ADVANCED MATERIALS

Supporting Information

for *Adv. Mater.*, DOI: 10.1002/adma.201104010

Bonding Assembled Colloids without Loss of Colloidal Stability

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Submitted to ADVANCED MATERIALS

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Glass transition temperature measurements:

The glass transition temperature of PMMA latex particles was measured with differential scanning calorimetry (HP DSC827e, Mettler-Toledo, USA). Prior to the measurements, the temperature and the heat flow were calibrated using certified indium and zinc samples. Samples were prepared in aluminum pans at room temperature. The samples were then hermetically sealed in aluminum pans to avoid any drying effects during measurements. Subsequently, masses of the particles were determined. 15.34 mg of dry particles, 16.72 mg of freshly *(*5 mins after the sample preparation, 42 wt%) prepared PMMA particles in CHB, and 16.22 mg of aged (7-8 days after the sample preparation, 42 wt%) PMMA particles were used for the measurements. Figure S1 shows differential scanning calorimetry data for 2.30 μm sized sterically stabilized PMMA particles heated at 10 ºC/min. The decrease in heat capacity at T_g exhibits overshoot typical of aged glasses, followed by a small exothermic peak. The glass transition temperature was taken as the middle of the base line shift. The glass transition temperatures of the dry, the freshly prepared dispersion of PMMA particles in CHB, and the aged dispersion of PMMA particles in CHB were 144.5 ºC, 142.5 ºC, 137.8 ºC respectively as seen in Figure S1.

