**Supplementary Information**

Structural rearrangements of Ru nanoparticles supported on carbon nanotubes under microwave irradiation

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**Experimental Section**

Ruthenium nanoparticles supported on carbon nanotubes (Ru/CNTs) were synthesized by a microwave thermolysis method. Ru₃(CO)₁₂ was mixed together with CNTs and grinded for 20 minutes. Argon was flowed on the mixture for 2 hours at a flow rate of 100 mL/min, followed by microwave treatment at a quartz-tube reactor with a frequency of 2.45 GHz (the power is 800 W) as the radiation source for 1, 3, and 5 min. Samples were then taken out after cooling to room temperature. The loading content of Ru is 5.0 wt%. They were named as Ru/CNTs-1min, Ru/CNTs-3min, and Ru/CNTs-5min, respectively.

X-ray diffraction (XRD) analysis of the CNTs and Ru/CNTs samples were carried out using a Rigaku D/Max-RB diffractometer with Cu $K_a$ monochromatized radiation
source (\(\lambda=1.54178\) Å) and a D8 Advance theta/theta diffractometer (Bruker AXS) with Cu \(K_{a1+2}\) radiation (\(\lambda=1.54186\) Å), respectively. A FEI Cs-corrected Titan 80-300 microscope operated at 300 kV was employed to conduct structural investigations of Ru/CNTs samples by using TEM, HRTEM, and STEM modes. HRTEM simulation was performed by using JEMS software and a supercell approach for the single crystal Ru nanoparticle (copyright P. A. Stadelmann, EPFL, Switzerland). The simulations of particle shapes were conducted by combing Atom software and HRTEM simulation. The Ru/CNT samples were ultrasonically dispersed in ethanol, and then a drop of the solution was deposited on a holey C/Cu TEM grid to be used for HRTEM and high-angle annular dark-field (HAADF)-STEM characterization.

**Fig. S1** HRTEM images of Ru/CNTs-5min sample (a, c) together with corresponding shapes with faceting configurations (b, d). Insets in Figs. S1a and S1c are the local FFT of the HRTEM images.
Fig. S2 The frequency of single crystal Ru nanoparticles against particle size for Ru/CNTs-1min (a), Ru/CNTs-3min (b), and Ru/CNTs-5min (c) samples.

Table S1 Measurement of central tendency (particle number, mean, median, and standard deviation) for Ru/CNTs-1min, Ru/CNTs-3min, and Ru/CNTs-5min samples by using STEM images.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Particle number</th>
<th>Mean (nm)</th>
<th>Median (nm)</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ru/CNTs-1min</td>
<td>673</td>
<td>1.8</td>
<td>2.2</td>
<td>0.8</td>
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<tr>
<td>Ru/CNTs-3min</td>
<td>683</td>
<td>2.3</td>
<td>2.3</td>
<td>1.1</td>
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<tr>
<td>Ru/CNTs-5min</td>
<td>675</td>
<td>2.6</td>
<td>2.3</td>
<td>1.5</td>
</tr>
</tbody>
</table>