660 cm<sup>-1</sup> on the ATR-FTIR spectrum. Meantime, the peak at 1160 cm<sup>-1</sup> on the Raman spectrum is attributed to the B-O bonds and that at 845 cm<sup>-1</sup> to the B-O rings. The results prove the successful synthesis of SN-PA.



**Fig. S3** Characterization of the single networks and ILNs. (a) ATR-FTIR spectra of SN-PU, SN-PA, ILNs and recycled ILNs. (b) Raman spectra of SN-PU, SN-PA and ILNs.

# **2.2** Synthesis of Polyurethane Single Network (SN-PU, i.e. Crosslinked Polyurethane Containing Reversible Disulfide Bonds)

The PU single network was synthesized according to the following procedures (Fig. S4). Typically, the dried PTMEG1000 (5.00 g, 5 mmol) was dissolved in DMF (10 mL) and then the mixture of isophorone diisocyanate (IPDI, 2.67 g, 12 mmol) and dibutyltin dilaurate (DBTDL, three drops) were added. The mixture was stirred at 60 °C for 12 h in Ar atmosphere. Next, 2,2'-disulfanediyldianiline (DTDA, 0.993 g, 4 mmol) was added and the solution was kept for stirring for 12 h. The wight-average molecular weight of the prepolymer was  $2.5 \times 10^4$  g/mol. Subsequently, triethanolamine (TEOA, 0.298 g, 2 mmol) was added, allowing for the crosslinking reaction for 12 h. After that, a designed amount of  $ZrCl_4$  was dissolved in DMF

for 48 h in vacuum to obtain a yellow transparent film of SN-PU. Here, SN-PU refers to the samples containing Zr<sup>4+</sup> by default unless otherwise specified. The control, SN-PU excluding Zr<sup>4+</sup> (abbreviated as SN-PU<sub>Zr free</sub>), was prepared following similar procedures without the addition of ZrCl<sub>4</sub>. The structure of the resultant SN-PU was confirmed by ATR-FTIR and Raman spectra. As shown in Fig. S3, the peaks at 3343 cm<sup>-1</sup> and 1551 cm<sup>-1</sup> on the ATR-FTIR spectrum are attributed to the stretching vibration of N-H, while the peak at 480 cm<sup>-1</sup> on the Raman spectrum is assigned to S-S bonds. The results confirm the successful



Fig. S4 Synthesis of SN-PU.

synthesis of SN-PU.

#### 2.3 Synthesis of the interlocked macromolecular networks (ILNs) from SN-PA and SN-PU

According to the requirements of preparing interlocked macromolecular networks,<sup>2-4</sup> DMF was chosen to act as the common solvent for dissolving SN-PA and SN-PU. Besides, 100 °C was selected as the preparation temperature because the exchange reaction temperatures of boronic ester bonds and aromatic disulfide bonds are between room temperature and 100 °C.

Firstly, powdered SN-PU (5 g) and SN-PA (5 g) were soaked in DMF (50 mL) at room temperature under strong stirring for 2 h, and then the solution was heated to 100 °C. Next, a little water (3 mL) was added under stirring for 20 min with reflux condensation in argon until a homogeneous solution was formed. Eventually, the resultant solution was filtered, concentrated in vacuum, and poured into a PTFE mold for curing at 80 °C for 48 h. The obtained film was ILNs11, meaning the weight ratio of SN-PU to SN-PA is 1:1. Accordingly, ILNs31 and ILNs13 were synthesized following the same routes like ILNs11 except that different proportions of SN-PU and SN-PA were used. The leading ILNs studied in the current work is ILNs11, so that it is simply referred to as ILNs unless otherwise specified.

The chemical structure of the resultant ILNs was verified by ATR-FTIR and Raman spectra (Fig. S3). In comparison with the ATR-FTIR and Raman spectra of SN-PU and SN-PA, the spectrum of ILNs shows that no new covalent bonds were formed during the topological re-arrangement induced interlocking. Evidently, both SN-PU and SN-PA are independent in the ILNs, and no chemical reaction occurred between them.

### 2.4 Preparation of the control of the ILNs (i.e., interpenetrating polymer networks, IPNs)

To highlight the critical importance of our ILNs, simultaneous IPNs were prepared from the same raw materials of ILNs and then characterized. Typically, dried PTMEG1000 (5.00 g, 5 mmol), IPDI (2.67 g, 12 mmol), DBTDL (three drops), DTDA (0.99 g, 4 mmol), TEOA (0.29 g, 2 mmol), zirconium (IV) chloride (0.08 g, 0.34 mmol), BA (8.17 g, 63.4 mmol), AA (2.15 g, 31.67 mmol), DHMA monomer (0.80 g, 5 mmol), AIBN (16.40 mg, 0.1 mmol), and 1,4-benzenediboronic acid (0.33 g, 2 mmol) were dissolved in DMF (50 mL). The polymerization was carried out at 60 °C for 24 h under stirring in dry argon. Finally, the product was poured into a PTFE mold and cured at 60 °C for 48 h.

# 2.5 Gluing of tempered glass and fluorine polymer covered TPT back sheet together with ILNs as adhesive

A little DMF (2.78 μL/cm<sup>2</sup>) was dropped on the ILNs film and then the latter was tightly sandwiched between tempered glass and TPT. The glass/ILNs/TPT assembly was secured by clamps, sealed in a plastic bag, and ultrasonicated at 60 °C for 1 h. Subsequently, the assembly was hot-pressed at certain

temperature under 0.3 MPa for a period of time in vacuum, and then incubated at room temperature for 48 h.

