**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.](image)

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