

Supplementary Information

Shape and structural motifs control of MgTi bimetallic nanoparticles using hydrogen and methane as trace impurities

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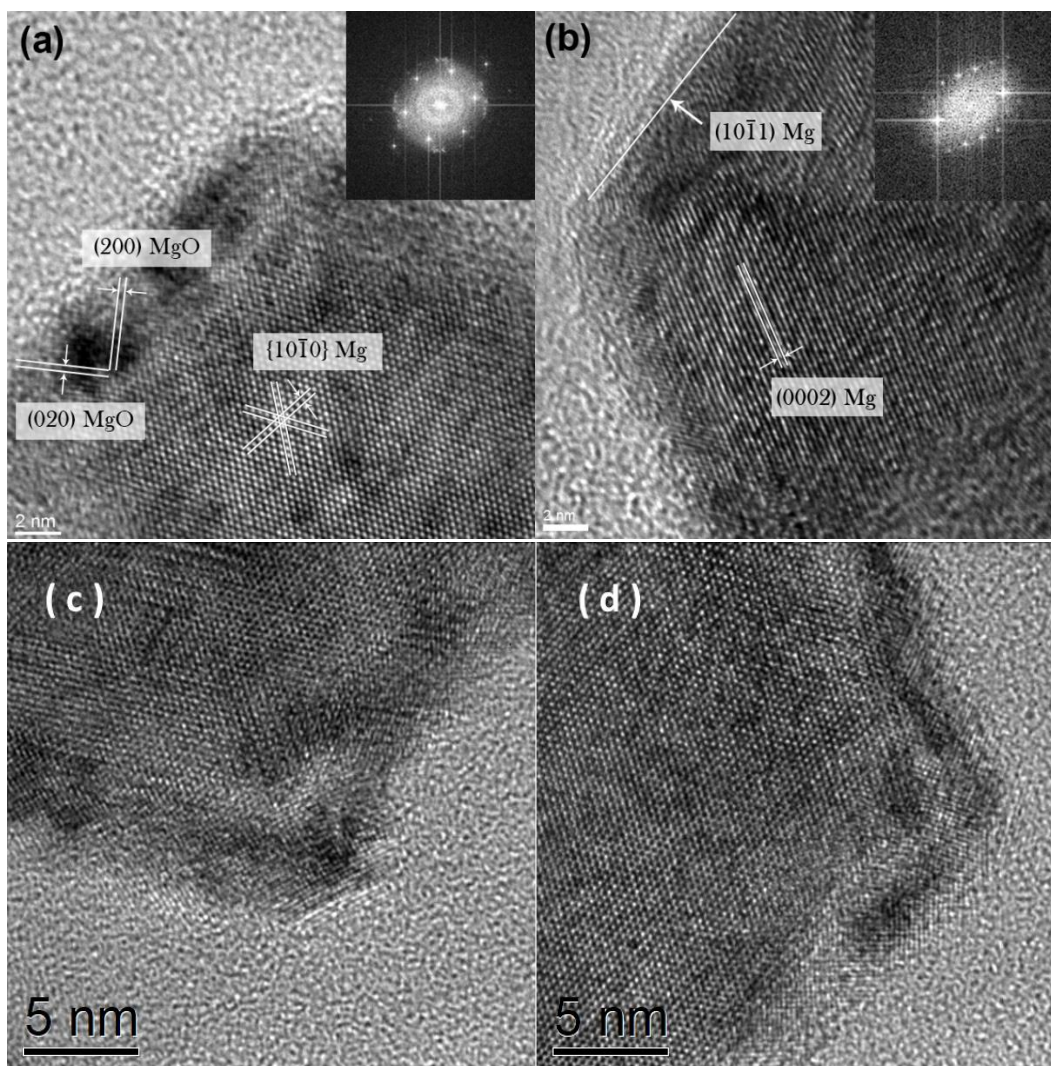


Figure S1: HRTEM bright field images of $\text{Mg}_{88}\text{Ti}_{12}$ nanoparticles (a) along the $[0001]$ zone axis that shows the OR between Mg and MgO (b) along the $[\mathbf{11\bar{2}0}]$ axis orthogonal to the $[0001]$ axis where the $\{0002\}$ and $\{\mathbf{10\bar{1}1}\}$ planes are parallel to a facet. The FFTs are shown in the insets. (c,d) HRTEM image of the MgTi NPs showing an crystalline MgO shell.