The reason of using DMF lies in that it is a good solvent for the ILNs so that it would help to achieve better wetting of the ILNs on the substrates. It is only an experimental trick and the quantity is too little to influence the interior of the adhesive. The comparison test without using DMF was conducted, and the average peeling strength was found to be 50.32 N/cm, which is slightly lower than that with DMF (64.86 N/cm) but still much higher than the controls and the values reported in literature (refer to the corresponding part of the main text for more details). Therefore, the use of DMF exerts rather limited influence to the test results.

#### 2.6 Recycling and reuse of ILNs

The glass/ILNs/TPT assembly was immersed in 95% ethanol solution and ultrasonicated at 60 °C. After 1 h, the ILNs were completely dissolved, leaving no remains on the adherends (i.e. glass and TPT). For regaining the ILNs, the ethanol solution containing the dissolved ILNs was filtered and dried to constant weight. Then, the recycled ILNs were used to glue glass and TPT together again under the same conditions as those applied to the virgin ILNs mentioned above.

## 3. General Characterizations

Fourier transform infrared (FTIR) spectra were collected with a Bruker EQUINOX55 spectrometer.

Proton nuclear magnetic resonance (<sup>1</sup>H NMR) spectra were recorded at room temperature by an AVANCE III 400 MHz using CDCl<sub>3</sub> as solvent.

Raman spectra were measured in the single spot mode using a confocal Raman microscope (Thermo Fisher DXR3xi) with the wavelength of the excitation laser of 785 nm, laser intensity of 30 mW, exposure time of 0.1 s, and accumulation times of 1000.

Molecular weights of the prepolymers of SN-PA and SN-PU were measured by gel permeation chromatography (GPC, Breeze 2, Waters, USA) with THF as the eluent (concentration: 1 mg/mL).

Differential scanning calorimetry (DSC) tests were carried out using a TA Instrument DSC Q10 at a rate of 20 °C/min in nitrogen atmosphere. The samples were firstly heated up to 100 °C and kept for 5 min to

eliminate the thermal history, and then they were quenched to -80 °C. Next, the samples were heated from -80 °C to 100 °C.

Dynamic mechanical analysis (DMA) was conducted on a TA Instrument DMA 8500 using tension mode under 1 Hz at a heating rate of 3 °C/min.

Atomic force microscope (AFM) measurement was performed via the peak force quantitative nanomechanical (QNM) mode at room temperature. Prior to the test, the sample was dissolved in DMF and dropped onto a piece of silicon plate to obtain a smooth surface.

Optical photographs were taken using the KEYENCE VHX-1000C digital microscope.

The fractional free volume and the pore radius were tested using a positron annihilation lifetime spectrometer (PALS, ORTEC 0107A) with <sup>22</sup>Na as the radiation source. The radiation source was sandwiched between two samples and the lifetime spectra were recorded at room temperature until more than one million accumulated counts were achieved. The resulting lifetime spectra were analyzed and calculated.<sup>2</sup>

Contact angle measurements were performed on a DSA-100 contact angle analyzer (Krüss GmbH) using water and diiodomethane as test liquids. The surface free energies of the material was estimated from:<sup>2</sup>

$$\cos \theta = -1 + \frac{2\sqrt{\gamma_S^d \gamma_L^d}}{\gamma_L} + \frac{2\sqrt{\gamma_S^p \gamma_L^p}}{\gamma_L}$$
(S1)

where  $\theta$  is the measured contact angle,  $\gamma_L$  and  $\gamma_S$  are the total surface free energies of test liquid and solid,  $\gamma_S^d$  and  $\gamma_l^d$  refer to the dispersion components of the surface energies of solid and test liquid,  $\gamma_S^p$  and  $\gamma_l^p$  refer to the polar components of surface energies of solid and test liquid.

The interfacial tension,  $\gamma_{12}$ , was calculated using the following Harmonic Mean equation proposed by Wu,<sup>3</sup> which is preferred for the low energy systems such as polymers.

$$\gamma_{12=} \gamma_{12=} -\frac{4\gamma_1^d \gamma_2^d}{\gamma_1^d + \gamma_2^d} - \frac{4\gamma_1^p \gamma_2^p}{\gamma_1^p + \gamma_2^p}$$
(S2)

where  $\gamma_1$  and  $\gamma_2$  represent the surface energies of component 1 and component 2.

Rheological measurements were conducted on a rheometer (Kinexus pro+) with parallel plate geometry (diameter: 20 mm). Temperature sweeping was performed at the shear strain of 1% and

frequency of 1 rad s<sup>-1</sup>, while the frequency sweeping was conducted at different temperatures in a frequency range of 0.01-100 rad s<sup>-1</sup> at shear strain of 1%.

Element distributions of Zr and F of the glass/ILNs/TPT assembly were examined by using a ZEISS Gemini SEM250 thermal field-emission scanning electron microscope (SEM) equipped with an energy dispersive spectroscope (EDS) detector. Before the experiment, the glass/ILNs/TPT assembly was quenched in liquid nitrogen and the glass can be easily separated from ILNs/TPT. Then, the remaining ILNs/TPT was cut to expose fresh cross-section, which was further coated by carbon sputtering. Because the amount of Zr<sup>4+</sup> of ILNs (0.45 wt%) was too low to be detected by the EDS, the ILNs with higher Zr<sup>4+</sup> loading was prepared and measured instead.

