

## Electronic Supplementary Information for

### Solid-solution alloying of immiscible metals at a nanoscale: Ir and Au

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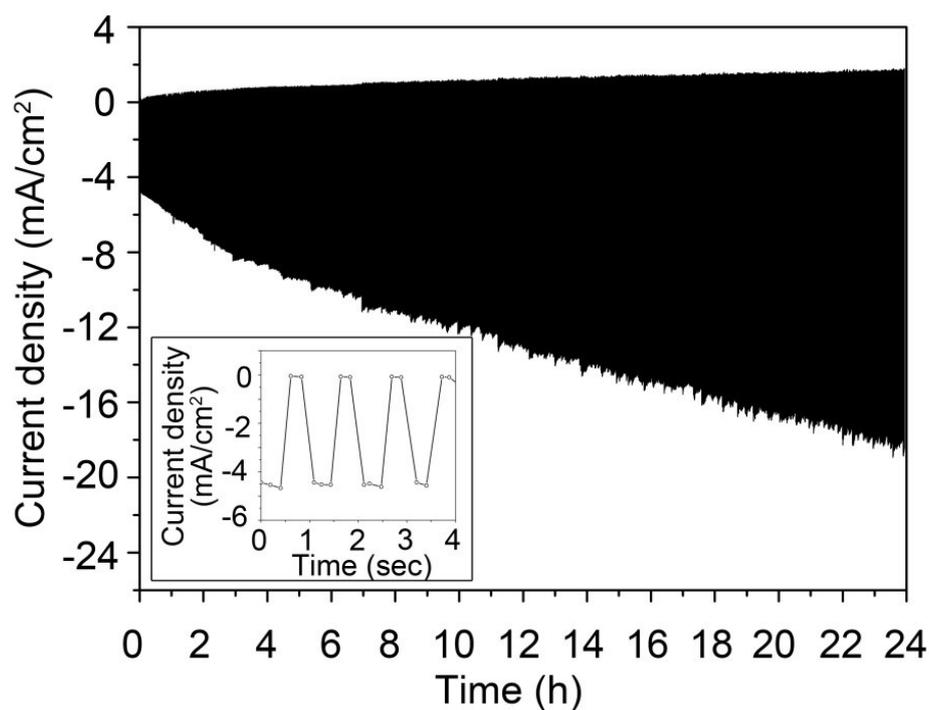


Figure S1. Variation of current density with time under a reverse-pulse potentiodynamic mode (1Hz,  $V_R = 10$  V and  $V_O = 0$  V for 24 hr) during the growth of Ir-Au alloys on a Au-coated substrate. The inset indicates the enlarged variation of current density at the initial state.

