Supplementary Data 1

Gold particles were attached to the original surface of zeolite A to distinguish the original from the cleaved surfaces. At first, 0.01g of zeolite A and 0.1 ml of a solution of gold particles in ethanol (0.05wt%, ca. 5 nm, Nippon Paint Co., Ltd.) were mixed. Then, the mixture was dried in an oven at 323 K for 24 hours. Schematic drawings of zeolite A with gold particles are shown in Figure S1. It is possible to distinguish the original from the cleaved surface. An FE-SEM image of the surface of zeolite A coated with gold particles is shown in Figure S2. Gold particles are well dispersed on the surface of zeolite A. The crystals recovered were crushed, and fixed on a Cu mesh grid before the observation by HRTEM.



Figure S1. Schematic drawings of zeolite A with gold particles.



Figure S2. FE-SEM image of the surface of zeolite A coated with gold particles.

Supplementary Data 2



- (a) HRTEM image of the internal structure of zeolite A taken from the [100] direction. We successfully observed a very clear HRTEM image. A schematic of the zeolite A is inserted in the lower part of the image.
- (b) Signal noise of the TEM photograph was removed by image calibration using the FFT method.
- (c) Computed image of the HRTEM image as derived by multislice calculations. A perfect match to (b) can be seen, indicating that it is possible to confirm the structure of zeolite A directly, and that it consists of sodalite cages connected with double four-membered rings (D4Rs).

Supplementary Data 3



Low magnification image of the sample. Cleaved surface has no gold particles and is rougher than an original one.