X-ray photoelectron spectroscopy (XPS) study was carried out by using a Thermo Fisher Scientific ESCALAB 250 instrument with a monochromatic Al K $\alpha$  source. To characterize the interfacial interaction between SN-PU and TPT by XPS, the DMF solution of SN-PU (concentration = 50%), which had been prepared by heating at 100 °C in advance, was coated onto TPT and then the treated TPT was dried, frozen and ground into fine powders for the measurement. The sample was abbreviated as SN-PU/TPT. Besides, the SN-PU without Zr ions, SN-PU<sub>Zr free</sub>, was also coated onto TPT and pulverized in the same way for the contrastive analysis (called SN-PU<sub>Zr free</sub>/TPT). As for the investigation of the interfacial interaction between SN-PA and glass, the tempered glass sheet was ground into powders in advance, which were then mixed with the DMF solution of SN-PA (concentration = 50%) prepared by adding a little water during the dissolution. Then, the mixture (SN-PA/glass) was dried to constant weight and measured by XPS.

Analysis of the coordination of zirconium ions was conducted by using density functional theory (DFT) calculation and molecular dynamic (MD) simulation with the aid of Materials Studio software. (i) Firstly, the ligands were constructed and optimized by the DMol3 basis set<sup>4,5</sup> via DFT calculations at the level of gradient-corrected functional (GGA/BP). Then, a series of amorphous cells were constructed by  $ZrCl_4$  and the relevant ligands at the desired stoichiometries to explore the dynamics of the polymers. After simulated annealing of the above systems between 300 K and 800 K, the acquired energy-minimized monomer structures were available for the next MD simulations. (ii) The dynamics calculation based on NVT ensemble (isothermal and isochoric relaxation) was applied to balance the system structures for 50000 ps at ambient temperature for purposes of understanding the kinetic interactions of the active sites

of the polymers and metal ions. Afterwards, the dynamic calculation in terms of NVE ensemble (isoenergy and isochoric relaxation) was further carried out based on the balanced structures for 50000 ps within 50000000 steps at ambient conditions and the frames were output every 500 steps. (iii) The radial distribution function (RDF) analysis of the two groups between the active sites (ligands) and metal ions of all the frames was carried out within the cut-off radius of 10 Å at the interval of 0.02 Å. (iv) By taking advantage of the RDF data, the average distance between atoms and coordination structural unit (pair correlation function, PCF) was obtained and the coordination number was calculated from:

$$\int_{0}^{r} r^{2} * g(r) dr$$
(S3)

where *N* is the number of ions in the box, *r* the distance between the ions,  $\rho$  the particle density of the ions, and *g*(*r*) the pair correlation function.

Tensile tests were performed using dumbbell-shaped specimens (20×4×2 mm<sup>3</sup>) on a SANSCMT6103 universal tester at a crosshead speed of 100 mm min<sup>-1</sup>. The cyclic tensile tests were conducted on the same instrument at the strain of 100% and there was no waiting time between consecutive ten cycles.

Adhesive strengths of the materials were characterized by peeling strengths of the bonded versions. Firstly, all the adherends were cleaned through ultrasonication in distilled water and ethanol for three times, respectively. Then, the cleaned adherends were placed in an oven preset at 100 °C for 1 h until completely dry.

For making the glass/ILNs/TPT assembly for 180° peeling test (Fig. S5a), the ILNs film (60 mm × 60 mm) coated with a little DMF was firstly sandwiched between the tempered glass (80 mm × 60 mm) and TPT (130 mm × 60 mm). Then, the assembly was secured by clamps, sealed in a plastic bag and ultrasonicated at 60 °C for 1 h. Finally, the assembly was hot-pressed under 0.3 MPa at certain temperature for a given period of time in vacuum and incubated at room temperature for 48 h. In the case of 180° peeling test, the ends of the glass and TPT, which were not glued, were attached to the universal testing machine (WB-5A, China), and pulled to failure at an angle of 180° under a constant crosshead speed of 100 mm min<sup>-1</sup> (reference: Chinese standard GB/T 2790-1995). For understanding the performances of the control adhesives (i.e., SN-PA, SN-PU, IPNs, EVA, and 3M VHB adhesive) in gluing glass and TPT together, similar assemblies were fabricated using the same method.

On the other hand, the adhesion of SN-PA towards glass and that of SN-PU towards TPT were also quantified. For the former, the glass/SN-PA/glass assembly should have been used, but a substitute glass/SN-PA/stainless steel mesh was manufactured in reality. The reason lies in the fact the assembly of glass/SN-PA/glass is not suitable for the 180° peeling test due to the rigidity of glass. Therefore, one side of the glass/SN-PA/glass assembly was replaced by a flexible hydrophilic adherend, stainless-steel mesh (Fig. S5b).<sup>6</sup> Moreover, the glass/SN-PA/stainless-steel mesh assembly was directly incubated at room temperature for 48 h prior to the test. This is because the bonding between glass and SN-PA is very strong and the failure after the 180° peeling test of the glass/SN-PA/stainless-steel mesh assembly, which had been hot-pressed like the glass/ILNs/TPT assembly, always occurred at the interface with the stainless-steel mesh. The measurement result cannot tell the real adhesion of SN-PA towards glass. Meantime, the TPT/SN-PU/TPT assembly (Fig. S5c) was made following the same procedures described above. Because of the flexibility of TPT, the 180° peeling test of TPT/SN-PU/TPT consisting of TPT at both sides naturally turned into T-type peeling test. The peeling strength was calculated from the maximum force divide by the sample width.<sup>7,8</sup> All the tests were repeated for at least three times to obtain the averaged results.



**Fig. S5** Schematic illustrations of the samples for the peeling tests. (a, b) 180° peeling test. (c) T-type peeling test.



