

Supporting Information for *Materials Horizons*

‘Pressure-Sensitive Adhesive Powder’

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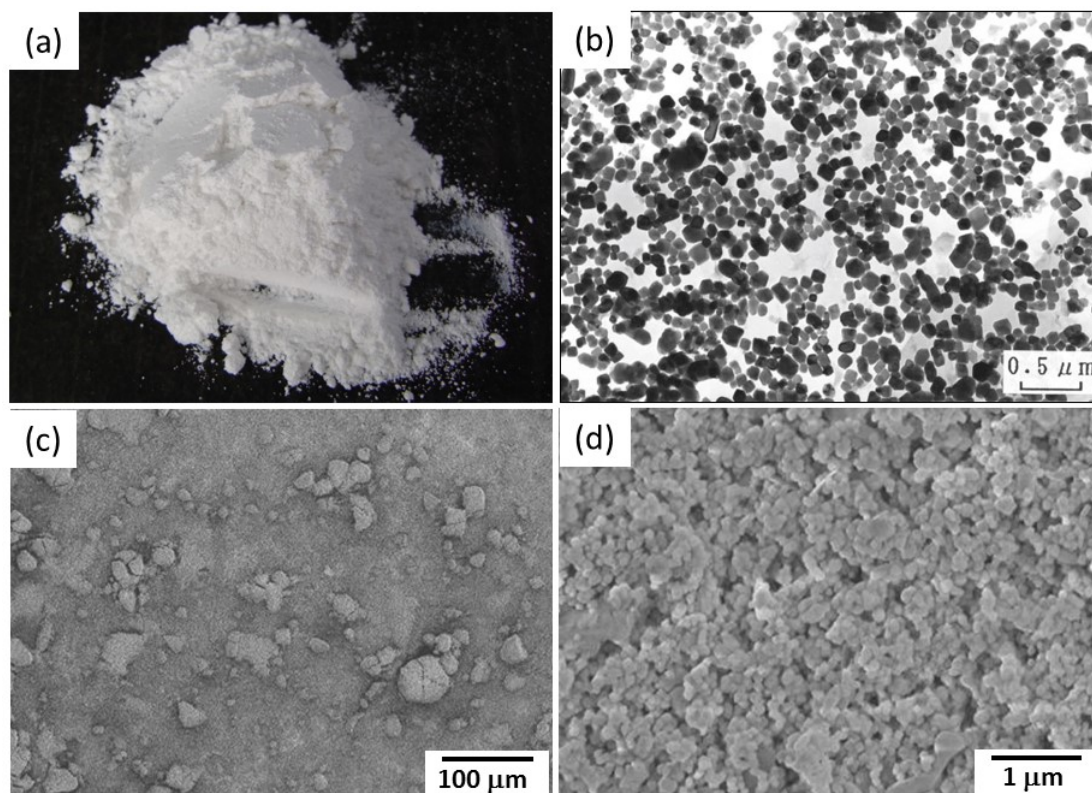
*5-16-1, Omiya, Asahi-ku, Osaka, 535-8585, Japan*

