

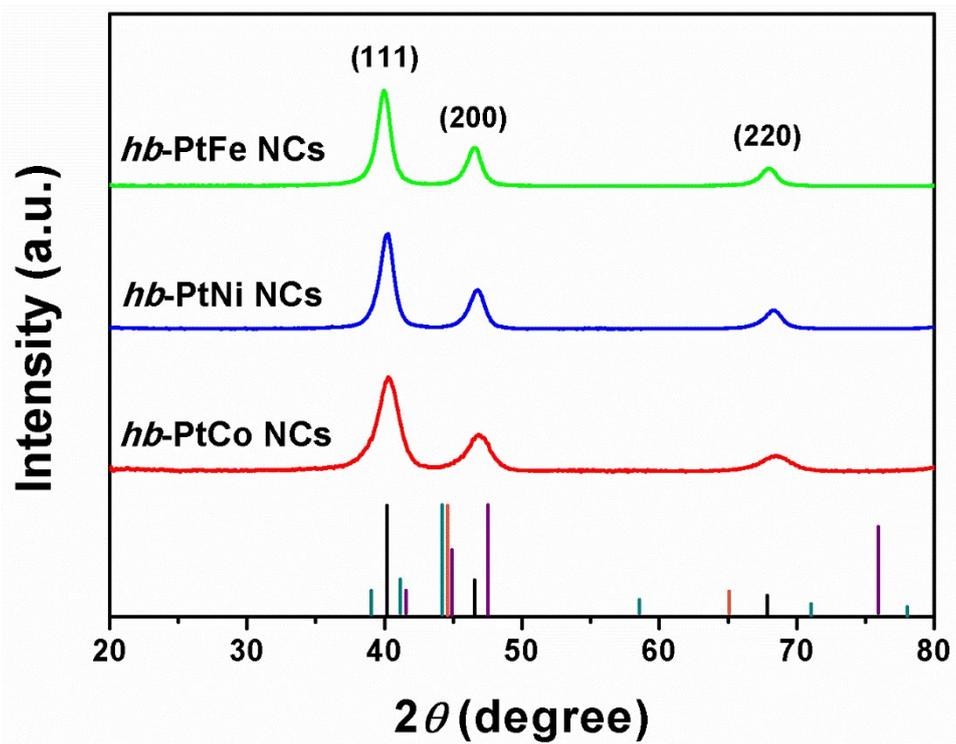
Electronic Supplementary Information (ESI) for