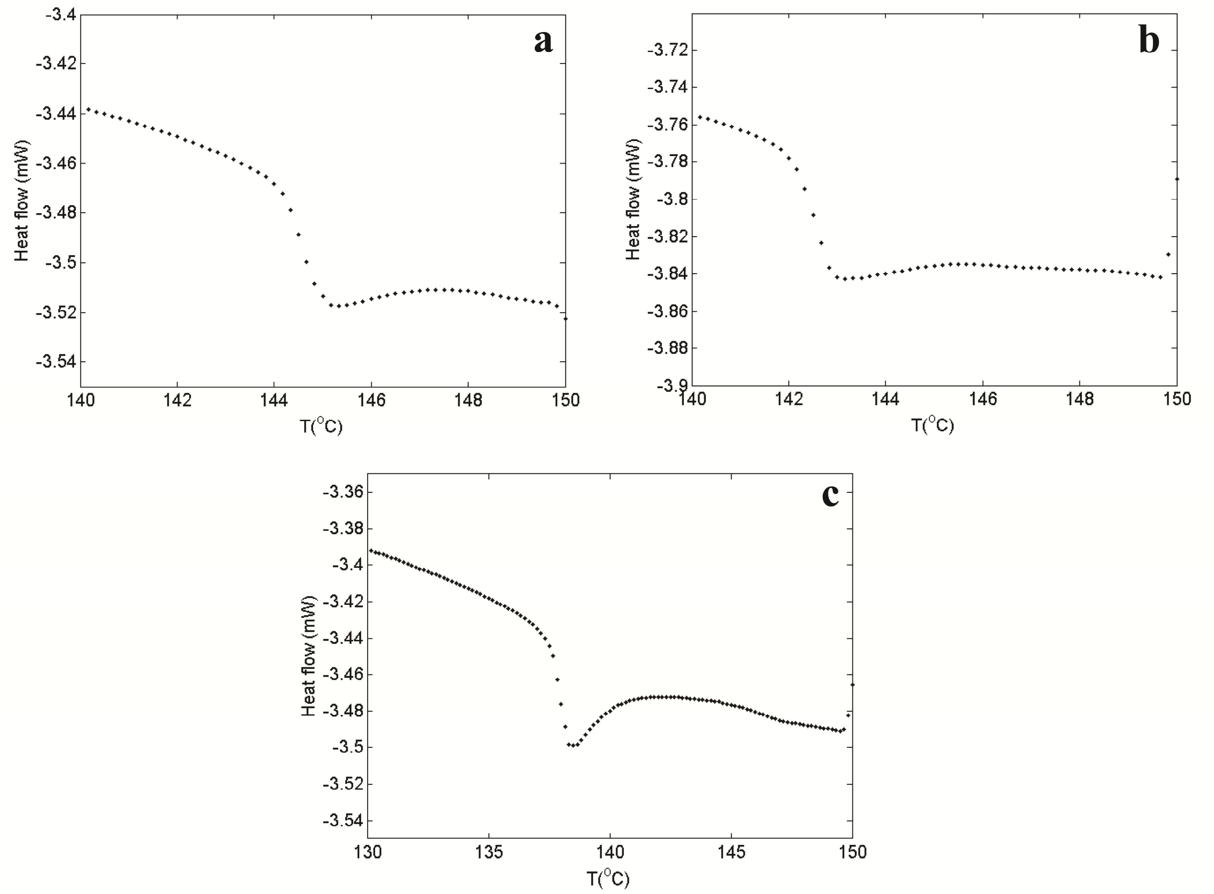

Figure S1. Differential scanning calorimetric diagrams of 2.30 µm sized sterically stabilized PMMA particles heated at 10 ºC/min. **a**, Dry PMMA particles. **b-c**, PMMA particles in cyclohexyl bromide (CHB) after different waiting times. **b,** 5 mins after the sample preparation. **c,** 7-8 days after the sample preparation.

Colloidal stability of bonded structure:

Here, we demonstrate that the permanently bonded and dried structures might be useful either as seeds for crystal nucleation and growth studies or for a successive self-assembly process. Permanently bonded and dried close-packed structures were first cut into small pieces with a sharp razor blade. Subsequently, these pieces were dispersed into a suspension of host particles (18 % by volume of 2.05 μm particles in salt saturated CHB). Note that the size of the host particles was chosen such that exactly one particle could fit in between neighboring seed particles as can be seen in Figure S2. This dispersion was then transferred to a 0.1 X 2.0

mm capillary cell and was studied with confocal microscopy. After 2 - 3 hrs, colloidal crystals of the host-particles were observed around the permanently bonded seed-structures (Figure S2**,** see also supplementary movie S1). We note that the orientation of host particles is different from the bonded seed-structure. Therefore this bonded structure acted as an impurity rather than a seed. However, we believe that by carefully tuning suspension parameters (e.g. particle diameter and Debye screening length) bonded structures can be used for nucleation and growth studies.

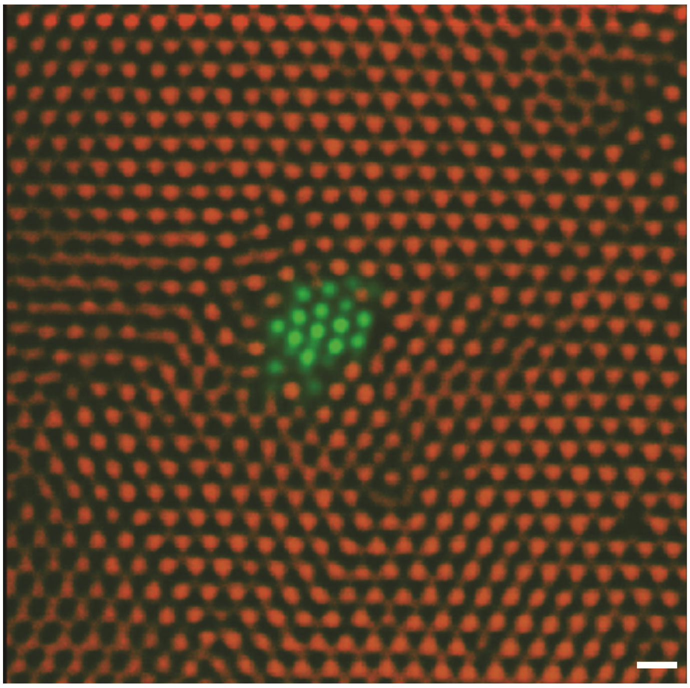

Figure S2. Colloidal stability of bonded structure. Confocal micrograph clearly reveals the permanently bonded particles (2.35 μm particles, green color) acted as an impurity in a dispersion of host particles (2.05 μm particles in CHB, red color). Scale bar is 5 μm.

Supplementary Movie S1. This movie illustrates the colloidal stability of permanently bonded structures (green color) and their second self-assembly in the sea of a different size particles (red color). The movie is composed of a stack of confocal micrographs (*xy-z*) and played at 2.1 μm/s.