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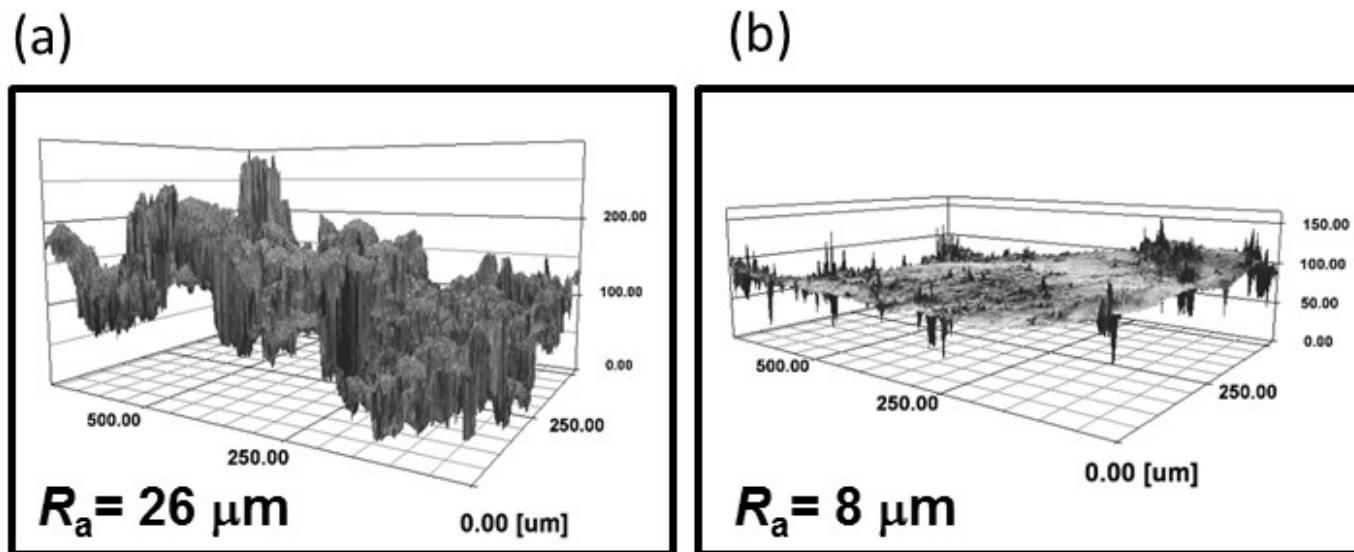
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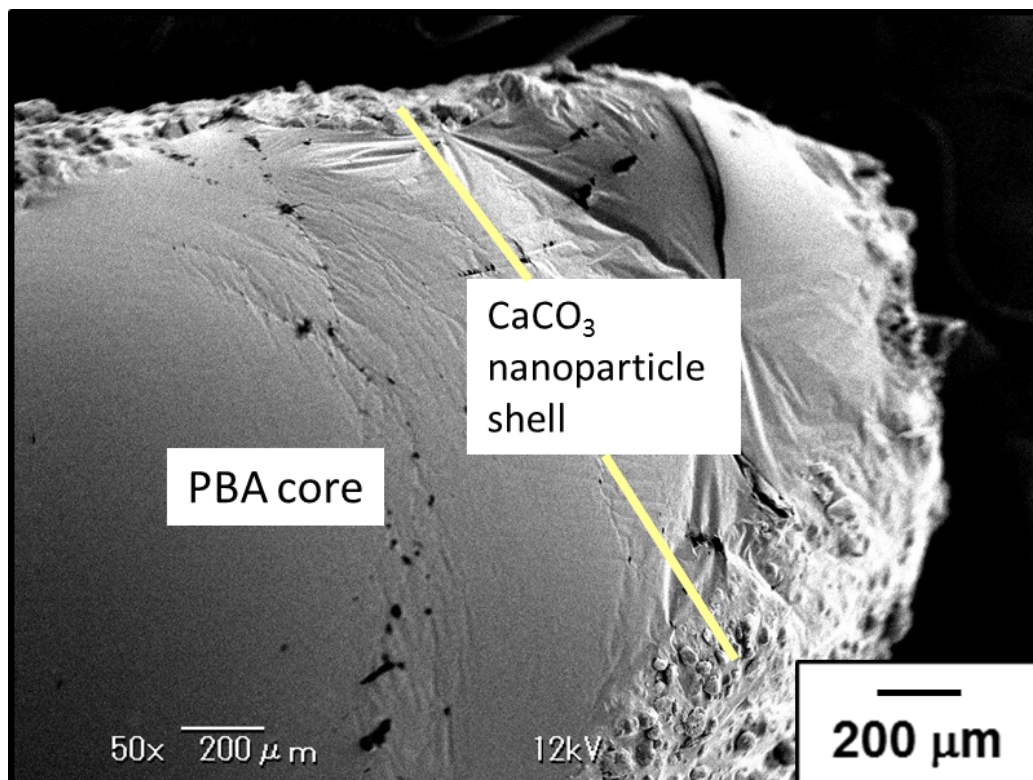
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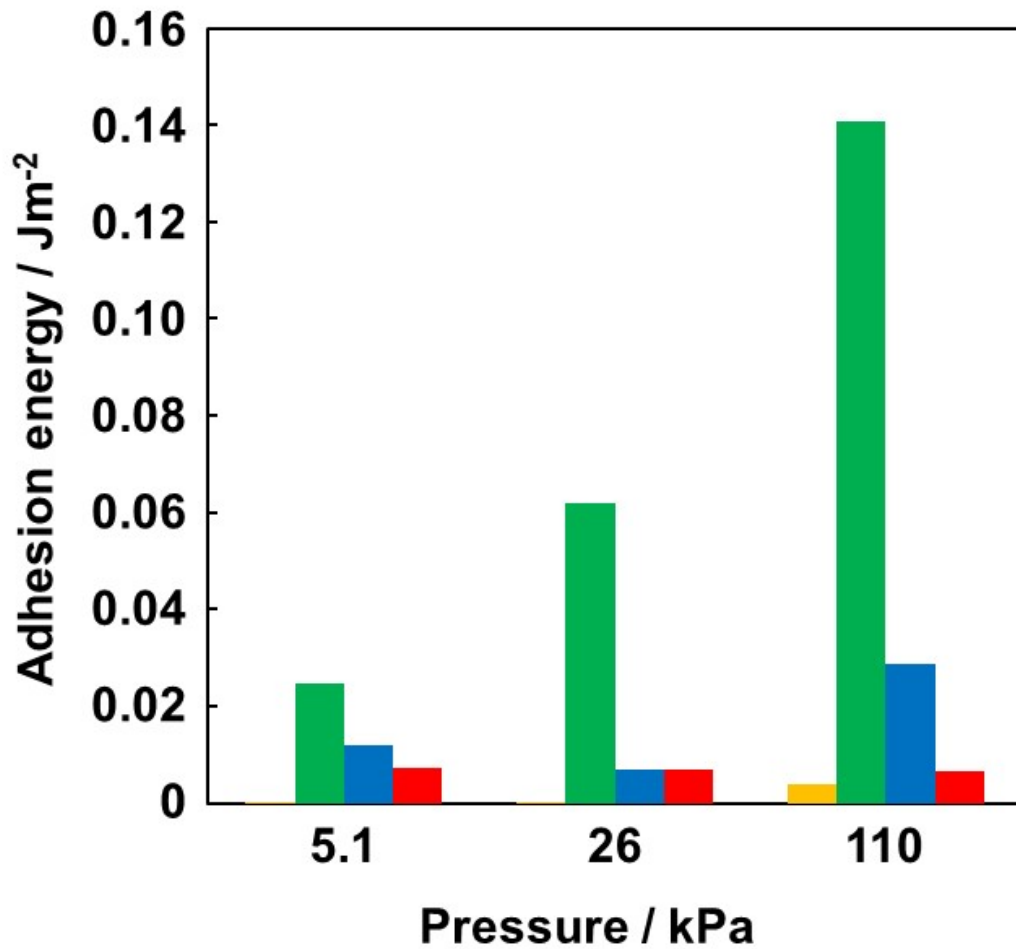
**Figure S1.** (a) Digital photograph, (b) transmission electron microscope and (c,d) scanning electron microscope images of CaCO<sub>3</sub> nanoparticles. Fig. S1(d) is a magnified image of Fig. S1(c).