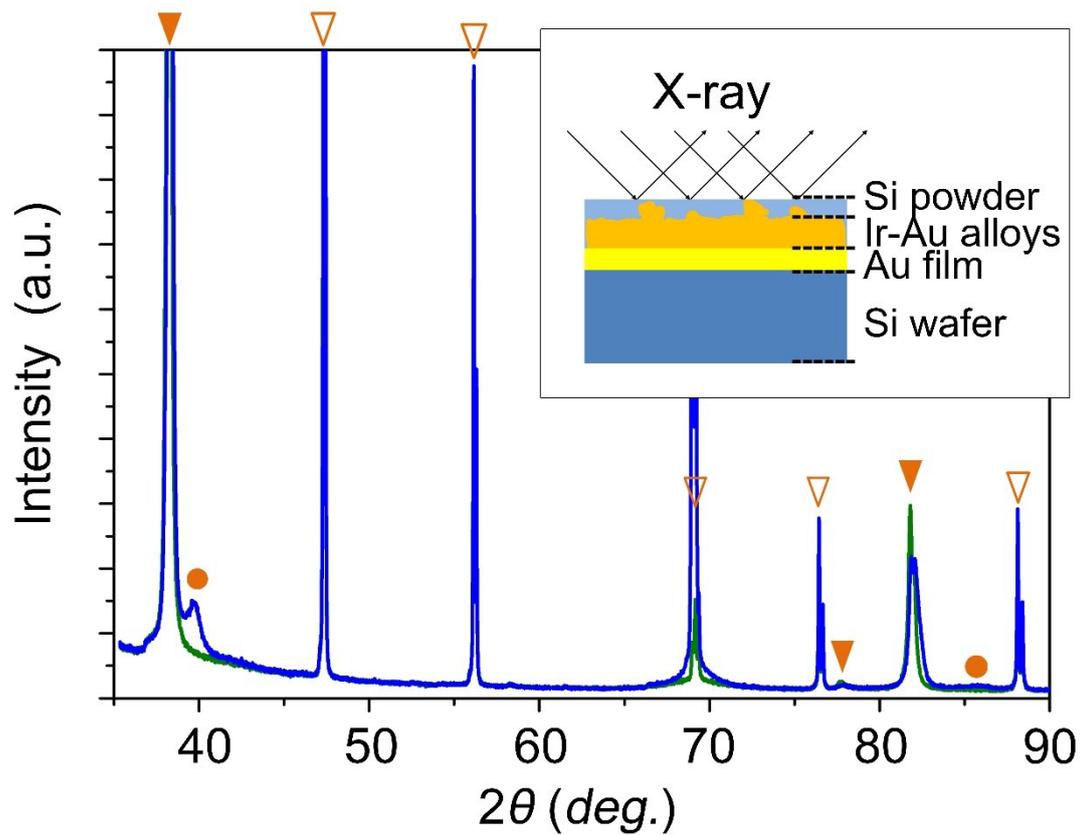


Figure S2. XRD patterns in the  $2\theta$  range  $35\text{-}90^\circ$  of Ir-Au alloys (blue line) and the Au-coated Si substrate as a substrate (green line). The inset denotes the schematic for preparing XRD samples. The solid and open upside-down triangles indicate Au film and Si powder, respectively. The solid circles denote the nanostructured Ir-Au alloys.

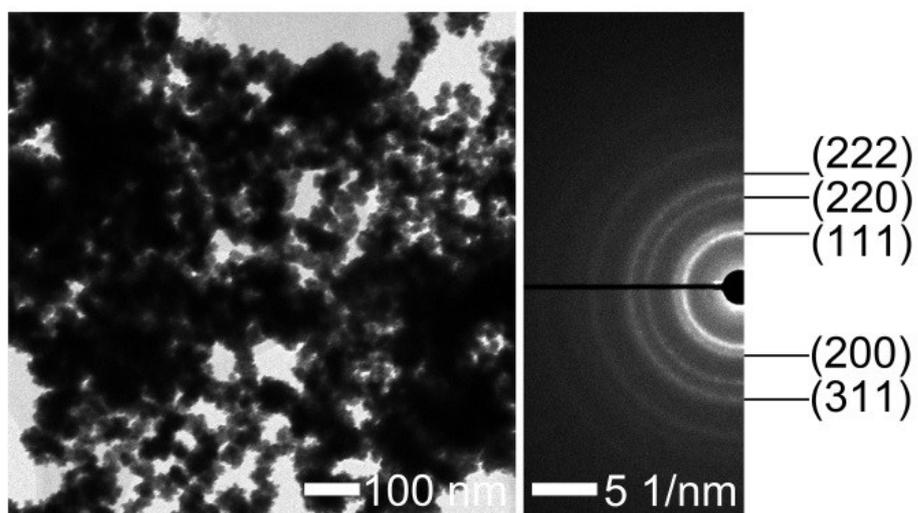


Figure S3. BFTEM image and electron diffraction pattern of Ir-Au alloy after detaching from Au substrate.