**Controlled synthesis of highly multi-branched Pt-based alloy nanocrystals  
with high catalytic performance**

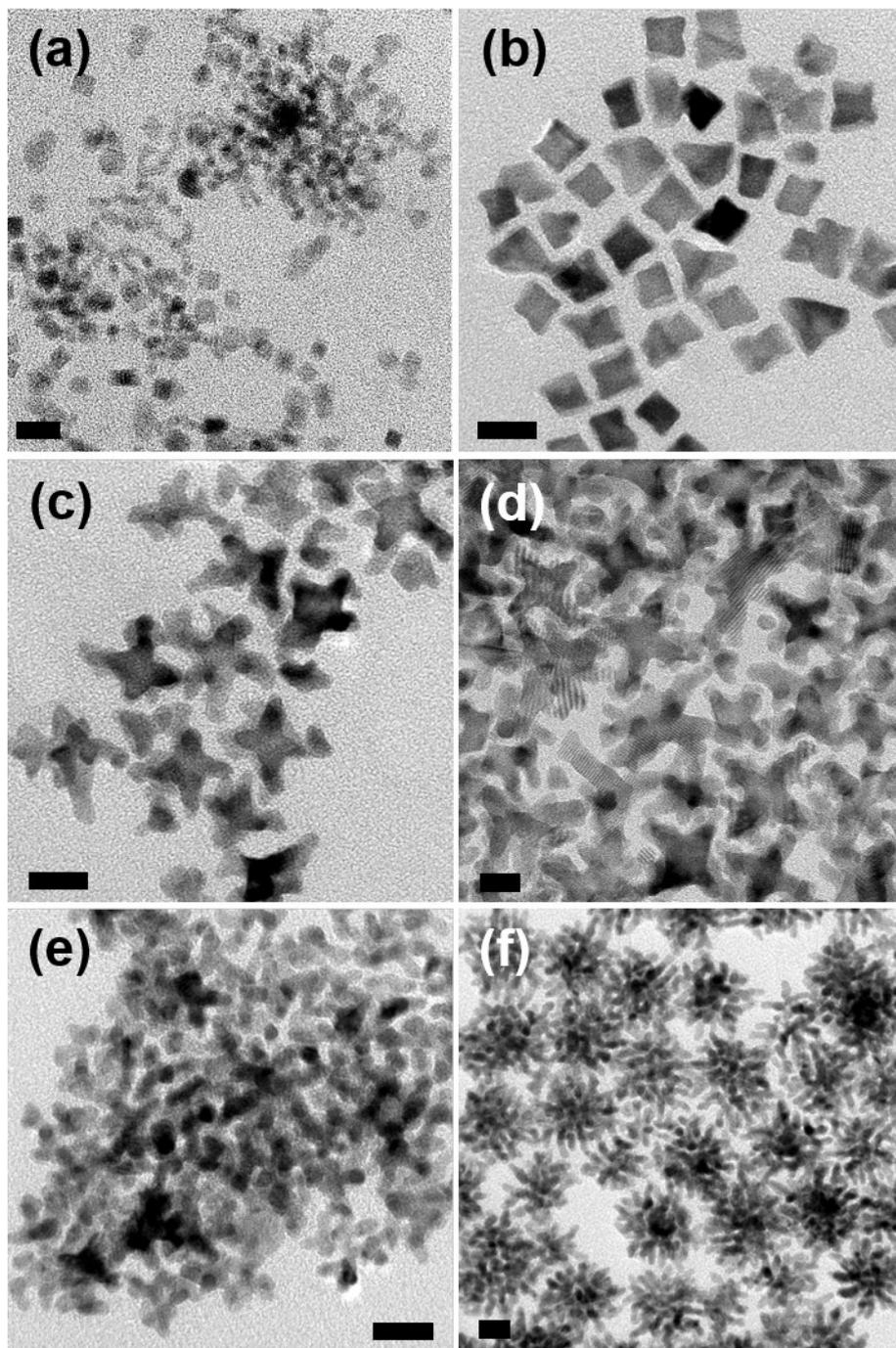
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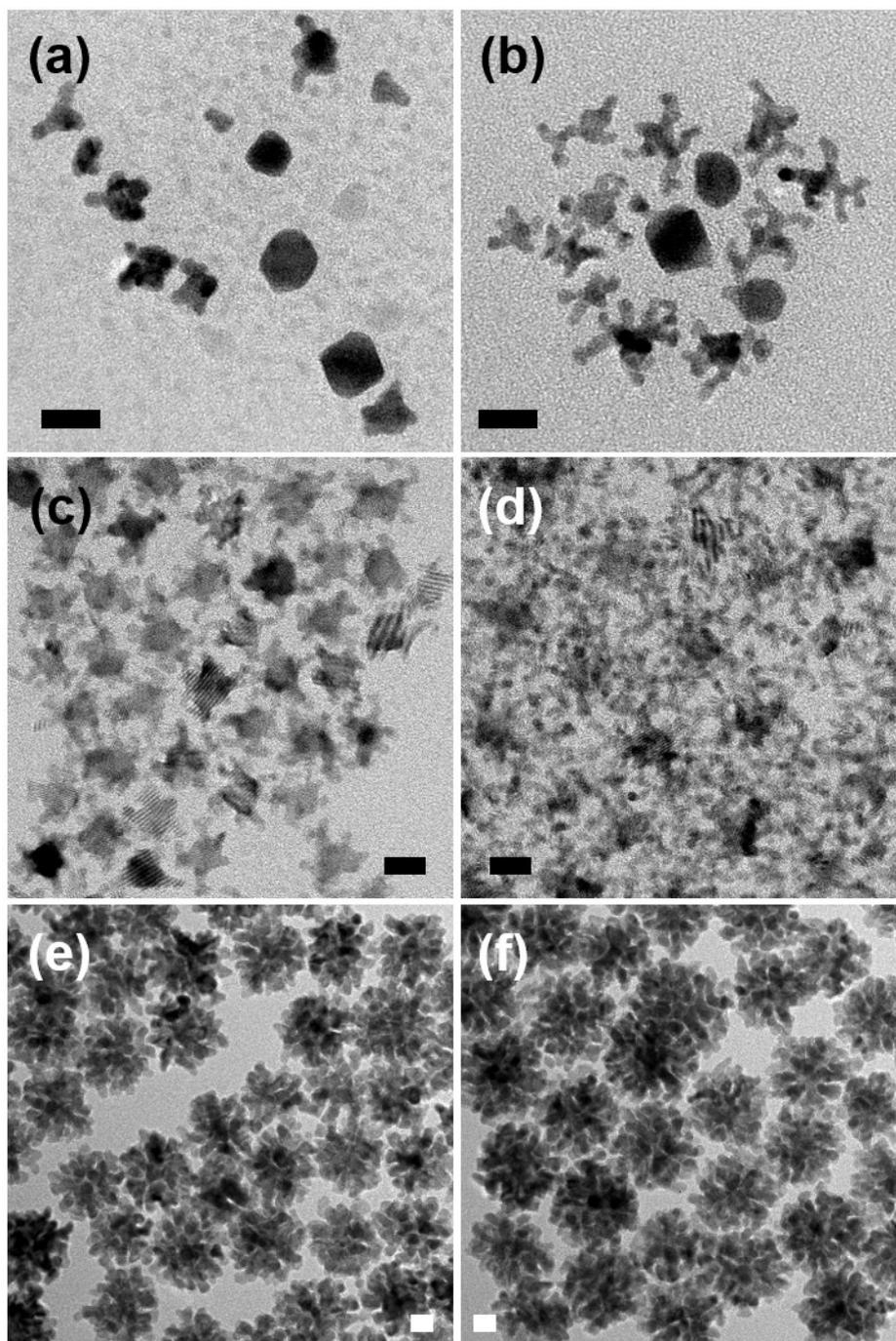
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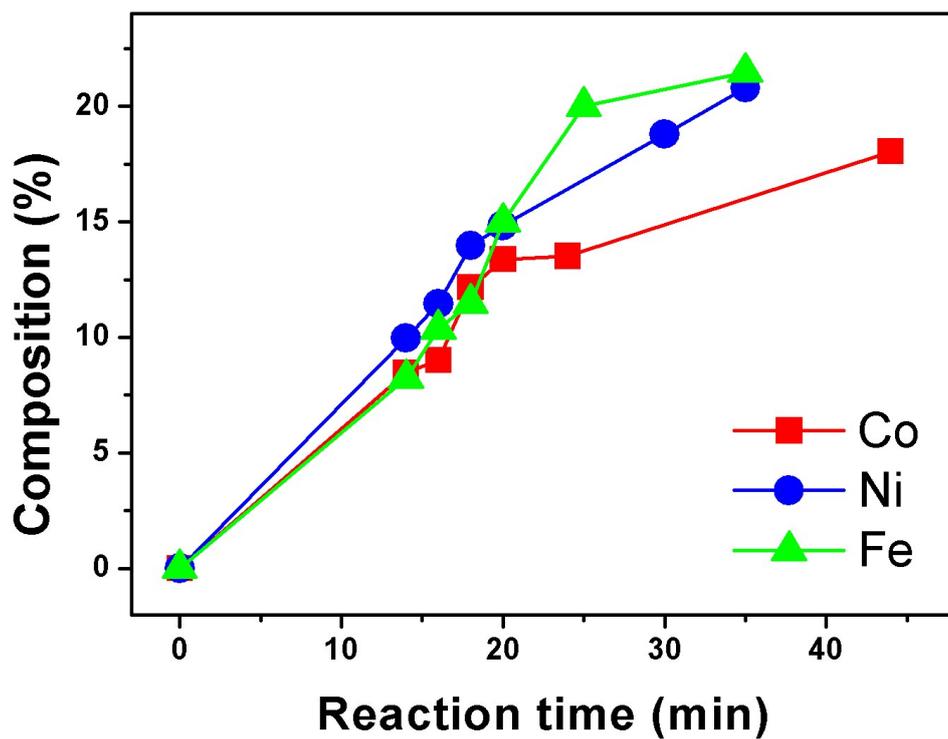
**Fig. S1** XRD patterns of the *hb*-PtCo, *hb*-PtNi, and *hb*-PtFe NCs. The intensities and positions for Pt, Co, Ni, and Fe references were taken from the JCPDS database: Pt (black, No. 01-1194), Co (purple, No. 00-005-0727), Ni (green, No. 00-045-1027), Fe (orange, No. 00-006-0696).