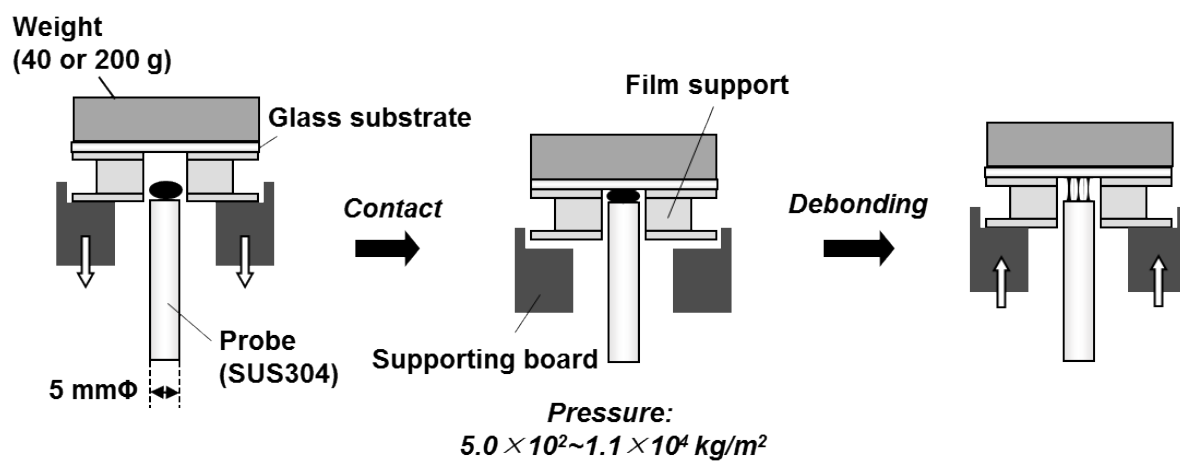
**Figure S2.** Laser microscope images of a particle with a soft adhesive PBA core and a  $\text{CaCO}_3$  hard nanoparticle shell morphology (a) before and (b) after application of shear stress. Laser microscope studies were conducted for the area of  $700 \mu\text{m} \times 500 \mu\text{m}$  and surface roughness ( $R_a$ ) of the adhesive materials are calculated using the Keyence software.



