Highly Stable and Active Solid-Solution-Alloy Three-Way Catalysts by Utilizing the Entropy Effect

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A three-way catalyst (TWC) that concurrently converts three harmful gases, carbon monoxide, hydrocarbons, and nitrogen oxides (NO_x) is an essential technique for a sustainable society. Rh has been an essential element in TWC because only Rh can efficiently catalytically reduce NO_x. However, since Rh is scarce metal and its price is very fluctuating, significant efforts have been made to develop alternative TWC catalysts to Rh. Nevertheless, Rh is still irreplaceable, and very recently, the price of Rh marked a record high. We reported that solid-solution alloy NPs of Pd and Ru, which are located next to Rh in the periodic table of elements, exhibit an excellent NO_x reduction activity in the TWC reaction superior to Rh.[1] However, the PdRu homogeneous solid-solution alloy structure is transformed at high temperatures into an equilibrium segregated structure due to the immiscibility of Pd and Ru, and the catalyst loses its excellent NO_x reduction activity. To overcome this thermal stability issue, we have focused on the high-entropy effect. Recently, bulk high-entropy alloys consisting of five or more elements with equal or relatively large compositions have drawn much attention as a new type of structural material with high phase stability at higher temperatures.[2] Here, we introduced a third element (M) to stabilize the solid-solution structure in the form of PdRuM NPs at high temperatures (Figure). We demonstrated that PdRuM ternary solid-solution alloy nanoparticles exhibit a highly durable and active TWC performance. Our work provides insights into the design of highly durable and efficient alloy catalysts, guiding how to take the most advantage of the configurational entropy in addition to the mixing enthalpy.

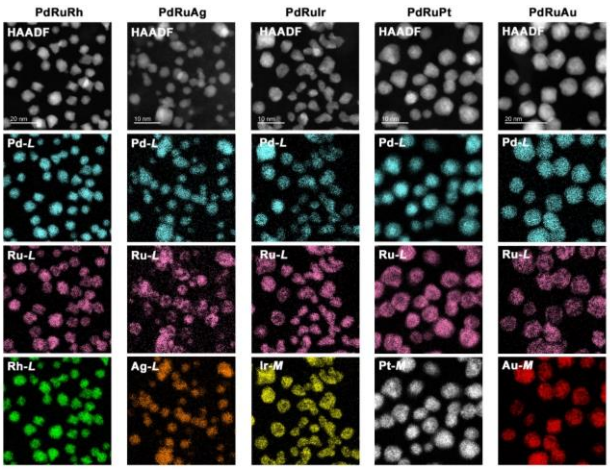

Figure HAADF-STEM images, Pd-L, Ru-L, and M elemental maps.

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