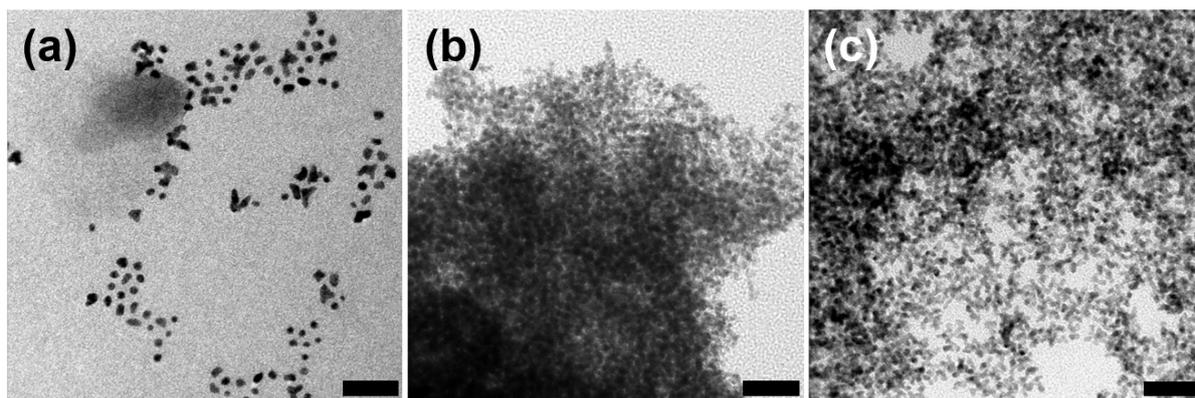
**Fig. S2** TEM images of the samples taken at different reaction stages for the case of the *hb*-PtNi NCs: (a) 200 (14 min), (b) 210 (16 min), (c) 220 (18 min), (d) 230 (20 min), (e) 230 (30 min), and (f) 230 (35 min) °C. Scale bars indicate 10 nm.