**Fig. S6** Homogeneity characterization of ILNs. (a) DSC heating curves recorded at a heating rate of 20 °C min<sup>-1</sup>. (b) Temperature dependences of loss factor, tan  $\delta$ , measured under 1 Hz. (c) Modulus mapping of ILNs measured by AFM



**Fig. S7** Homogeneity characterization of IPNs. (a) Light microscope images, (b) modulus mappings measured by AFM, (c) DSC heating curve recorded at a heating rate of 20 °C min<sup>-1</sup>, and (d) temperature dependence of loss factor, tan  $\delta$ , measured under 1 Hz of the simultaneous IPNs.

### 4. Forced miscibility of ILNs (in comparison with the phase-separated IPNs)

Fig. S7a shows that there is obvious regional inhomogeneity due to serious phase separation in the IPNs. The phase separation further leads to heterogeneous modulus distribution of the material (Fig. S7b).

The DSC (Fig. S7c) and DMA (Fig. S7d) curves show two glass transition temperatures, which agrees with the phase separation phenomenon. The results suggest that the IPNs are completely different from ILNs in essence, despite the fact that they are made by the same monomers.



## 5. Surface performances

**Fig. S8** Surface properties. (a, b) Photographs of water contact angles of (a1) glass, (a2) TPT, (b1) SU-PA, (b2) SN-PU, and (b3) ILNs. (c) Surface energies of glass, TPT, SN-PA and SN-PU. (d) Interfacial tensions of SN-PA/glass, SN-PU/glass, SN-PA/TPT, and SN-PU/TPT pairs.



**Fig. S9** Validation of self-stratification by Raman spectroscopy. Depth scanning Raman spectra of the ILNs in the glass/ILNs/TPT assembly collected from the glass side to the TPT side. Preparation conditions of the glass/ILNs/TPT assembly: hot-pressing temperature = 120 °C; hot-pressing pressure = 0.3 MPa; hot-pressing time = (a) 0 min, (b) 5 min, (c) 10 min, and (d) 15 min.

## 6. Self-stratification behaviors



**Fig. S10** Validation of self-stratification by XPS. (a) Low-resolution XPS spectra of the ILNs near glass and TPT. Note: The glass/ILNs/TPT assembly was hot-pressed at 120 °C under 0.3 MPa for 15 min in advance. Then, the sandwiched ILNs were peeled off and the surface close to the glass and that close to the TPT were measured by XPS, respectively. (b, c) Contents of the characteristic elements of (b) SN-PA and (c) SN-PU on both sides of the peeled ILNs. (d) High-resolution XPS O1s spectra of the ILNs near glass and TPT.

Based on the low-resolution XPS spectra of the ILNs near glass and TPT (Fig. S10a), B and O are chosen to represent SN-PA (there are more O in SN-PA than SN-PU according to their structures), and N and S act as the characteristic elements of SN-PU. As shown in Fig. S10b, the contents of O and B of the ILNs close to the glass side are higher than those close to the TPT side. Meantime, the contents of N and S have the opposite result (Fig. S10c). It is thus known that in the glass/ILNs/TPT assembly, SN-PA is enriched near the glass and SN-PU is concentrated near the TPT. Afterwards, the O1s peaks are split into three parts:<sup>9</sup> C=O, Zr<sup>4+</sup>-O and C-O, respectively (Fig. S10d). It is seen that on the surface separated from TPT, the peak area of C-O is larger than that of C=O, indicating that there are more SN-PU in the ILNs near the TPT (as SN-PU contains more C-O contributed by PTMG). Conversely, the peak area of C-O is obviously lower than that of C=O on the ILNs surface close to the glass (as SN-PA contains more C=O contributed by acrylate monomers). The analysis supports the above discussion about the self-stratification effect of the ILNs.



**Fig. S11** Rheological properties of ILNs. (a) Temperature dependences of storage and loss moduli measured at 1 Hz. (b) Complex viscosity of ILNs versus temperature measured at 1 Hz. (c) Frequency dependences of storage and loss moduli of ILNs measured at 120 °C. (d) Frequency dependences of storage and SN-PU measured at 120 °C.

Fig. S11a shows that the crossover of the storage modulus, G', and loss modulus, G'', of ILNs appears when temperature is raised to 82 °C, owing to the switch from elastic-like (G' > G'') to viscous-like (G' < G'') status at temperature higher than 82 °C. The reversible reactions of the built-in reversible boronic ester and disulfide bonds account for the variation as the disconnected networks need time to be reconstructed during metathesis. In this context, the hot-press of the glass/ILNs/TPT assembly at 120 °C is reasonable. The ILNs can be largely de-crosslinked and become liquid-like fluid under the circumstances, which benefits (i) wetting at the interfaces and (ii) stratification of the fragmented SN-PA and SN-PU. The

significant drop of the complex viscosity with a rise in temperature (Fig. S11b) confirms the analysis from another angle.

On the other hand, the flow transition relaxation time,<sup>10</sup>  $\tau_f$ , of ILNs, determined from the reciprocal frequency at the intersection of the frequency dependences of G' and G'' (Fig. S11c), is found to be 0.4 s at 120 °C. It falls between the values of SN-PA (0.25 s) and SN-PU (0.63 s) (Fig. S11d), which well agrees



**Fig. S12** Stratification effect of IPNs. (a, c) Typical depth scanning Raman spectra of different regions and (b, d) the corresponding relative intensities of the characteristic peaks of SN-PA and SN-PU of the IPNs isolated from the glass/IPNs/TPT assembly. The measurements are conducted from the glass side to the TPT side. Note: The preparation conditions of the glass/IPNs/TPT assembly are the same as the optimal ones applied for making the glass/ILNs/TPT assembly: hot-pressing temperature = 120 °C, hot-pressing time = 15 min and hot-pressing pressure = 0.3 MPa.

with the homogeneity of the ILNs.

