

Supplemental Material

In terms of the discrepancy of thresholds for acrylic polyurethane coatings, it is necessary to explore the effect of substrate on the surface. The high roughness combined with good adhesion of Y-0 specimen endows acrylic polyurethane coatings with a high laser complete removed threshold, as illustrated in Fig. S1. For 2024 aluminum substrate without anodizing treatment, adhesive failure is more likely to occur due to no mechanical association between the coatings and the smooth substrate causing a shedding layering or stripping at the interface [1]. However, good adhesion strength at the coating-substrate interface hinders the laser removal process. Fig. S1 shows typical normal and frictional forces and penetration depth as a function of scratch track length during scratch testing. As the normal forces increase, the frictional forces and penetration depth increase accordingly and at a maximum. It is apparent from Fig. S1(a) that as the applied normal force increases above 5.31 N, a more rapid rate of increase in the penetration depth and the friction coefficient is observed. And the penetration depth reaches a value of $\sim 10\text{ }\mu\text{m}$ at a normal force of 5.31 N, representing that the coatings are almost penetrated by the indenter. Fig. S1(b) explains that the penetrated force of substrate with anodized treatment is 9.80 N, with $\sim 18\text{ }\mu\text{m}$ penetration depth with a narrower width. From the optical image of the scratch track, the width of the scratch track increases with the lateral displacement of the indenter and at a normal load of 5.31 N and 9.80 N, the superficial coating begins to peel at both sides of the scratch track. That suggests the critical load (L_c) for acrylic polyurethane coatings on different substrates is about 5.31 N and 9.80 N, respectively. The L_c value of the coating on an anodized treatment substrate is 84.55% larger than untreated substrates under identical experimental conditions, which means that Y-0 specimen has excellent adhesion.

A rougher surface increases energy absorption and thus is in need of less energy density of the laser. Simultaneously, good adhesion tends to attach to the substrates, leading to damage while the ablation mechanism is dominant. Therefore, in order to ensure surface quality, it is necessary to adjust the laser removal process to enhance the vibration effect.

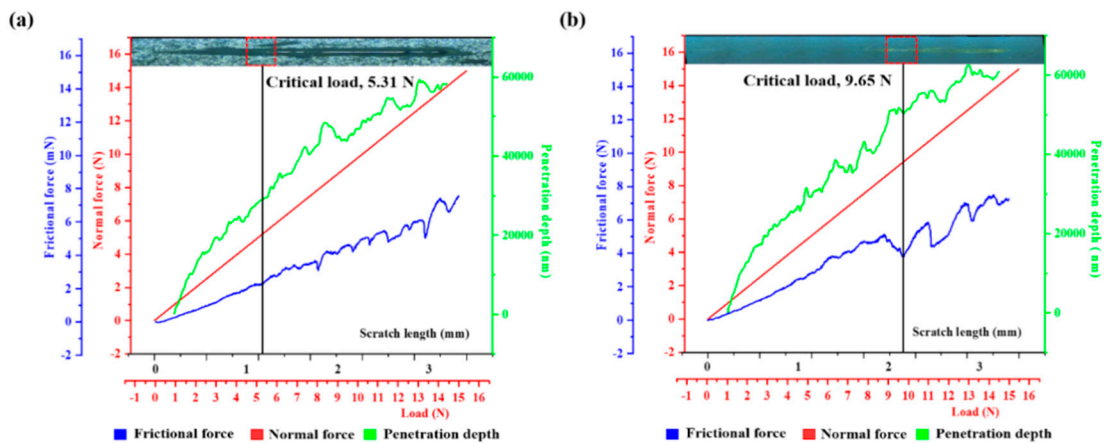


Figure S1. Micro-scratch test of acrylic polyurethane coatings on different substrates: (a) 2024 aluminum alloy substrate, (b) anodized treated 2024 aluminum alloy substrate.

The microscopic reaction principle in laser removal needs to be further analyzed through XPS results. When the specimen is cleaned in the different laser fluence as well as overlapping ratios, smaller differences are found in the results of some XPS spectrum. The relative contents of C1s and O1s change the most obvious to illustrate the laser removal mechanism during laser treatment. High-resolution spectra of C1s and O1s are presented in Fig. S2, representing the variances in the surface composition. Fig. S2(a, b) pre-

sents the spectral feature peaks of C1s and O1s of as-received acrylic polyurethane coatings. C-N and C-O function groups are sharply eliminated after laser treatment, as explained in Fig. S2(b, d, e). Table S1 indicates that the bond energy of C-O is 3.71 eV, a little higher than C-N (3.16 eV), which means the energy of the photon is more than 3.71 eV in laser removal technology [2]. From Fig. S2(b), C1s spectrum, the C=C has been left, and the C=O spectrum is of insufficient binding energy. Although the bond energy of C=O is 7.46 eV, the energetic photons break a large proportion of the functional groups during enough power of a single pulse. The oxides on the surface are lost by evaporation, and the rest of the bonds C-O are broken by the high-energy laser due to their low binding energy, as Fig. S2(d) displays. And as shown in Fig. S2(e, g), the C1s are with the same functional groups in different laser intensities. This is due to the same substrates, and the variance in laser fluence is very subtle so the photoelectrons have almost the same energy. Nevertheless, the peak difference in the O1s spectrum lies in its surface oxide, both Y-1 surface and N-1 surface samples form thin layers of alumina, while the surface oxide composition of Y-2 is mainly newly generated titanium dioxide. However, Fig. S2(d, f) explains that the peak intensity of O1s in N-1 sample is 30000 higher than that of Y-1. It means that the generated Al_2O_3 layer in Y-1 sample is thinner further proving the protective effect of the original anodic oxide films.