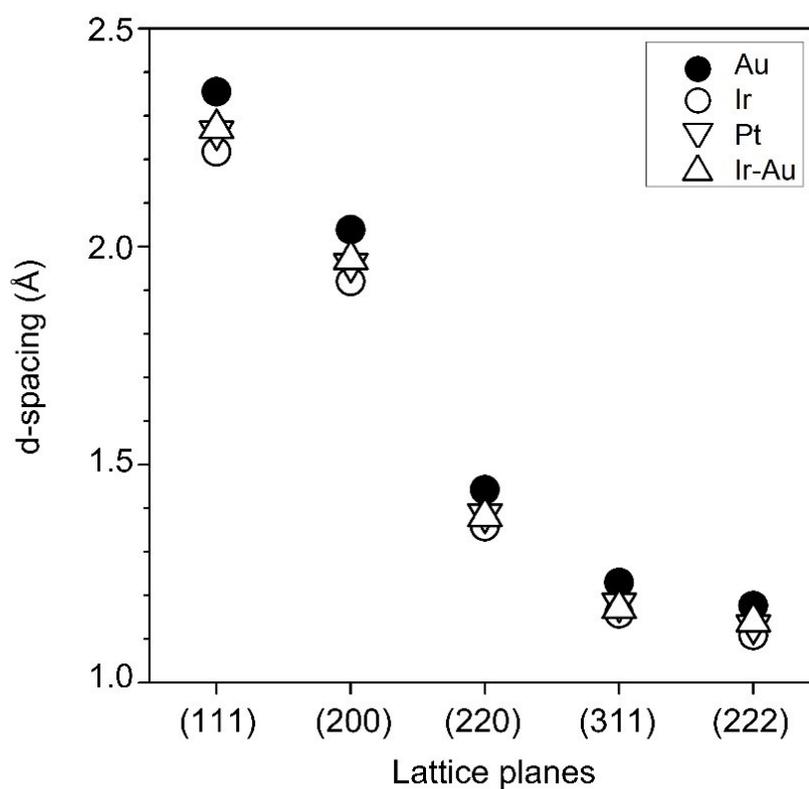


Figure S4. Variation of d-spacing with lattice planes (hkl). The d-spacing of Au (solid circles), Ir (open circles) and Pt (open upside-down triangles) come from JCPDS cards. The d-spacings of Ir-Au alloys (open triangles) were measured by TEM analyses.

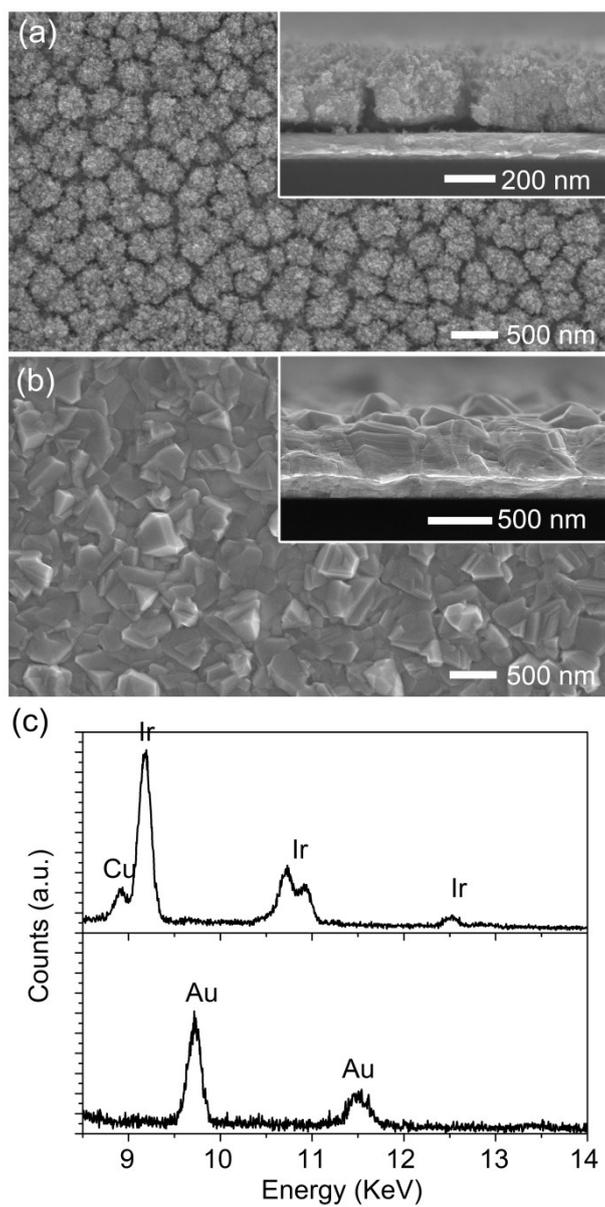


Figure S5. Top-view and cross-sectional SEM images of pure (a) Ir and (b) Au layers deposited on Au-coated substrate. (c) EDS spectra of pure Ir (upper) and Au (lower) metals.