There are two types of stratification of the IPNs isolated from the glass/IPNs/TPT assembly after hotpressing. That is, almost no stratification is detected in some regions, while slight stratification is found in other regions. As shown in Fig. S12a and b, all the characteristic peaks of either SN-PA or SN-PU are nearly unchanged along the scanning depth, which means that there is no stratification in this case. In contrast, the intensities of the characteristic peaks collected from the other region show the evidence of stratification (Fig. S12c and d). The peak intensity ratio of B-O rings/C-S to CH<sub>2</sub> of SN-PU regularly increases with decreasing the distance to TPT. Even so, the changes in the same peak intensity ratio of SN-PA are still too insignificant to summarize the trend. On the whole, the degree of stratification of IPNs is much lower than that of ILNs (Fig. 2b and c).

**Table S1** Tracer diffusion coefficients,  $D^*$ , of SN-PA and SN-PU in ILNs calculated from the depth scanning Raman spectra<sup>11,12</sup> (Fig. S9) of the ILNs hot-pressed between glass and TPT.

Hot-press time (min)	5	10	15
D* of SN-PA (cm <sup>2</sup> s <sup>-1</sup> )	4.45×10 <sup>-6</sup>	1.11×10 <sup>-6</sup>	5.74×10 <sup>-8</sup>
D* of SN-PU (cm <sup>2</sup> s <sup>-1</sup> )	5.53×10 <sup>-7</sup>	1.39×10 <sup>-9</sup>	4.27×10 <sup>-11</sup>

## 7. Adhesive and cohesive characteristics



**Fig. S13** Cohesive interactions. (a) FTIR spectra of SN-PA and ILNs. (b) FTIR spectra of SN-PU and SN-PU<sub>Zr free</sub>. (c) High-resolution XPS N1s spectra of SN-PU<sub>Zr free</sub> and SN-PU. (d) Raman spectra of ILNs and SN-PA. (e) High-resolution XPS O1s spectra of SN-PA and ILNs.

To understand the intra- and inter-macromolecular interaction in the self-stratified ILNs sandwiched between glass and TPT, FTIR, XPS and Raman spectra of original SN-PA, SN-PU and ILNs, which represent the SN-PA rich and SN-PU rich zones and the core part of the stratified ILNs, are collected in Fig. S13a-e, respectively. The carbonyl stretching at 1705 cm<sup>-1</sup> on the FTIR spectrum of SN-PA (Fig. S13a), which originates from the self-associated COOH dimers, is attributed to the intramolecular hydrogen bonds between carboxyl groups of polyacrylate, while the blue shift of the absorptions of N-H groups of ureido (at 3464 cm<sup>-1</sup>, 3343 cm<sup>-1</sup> and 1551 cm<sup>-1</sup>) of SN-PU with the addition of Zr<sup>4+</sup> (Fig. S13b) should be correlated to the coordination between Zr<sup>4+</sup> and ureido groups of SN-PU. The newly emerged Zr<sup>4+</sup>-N peak at 399.87 eV on the N1s spectrum of the SN-PU (Fig. S13c) testifies the interpretation. On the other hand, the FTIR spectrum of the original ILNs (Fig. S13a) implies that in the central zone of the self-stratified ILNs away from the interfacial regions, the carbonyl peak attributed to the intramolecular hydrogen bonds of SN-PA at 1705 cm<sup>-1</sup> is replaced by that representing the intermolecular hydrogen bonds between acid and ether at 1725 cm<sup>-1</sup> of ILNs. The intramolecular hydrogen bonds of SN-PA must have been broken to form intermolecular hydrogen bonds with the ether groups of SN-PU when the ILNs are produced. Furthermore, the symmetric stretching vibration of -COO<sup>-</sup> groups of SN-PA at 1394 cm<sup>-1</sup> is found to shift to 1402 cm<sup>-1</sup> on the ILNs' spectrum as a result of formation of the coordination bonds between  $Zr^{4+}$  and carboxyl, which coincides with the results of Raman (Fig. S13d) and XPS (Fig. S13e) spectroscopy study. The absorption of carboxyl groups of SN-PA shifts from 1705 cm<sup>-1</sup> to 1714 cm<sup>-1</sup> when SN-PA is involved in ILNs (Fig. S13d). Clearly, the intermolecular complexation between carboxyl of SN-PA and  $Zr^{4+}$  of SN-PU is built up. Meantime, the appearance of  $Zr^{4+}$ -O peak (531.68 eV) on the O1s spectrum (Fig. S13e) of ILNs is further evidence of the coordination bonds.



**Fig. S14** Adhesive interactions at the interfaces. (a, b) High-resolution XPS (a) O1s and (b) Si2p spectra of original glass and the mixture of SN-PA/glass. (c) FTIR spectra of SN-PA and SN-PA near glass. Note: To obtain the SN-PA near glass for the measurement, the assembly of glass/SN-PA/stainless steel wire cloth was hot-pressed at 120 °C under 0.3 MPa for 15 min. Then, the sandwiched SN-PA was peeled off and the surface near the glass was measured by FTIR in terms of ATR method. (d) High-resolution XPS O1s spectrum of the glass isolated from the glass/ILNs/TPT assembly. Note: The glass/ILNs/TPT assembly was hot-pressed at 120 °C under 0.3 MPa for 15 min. Then, the sandwiched ILNs were peeled off and the surface of remaining glass near ILNs was measured by XPS. (e) FTIR spectra of SN-PU and the mixture of SN-PU/TPT. (f) High-resolution XPS F1s spectra of original TPT and the TPT isolated from the glass/ILNs/TPT assembly. Note: The glass/ILNs/TPT assembly was hot-pressed at 120 °C under 0.3 MPa for 15 min. The glass/ILNs/TPT assembly was hot-pressed at 120 °C under 0.3 MPa for 15 min. Then, the sandwiched ILNs were peeled off and the surface of remaining glass near ILNs was f1s spectra of original TPT and the TPT isolated from the glass/ILNs/TPT assembly. Note: The glass/ILNs/TPT assembly was hot-pressed at 120 °C under 0.3 MPa for 15 min. Then, the sandwiched ILNs were peeled off remaining TPT near ILNs was measured by XPS.