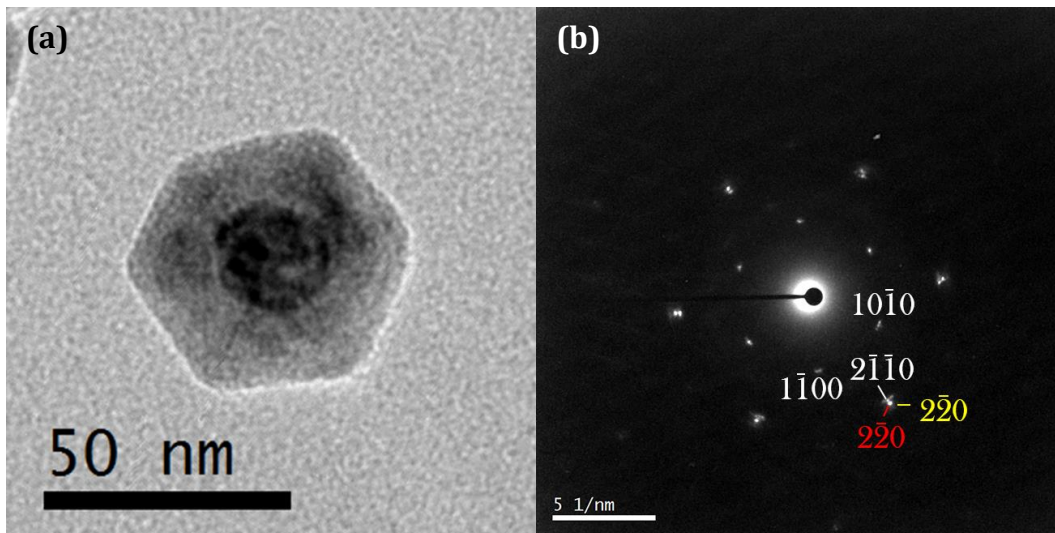


Figure S2: (a) High magnification bright field image of a core-shell $\text{Mg}_{85}\text{Ti}_{15}$ nanoparticle and (b) its correspond SAED pattern viewed along the Mg [0001] axis. HCP Mg, FCC TiC and FCC MgO planes are indexed in white, red and yellow, respectively.

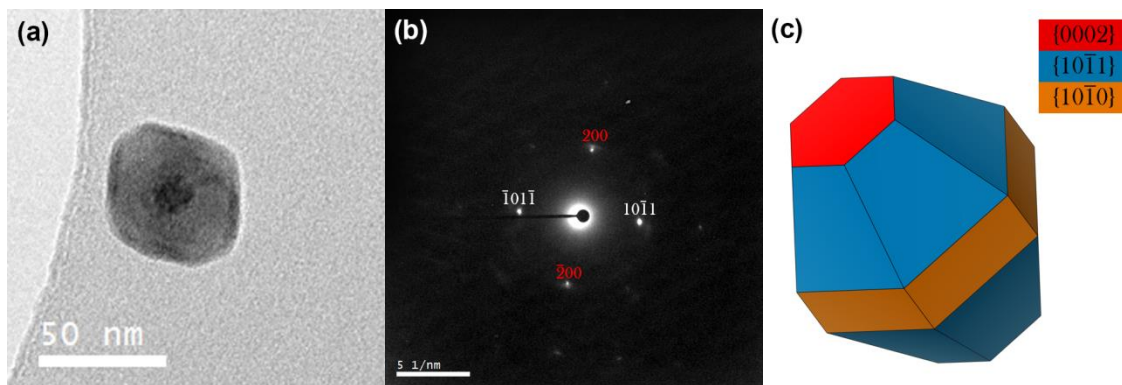


Figure S3: (a) High magnification bright field TEM image of a core-shell $\text{Mg}_{85}\text{Ti}_{15}$ nanoparticle (b) with its corresponding SAED pattern where Mg and TiC planes are indexed in white and red, respectively. (c) The 3D model in the equal orientation.