In this process, the substrate does not affect the molecular perturbation, so the high-resolution spectra C1s of N-1 Y-2 samples are the same in Fig. S2(c, e, g). And the difference between O1s is related to the performance of the substrates in Fig. S2(d, f, h).

Table S1. The binding energy of the molecule bond.

Type of bond	Bond energy (eV)
C-H	4.28
C=C	6.24
C-C	3.59
C-O	3.71
C-N	3.16
C=O	7.46

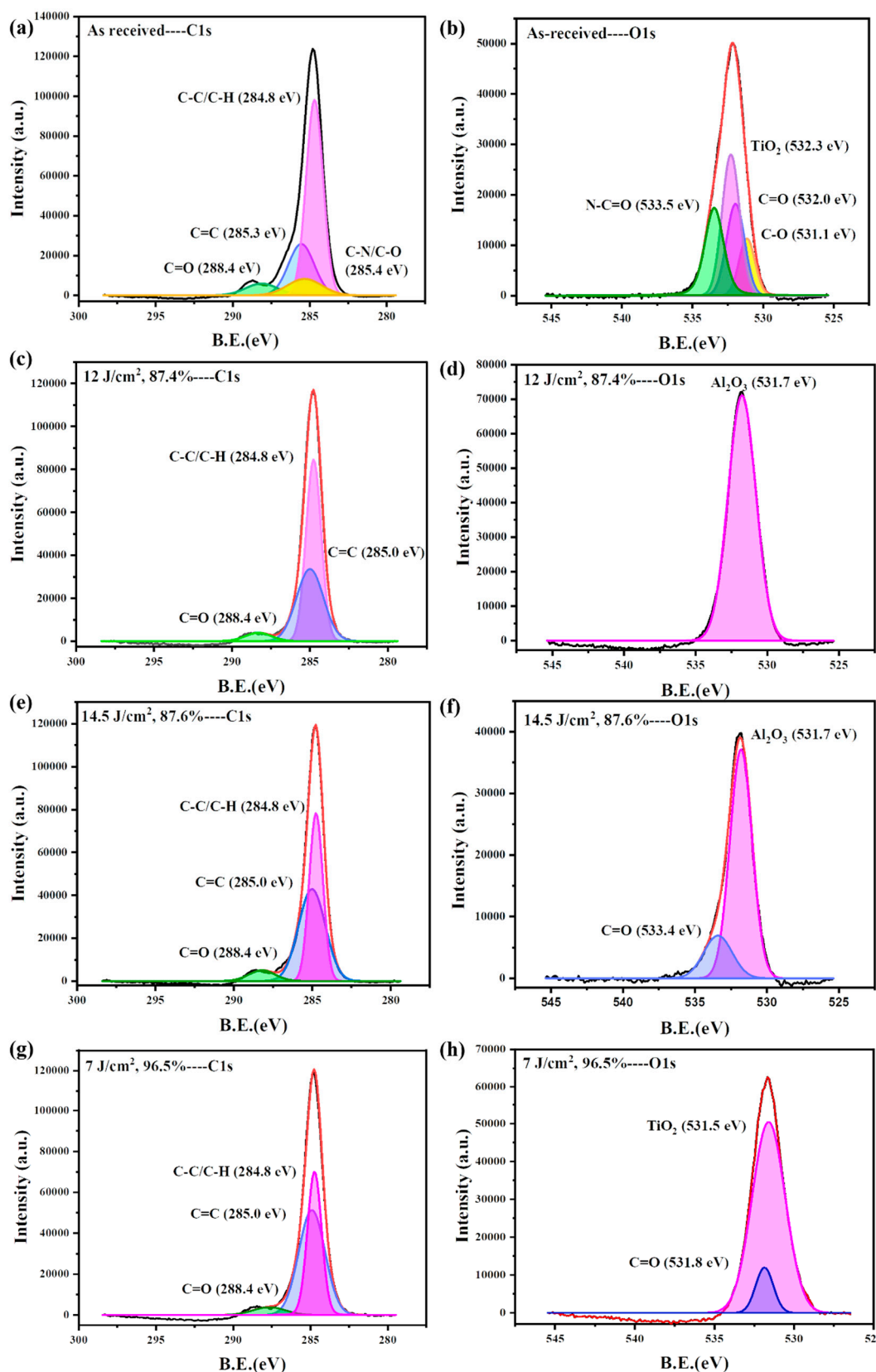


Figure S2. High-resolution spectra C1s of specimens: (a) as received, (c) N-1, (e) Y-1, (g) Y-2 and O1s of specimens: (b) as received, (d) N-1, (f) Y-1, (h) Y-2.

References

1. Miguel, M C; Thomas H; , N. Nina, Turning industrial paints superhydrophobic via femtosecond laser surface hierarchical structuring, *Prog. Org. Coat.* **2021**, 163, 106625.

2. Shakhawath, H; Salimullah, S; Mohammad, F. Ultra-high birefringent, highly nonlinear Ge₂₀Sb₁₅Se₆₅ chalcogenide glass photonic crystal fiber with zero dispersion wavelength for mid-infrared applications, *Optik*. **2020**, 225,165753.