Besides the investigation of the interfacial interaction between the SN-PA rich region of ILNs and glass using SN-PA as the substitution for the SN-PA rich region (Fig. S14a-c), the XPS O1s spectrum of the glass isolated from the glass/ILNs/TPT assembly is recorded for cross checking (Fig. S14d). It is found that the O1s peak is split into Si-O (532.70 eV), C=O (533.50 eV) and C-O (531.40 eV) under the circumstances. The presence of small amount of C=O and C-O indicates that a few ILNs are left on the glass after their separation. The Si-O peak of the original glass at 532.38 eV (Fig. S14a) moves to 532.70 eV (Fig. S14d) after being isolated from the glass/ILNs/TPT assembly, which resembles the case of SN-PA/glass mixture (Fig. S14a) and proves the existence of hydrogen bonds between the SN-PA rich region of ILNs and glass once

more. Moreover, unlike the result of the ILNs near glass (Fig. S10d), there is no peak of Zr<sup>4+</sup>-O on the O1s spectrum of the glass isolated from the glass/ILNs/TPT assembly (Fig. S14d). It means that the Zr<sup>4+</sup> in the SN-PA rich region of ILNs doesn't coordinate with hydroxyl of glass.

Similarly, the XPS F1s spectrum of the TPT isolated from the glass/ILNs/TPT assembly is also collected and compared to that of the original TPT (Fig. S14f). Like the F1s spectrum of the SN-PU/TPT mixture (Fig. S14c), the former consists of three components representing Zr<sup>4+</sup>-F, N-F and C-F, respectively. The peak of Zr<sup>4+</sup>-F demonstrates that the Zr<sup>4+</sup> has coordinated with the fluorine of TPT, while the N-F peak reflects the hydrogen bonds between the ureido groups of SN-PU of ILNs and fluorine of TPT. It is clear that the same conclusion can be concluded from the XPS results of the SN-PU/TPT mixture (Fig. S14c) and the TPT isolated from the glass/ILNs/TPT assembly (Fig. S14f). That is, both the envisaged Zr<sup>4+</sup>-F coordination and N-F hydrogen bonds between the SN-PU rich region of ILNs and TPT (Fig. 1b5) are established.



**Fig. S15** Mixed coordination interactions at the interface of TPT. (a, b, c) Optimized structures of the groups of (a) NHCONH, (b) COOH and (c)  $CF_2$  involved in the coordination of  $Zr^{4+}$ -N,  $Zr^{4+}$ -O and  $Zr^{4+}$ -F bonds. The dark grey, light grey, red, blue and purple spheres represent carbon, hydrogen, oxygen, nitrogen and fluorine, respectively. (d, e, f) Optimized structures of the coordination of (d)  $Zr^{4+}$ -N, (e)  $Zr^{4+}$ -O and (f)  $Zr^{4+}$ -F bonds for radial distribution function (RDF) calculation. The color coding of the spheres is the same as that of (a, b, c). (g) Bond length dependences of PCF (solid lines) and running coordination number (RCN, dotted lines) of  $Zr^{4+}$ -N,  $Zr^{4+}$ -O and  $Zr^{4+}$ -F bonds in the case of single coordination. (h, i) Bond length dependences of PCF (solid lines) and RCN (dotted lines) of  $Zr^{4+}$ -F bond in the case of mixed coordination.

To figure out the coordination situation, density functional theory (DFT) calculation and molucular dynamic (MD) simulation were utilized. The simplified characteristic groups (NHCONH, COOH, and CF<sub>2</sub>) were employed to represent the ligends in the adhesion system (Fig. S15a-c), which were then coordinated with Zr<sup>4+</sup> (Fig. S15d-f and Fig. 3d, e). As shown in Fig. S15g, the bond lengths of Zr<sup>4+</sup>-F, Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O coordination are 2.11, 2.41 and 2.27 Å, respectively, indicating that the Zr<sup>4+</sup>-F bond is the most stable one. In addition, the coordination numbers of Zr<sup>4+</sup>-F, Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O are 1.81, 6.57 and 2.93,

respectively. The results suggest that  $Zr^{4+}$  can be coordinate with N, O and F, while  $Zr^{4+}$  is more likely to coordinate with F under the same conditions.

It is worth noting that the above conclusion is made in the case of single coordination. Since multiple coordination of Zr<sup>4+</sup> coexists in the ILNs, when the latter serve as the Janus-like adhesive for gluing glass and TPT, the impact the Zr<sup>4+</sup>-F coordination bond receives should be considered. As shown in Fig. S15h and i, the bond lengths of Zr<sup>4+</sup>-F and Zr<sup>4+</sup>-N in the case of mixed Zr<sup>4+</sup>-F and Zr<sup>4+</sup>-N coordination are 2.13 and 2.37 Å, respectively, and those of Zr<sup>4+</sup>-F, Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O in the case of mixed Zr<sup>4+</sup>-F, Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O coordination are 2.13, 2.41 and 2.23 Å, respectively. The Zr<sup>4+</sup>-F coordination is still the most stable one, which ensures the interfacial bonding between the stratified ILNs and TPT. Accordingly, the coordination numbers of each ion pairs were estimated. There are 1.30 F and 3.7 N coordinated with Zr<sup>4+</sup>-O decrease to 0.66, 1.08 and 0.47 for the mixed Zr<sup>4+</sup>-F, Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O coordination. The calculation results coincide with the trend reflected by those of the single coordination, and show the importance of self-stratification again. The stronger coordination between Zr<sup>4+</sup> and F at the ILNs/TPT interface than that between Zr<sup>4+</sup> and N or O in the bulk ILNs benefits the adhesion to TPT.