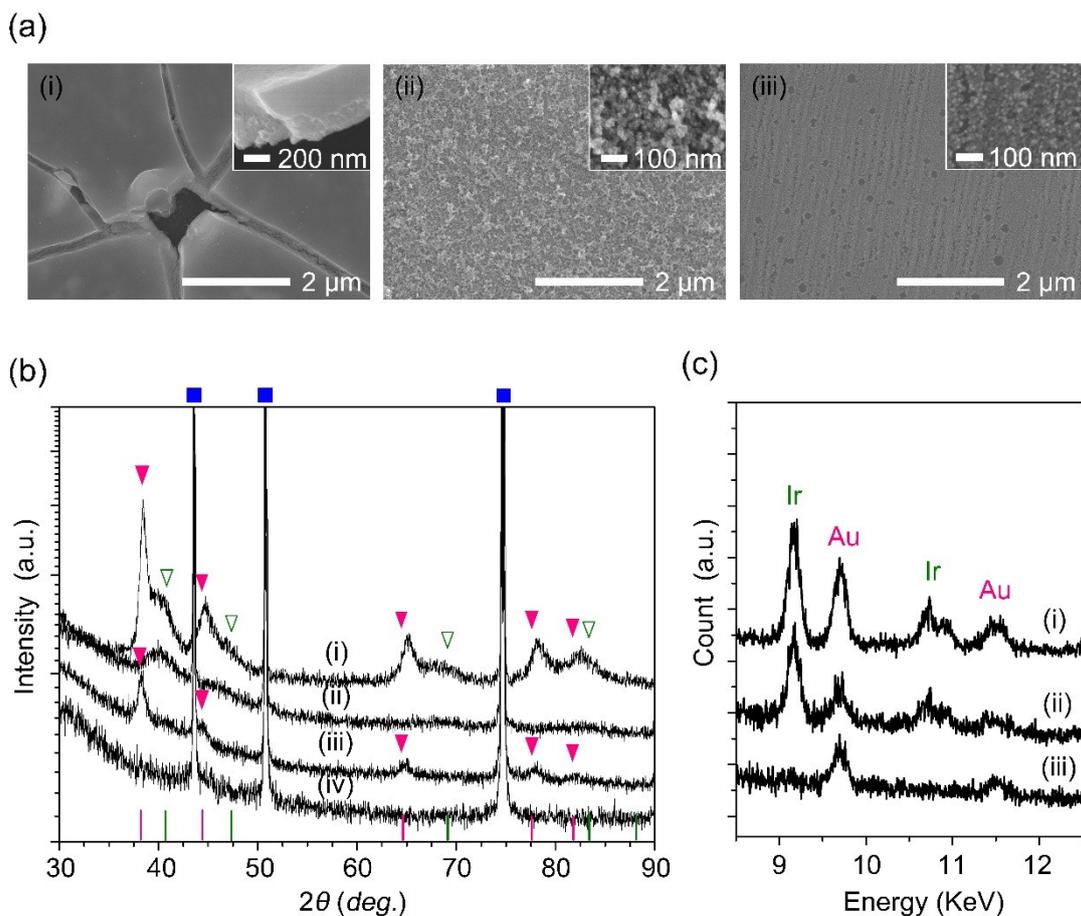


Figure S6. (a) SEM images, (b) XRD patterns and (c) EDS spectra of the deposits produced on stainless steel substrates under the aqueous electrolytes and the reduction potentials of (i) 5 mM  $\text{IrCl}_3 \cdot n\text{H}_2\text{O}$  and 0.1 mM  $\text{HAuCl}_4 \cdot n\text{H}_2\text{O}$  (0.5 Hz,  $V_R = 5$  V and  $V_O = 0$  V for 2 hr), (ii) 0.025 mM  $\text{IrCl}_3 \cdot n\text{H}_2\text{O}$  and 0.0125 mM  $\text{HAuCl}_4 \cdot n\text{H}_2\text{O}$  (1 Hz,  $V_R = 10$  V and  $V_O = 0$  V for 24 hr), which is identical to the electrodeposition condition of Ir-Au alloys shown in Figure 1. (iii) 0.025 mM  $\text{IrCl}_3 \cdot n\text{H}_2\text{O}$  and 0.0125 mM  $\text{HAuCl}_4 \cdot n\text{H}_2\text{O}$  (DC mode,  $V_R = 1$  V for 22 hr), (iv) the only stainless steel substrate as marked by the blue solid-rectangular. (JCPDS card #33-0397) (The pink upside-down solid triangles and green upside-down open triangles indicate Au and Ir phases, respectively.)