**Figure S3.** Scanning electron microscope cross-section image of cryo-fractured CaCO<sub>3</sub> nanoparticle-coated PBA particle.



**Figure S4.** Relationship between pressure applied to PSA materials and adhesion energy in tack measurement. Samples: Liquid marble PSA (yellow bar) before and (green bar) after application of shear stress, (blue bar) PBA latex film with a thickness of 45  $\mu\text{m}$  and (red bar) commercially available PSA tape (Scotch® Magic™ Tape 810)



**Figure S5.** Schematic illustrations of the test procedure and measurement component of the probe tack tester.

### *Gel-permeation chromatography (GPC)*

GPC measurements were performed using an RI detector equipped with three TSKgel MultiporeHXL-M columns (Tosoh Co.) and a TSKGuardcolumn MP(XL) column (Tosoh Co.) at 5.3 MPa. Tetrahydrofuran was used as the eluent at a flow rate of 1.0 mL/min at 40 °C.  $M_w$ (GPC) of the sample polymer was calibrated with polystyrene and poly(methyl methacrylate) standards.

### *Transmittance measurements*

Light transmittance percentages of dried adhesive liquid marbles before and after application of shear stress were recorded using a Shimadzu UV-1600 spectrophotometer with two 38- $\mu$ m thick PET films (Lintec Co., Japan) as a reference. A dried adhesive liquid marble before and after application of shear stress was put between the PET films and pressed using a press (MP-WNL250, Toyoseiki, Japan) at room temperature to obtain a mean adhesive film thickness of 0.5 mm, which was determined with a dial thickness gauge (H-MT, Ozaki Mfg. Co., Ltd). Transmittance% values were determined at 500 nm.

### *Pycnometer*

The solid-state density of the dried CaCO<sub>3</sub> nanoparticles was determined by helium pycnometry using a Micromeritics Accu Pyc II 1340 instrument.

### *3D laser scanning microscopy*

Laser microscope studies were conducted using VK-9510 (Keyence, Japan) for the area of 700 mm $\times$ 500 mm and surface roughness ( $R_a$ ) of the adhesive materials are calculated using the Keyence software.

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