**Fig. S16** Coordination interactions in ILNs. (a) Optimized structures for RDF calculations of the mixed  $Zr^{4+}$ -N and  $Zr^{4+}$ -O coordination. (b) Bond length dependences of PCF (solid lines) and RCN (dotted lines) of  $Zr^{4+}$ -F bond in the case of mixed coordination.

In addition to the Zr<sup>4+</sup>-F coordination at the ILNs/TPT interface, the Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O coordination is discussed to understand the coordination situations in the SN-PU rich zone (Fig. S15d) and ILNs cores (Fig. S16a). As shown in Fig. S15d, the coordination number of single Zr<sup>4+</sup>-N coordination is 6.57, which means about six nitrogen atoms are coordinated with Zr<sup>4+</sup> in the SN-PU rich layer. Similarly, the coordination numbers of Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O are 3.26 and 1.23 in the case of mixed Zr<sup>4+</sup>-N and Zr<sup>4+</sup>-O coordination (Fig. S16b), respectively, which means about three nitrogen atoms and one oxygen atom are coordinated with Zr<sup>4+</sup> in the interior of ILNs.



**Fig. S17** Adhesion properties. (a) Peeling strengths of the bonded pairs of SN-PA/glass, SN-PU<sub>Zr free</sub>/TPT and SN-PU/TPT. Note: The specimens of SN-PA/glass were prepared at room temperature and those of SN-PU<sub>Zr free</sub>/TPT and SN-PU/TPT were prepared by hot-pressing at 120 °C under 0.3 MPa for 15 min. (b) Peeling strengths and the sandwiched adhesive thickness of glass/ILNs/TPT assembly prepared under different hot-pressing temperatures but constant hot-pressing time of 15 min. (c) Peeling strengths of glass/ILNs/TPT assembly prepared under different hot-pressing times but constant hot-pressing temperature of 120 °C. Note: The pressure applied for making the specimens in (b, c) is 0.3 MPa.



**Fig. S18** Illustrations of the peeling tests and failure appearances of the samples. (a) 180° peeling test of glass/SN-PA/stainless steel mesh. (b) T-type peeling test of TPT/SN-PU/TPT assembly. (c) 180° peeling test of the assembly of glass/ILNs/TPT.



**Fig. S19** Macroscopic dissipation property. (a, b, c) Cyclic tensile loading/unloading curves of (a) SN-PA, (b) SN-PU and (c) ILNs.



**Fig. S20** Microscopic dissipation property. (a, b, c) Dissipation mappings of (a) SN-PA, (b) SN-PU and (c) ILNs measured by AFM. Note: The dissipation mapping determined by the QNM mode of AFM reflects the area of hysteresis between the tracing and retracing curves during each testing period. Therefore, the loss of mechanical energy (i.e. dissipation of energy) can be evaluated by the dissipation mapping image in a microscopic level. The gradient color scale on the right of each image gives quantitative description of the dissipation energy.



**Fig. S21** Determination of the failure region. (a, b) Raman spectra of (a) SN-PU and SN-PA, and (b) ILNs and the cohesively failed ILNs sampled from the glass/ILNs/PPT assembly after 180° peeling test. (c) Raman peak area ratios of S-S bond/B-O ring and S-S bond/B-O bond of ILNs and the cohesively failed ILNs calculated using the spectra of (b). Note: The bonded glass/ILNs/TPT assembly was tested to failure by 180° peeling test and then the exposed failure surface was measured by Raman spectroscope.



**Fig. S22** Failure features. (a) SN-PA (failure mode: adhesion failure on TPT, indicating the incompetent of SN-PA to bond TPT. The original cloth texture on the glass surface is covered by the failed SN-PA so tightly that it could not be seen at all). (b) IPNs (failure mode: mixed failure on both sides, indicating the inhomogeneous structure of IPNs). (c) EVA (failure mode: adhesion failure on TPT: indicating the necessity of surface pre-treatment). (d) 3M VHB tape (failure mode: adhesion failure on failure on glass).

When SN-PA is used for bonding glass and TPT, it is firmly sticked to glass but nothing is left on TPT after the peel test (Fig. S22a), indicating that SN-PA is not fit for bonding TPT. With respect to IPNs (Fig. S22b), a few remains of the adhesive can be found on both glass and TPT. This because the inhomogeneous microstructure of IPNs leads to uneven distribution of SN-PU and SN-PA. In the case of EVA, it is all on the glass after the measurement (Fig. S22c), which indicates that EVA is hard to realize the bonding of TPT without surface treatment of the latter. The failure of the assembly bonded by 3M VHB tape occurs on the surface of glass (Fig. S22d). It means that the adhesion strength to TPT is remarkably larger than that to glass and the adhesive tape cannot implement the bonding of glass and TPT simultaneously.



**Fig. S23** Effect of hot-press temperature. Peeling strength of the glass/TPT combination sticked by commercial EVA encapsulant at different hot-press temperatures, constant hot-press time of 15 min and constant hot-press pressure of 0.3 MPa.