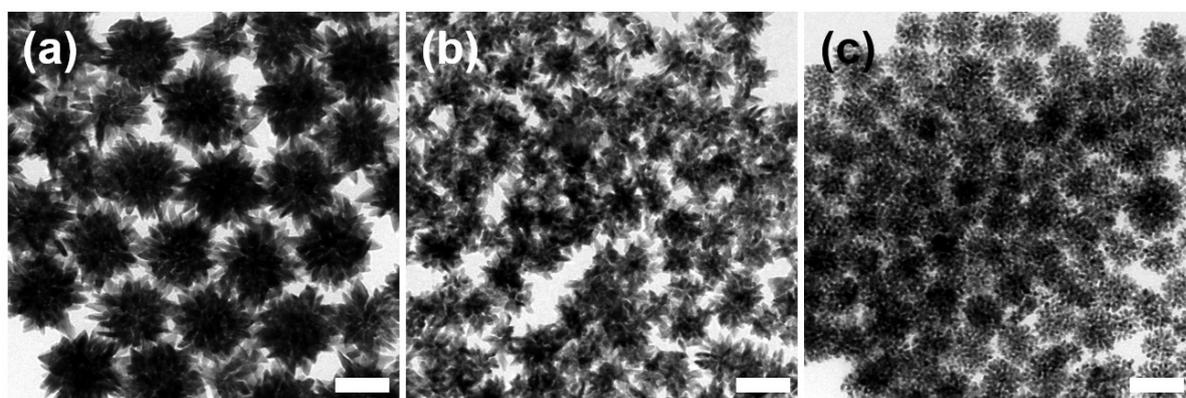
**Fig. S3** TEM images of the samples taken at different reaction stages for the case of the *hb*-PtFe NCs: (a) 200 (14 min), (b) 210 (16 min), (c) 220 (18 min), (d) 230 (20 min), (e) 230 (25 min), and (f) 230 (35 min) °C. Scale bars indicate 10 nm.



