

Fig. 1. Controlling the Pd content and thickness in Pd-Ru thin films. (a) ALD sequence including  $m \times [\text{Pd}(\text{hfac})_2 / \text{RuO}_4 / \text{H}_2^* \text{ plasma}]$  and  $(20-m) \times [\text{Pd}(\text{hfac})_2 / \text{H}_2^* \text{ plasma}]$ ,  $m = 1, 2, 4, 10, 20$ , (b) Sequence variation allows to control the Pd content in Pd-Ru thin films, ranging from  $\text{Ru}_{10}\text{Pd}_1$  to  $\text{Ru}_1\text{Pd}_1$  atomic stoichiometries, for which linear growth curves are obtained. In addition, while the  $\text{Pd}(\text{hfac})_2 / \text{H}_2^*$  process gives rise to very long incubation times ( $> 100$  cycles), introducing a  $\text{RuO}_4$  pulse every 20 ALD cycles, already leads to a swift nucleation of bimetallic thin film growth.

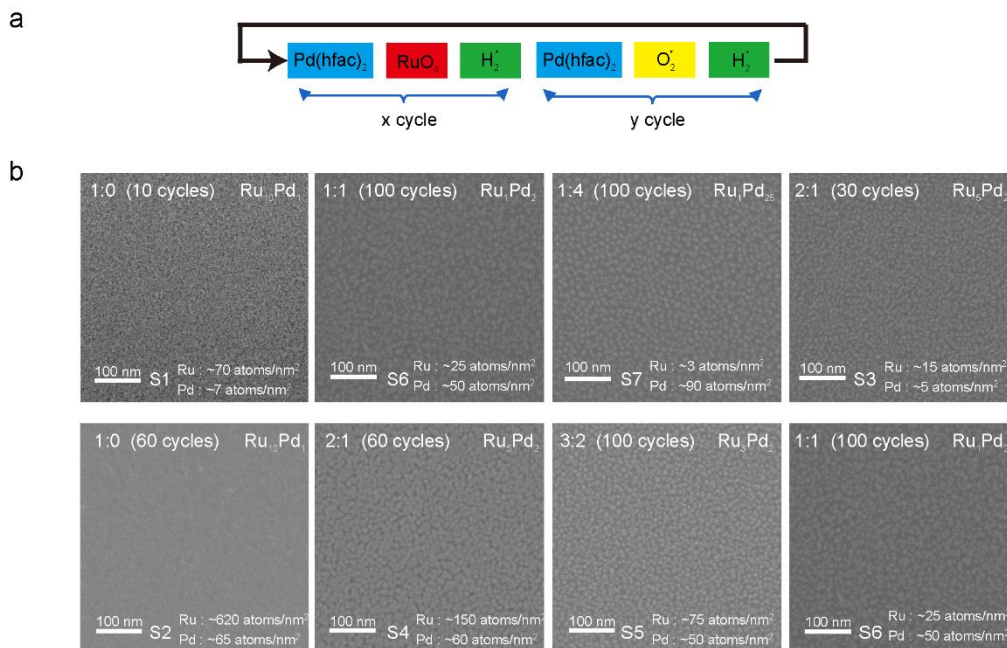


Fig. 2. (a) By replacing part of the  $\text{RuO}_4$  units in the three-step process by  $\text{O}_2^*$ , yielding  $x [\text{Pd}(\text{hfac})_2 / \text{RuO}_4 / \text{H}_2^* \text{ plasma}]$  and  $y [\text{Pd}(\text{hfac})_2 / \text{O}_2 / \text{H}_2^* \text{ plasma}]$  cycles, (b) the morphology of the Pd-Ru is transformed from thin films to bimetallic NPs, as observed from scanning electron microscopy (SEM) images. The top left ratios in each image corresponds to the  $x:y$  ratio in (a).