Recycled glass	Recycled TPT

**Fig. S24** Effect of recycling. Photographs of the recycled glass (left) and TPT (right) after removing ILNs in the warm ethanol solution.

Table S2 Comparison of peeling strengths of glass and fluoropolymers bonded by different adhesives

Adherend	Adhesive	Adhesion type	Peeling strength (N/cm)	Reference
Glass	Polyacrylic acid/polyvinyl alcohol-borax hydrogel	Glass/Adhesive	0.0348	13
Glass	Cross-linked poly(dimethylsiloxane)	Glass/Adhesive	0.25	14
Glass	Acrylic acid, 2-methacryloyloxyethyl phosphorycholine, tannic acid hydrogel	Glass/Adhesive	2.48	15
Glass	Poly(thioctic acid-phytic acid)s pressure- sensitive adhesives	Glass/Adhesive	2.89	16
Glass	Poly((N-(3-aminopropyl)methacrylamide hydrochloride-co-N- [tris(hydroxymethyl)methyl]acrylamide))	Glass/Adhesive	4.22	17

#### adhesive hydrogel

Glass	Poly( <i>tert</i> -butyl acrylate, ethylene glycol dimethacrylate) innogel	Glass/Adhesive	4.80	18
Glass/PET	Poly(2-hydroxyethyl acrylate, 2-ethylhexyl acrylate) pressure sensitive adhesives	Glass/Adhesive/PET	0.45	19
Glass/PET	Poly(methacrylated lysine, acrylamide) hydrogel	Glass/Adhesive/PET	1.18	20
Glass/PET	Poly(2-ethyl hexylacrylate, methyl methacrylate, acrylic acid) pressure sensitive adhesives	Glass/Adhesive/PET	2.6	21
Glass/PET	UV-curable polyurethane	Glass/Adhesive/PET	3.12	22
Glass/PET	Bovine serum protein/poly acrylamide organohydrogels	Glass/Adhesive/PET	5.49	23
Glass/PET	Poly(ethyl acrylate, 1-ethynylcyclohexyl acrylate) elastomers	Glass/Adhesive/PET	16.21	24
Glass/PET	Poly(butyl acrylate, isobornyl acrylate) elastomers	Glass/Adhesive/PET	20.26	25
Glass/Cloth foil	Poly(acrylated adenine, acrylated thymine, acrylamide) adhesive hydrogel	Glass/Adhesive/Cloth foil	1.66	26
Glass/Cloth foil	Poly(acrylated adenine, acrylated uracil, acrylamide) hydrogel	Glass/Adhesive/Cloth foil	3.96	27
Glass/Textile	Polyacrylic acid-enhanced carboxymethyl cellulose hydrogel	Glass/Adhesive/Texile	0.196	28
Glass/PAAm hydrogel	Mussel-mimetic polyurethane glue solution	Glass/Adhesive/PAAm hydrogel	3.53	29
Glass/PU film	Hyperbranched polymer elastomer pressure sensitive adhesives	Glass/Adhesive/PU film	13.09	30
Glass/Inelastic tape	Polyacrylamide@ĸ- carrageenan hydrogel	Glass/Adhesive/Inelastic tape	14	31
Glass/Stainless- steel mesh	SN-PA	Glass/Adhesive/Stainless- steel mesh	33.87	This work
Teflon	Synthetic gecko tape	Teflon/Adhesive	0.075	32
Teflon	Polydimethylsiloxane-poly(methyl methacrylate) block copolymers blended with poly(methyl methacrylate)	Teflon/Adhesive	6.09	33
PTFE	Solvent-borne acrylic pressure sensitive adhesives	PTFE/Adhesive	0.31	34
PTFE	Poly(thioctic acid-phytic acid)s pressure- sensitive adhesives	PTFE/Adhesive	1.24	16
PTFE	Silicone pressure sensitive adhesives	PTFE/Adhesive	2.2	35
TPT (PVDF)	SN-PU	TPT (PVDF)/Adhesive	38.79	This work
PTFE/PET	Poly(methacrylated lysine, acrylamide) hydrogel	PTFE/Adhesive/PET	0.144	20
PTFE/PET	Bovine serum protein/poly acrylamide organohydrogels	PTFE/Adhesive/PET	4.89	23
Teflon/PET	(Styrene-isoprene-styrene)-based hot-melt	Teflon/Adhesive/PET	11.5	36

PTFE/Cloth foil	Poly(acrylated adenine, acrylated uracil, N,N'- methylene bis-acrylamide) adhesive hydrogels	PTFE/Adhesive/Cloth foil	1.88	37
PTFE/Cloth foil	Poly(acrylated adenine, acrylated thymine, acrylamide) adhesive hydrogel	PTFE/Adhesive/Cloth foil	2.24	26
PTFE/Cloth foil	Poly(acrylated adenine, acrylated uracil, acrylamide) hydrogel	PTFE/Adhesive/Cloth foil	3.49	27
PTFE/PAAm hydrogel	Mussel-mimetic polyurethane glue solution	PTFE/Adhesive/PAAm hydrogel	1.07	29
PTFE/PDMS	Tetrapodal ZnO	PTFE/Adhesive/PDMS	1.9	38
PTFE/PU	Undisclosed adhesive	PTFE/Adhesive/PU	2.12	39
PTFE/Cu	Polydopamine coating + polybutadiene adhesive	PTFE/Adhesive/Cu	4.20	40
PVDF/Pig abdominal wall	Polyurethane-based adhesives	PVDF/Adhesive/Pig abdominal wall	10.30	41
TPT/Glass	ILNs	TPT/Adhesive/Glass	64.86	This work

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