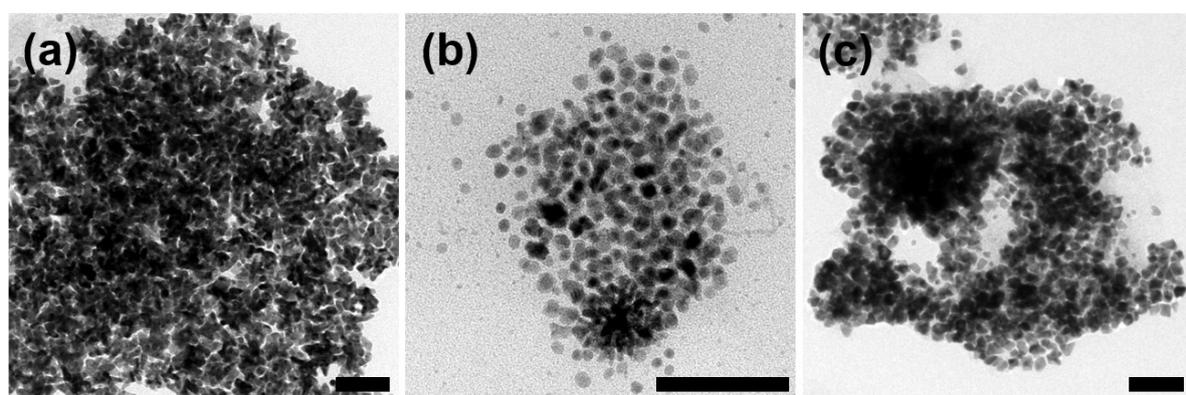
**Fig. S4** The changes in the M composition of the PtM NCs at different reaction times, which were evaluated by the EDS measurements.



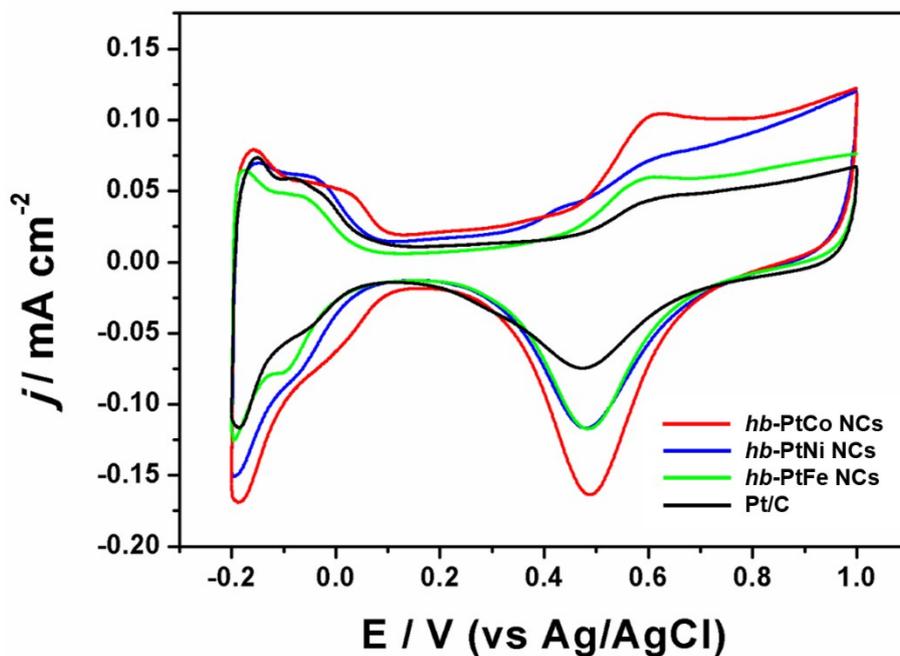
**Fig. S5** TEM images of nanostructures prepared with 1-octadecene instead of OAm while keeping the other reaction conditions, which were employed for the synthesis of the (a) *hb*-PtCo, (b) *hb*-PtNi, and (c) *hb*-PtFe NCs, unchanged. Scale bars indicate 20 nm.



