



Supplementary Material

Spontaneous Formation of Core@shell Co@Cr Nanoparticles by Gas Phase Synthesis

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Figure S1. Scheme of the MICS system for the fabrication of the nanoparticles.

Figure S1 presents a scheme of the MICS system used for the fabrication of the nanoparticles. In this scheme, the sputtering gas inlets (Argon in this work) are depicted in order to explain the differential fact of this equipment, in which Ar can be injected through the three magnetrons present in the MICS.

Figure S2 presents the ZFC-FC magnetization curves. The absence of a maximum hinders the possibility to extract a blocking temperature of this system.



Figure S2. ZFC-FC magnetization curves of (**a**) NPs d = 5.6 nm at Hapl = 100 Oe and (**b**) NPs of d = 6.9 nm at H_{apl} = 50 Oe.

Figure S3 presents the evolution of the elements detected in the XPS analysis as a function of the sputtering time. Note that the carbon arises from air exposure due to exsitu XPS analysis.



Figure S3. Evolution of the chemical composition extracted from the wide scan spectra taken at different sputtering times with Ar⁺.



Figure S4. Evolution of the cobalt components with sputtering time.

Figure S4 presents the evolution of the cobalt components with sputtering time extracted from the analysis of the XPS Co 2p core level. It must be taken into account that the presence of cobalt oxides does not decay with etching time, as the sample is formed by a multilayer of NPs. The stacking sequence of these NPs is not homogeneous and, therefore, there are contributions coming from different areas of several NPs at the same time.



Figure S5. Comparison of the NPs formed from a Co₉₀Cr₁₀ and Co₈₀Cr₂₀ target.

The comparison of the $C_{s-corrected}$ HR-STEM images depicted in Figure S5 clearly evidenced the thicker shell obtained in the $C_{080}Cr_{20}$ system. The larger presence of Cr favoured the thicker shell formation.



Figure S6. log (MFC-MZFC) vs T of the Co₈₀Cr₂₀ 7 nm NPs. A change in the slope can be observed at T \approx 70 K, indicating the blocking temperature.