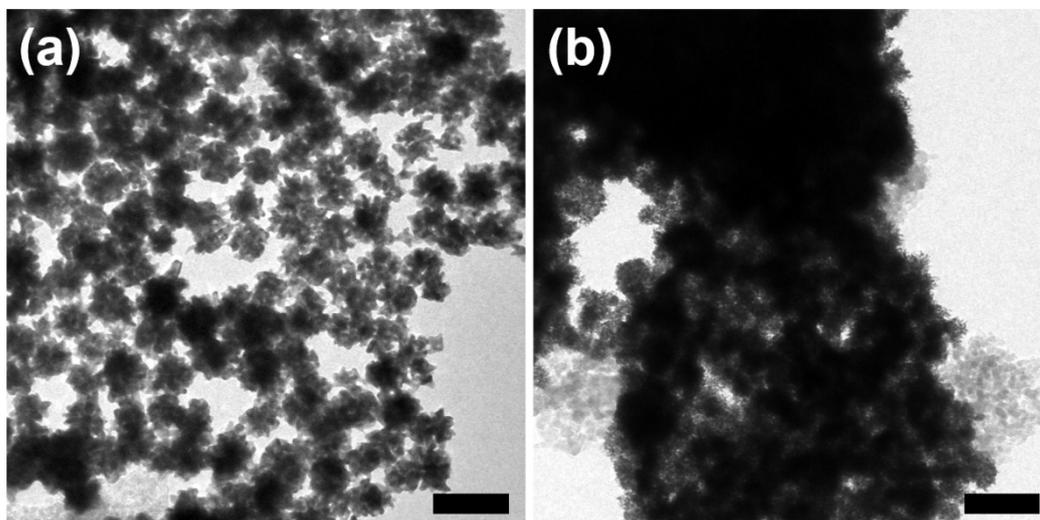
**Fig. S6** TEM images of nanostructures prepared in the absence of OA while keeping the other reaction conditions, which were employed for the synthesis of the (a) *hb*-PtCo, (b) *hb*-PtNi, and (c) *hb*-PtFe NCs, unchanged. Scale bars indicate 50 nm.



**Fig. S7** TEM images of nanostructures prepared in the absence of ACA while keeping the other reaction conditions, which were employed for the synthesis of the (a) *hb*-PtCo, (b) *hb*-PtNi, and (c) *hb*-PtFe NCs, unchanged. Scale bars indicate 50 nm.



**Fig. S8** CVs of the various catalysts recorded at room temperature in  $N_2$ -saturated 0.1 M  $HClO_4$  at a scan rate of  $50 \text{ mV s}^{-1}$ . The current values were normalized to the ECSAs of the catalysts.



**Fig. S9** TEM images of the (a) *hb*-PtCo and (b) *hb*-PtNi NCs after the electrochemical test. Scale bars indicate 100 nm. The Pt:M atomic ratios of the *hb*-PtCo and *hb*-PtNi NCs after the test were determined to be 79:21 and 77:23 by EDS measurements, respectively.