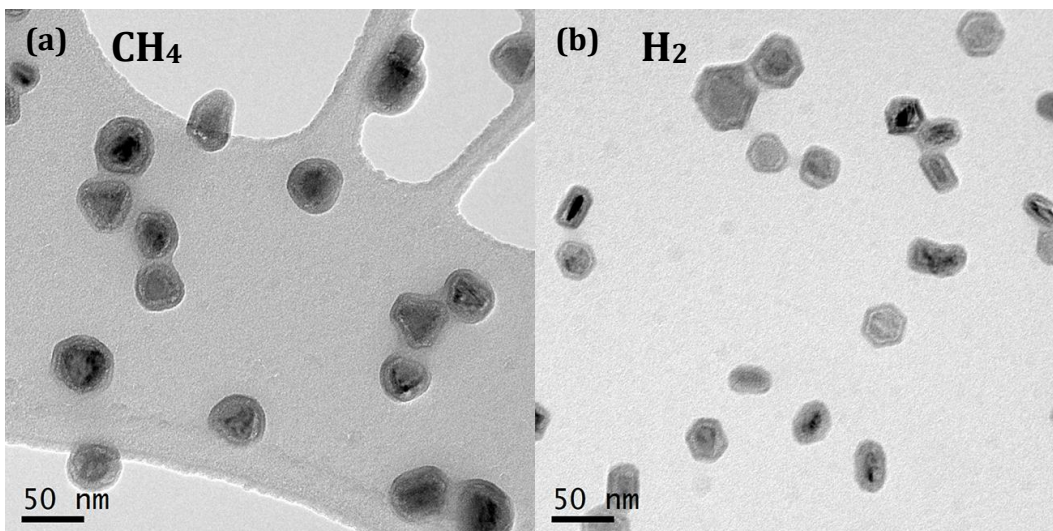


Figure S4: (a) Bright field TEM image of $Mg_{45}Ti_{55}$ nanoparticles grown in an Ar- CH_4 gas environment with porous (b) Bright field TEM image of $Mg_{64}Ti_{36}$ nanoparticles grown in an Ar- H_2 environment with porous subshell.

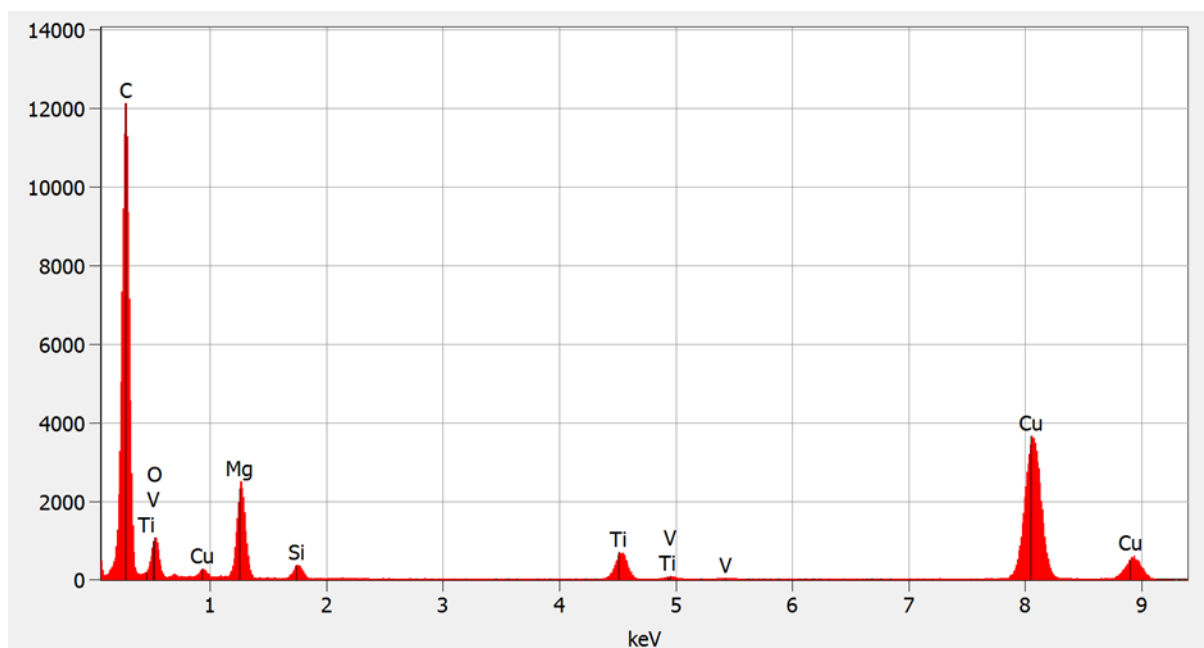


Figure S5: The chemical composition of the MgTi NPs was determined by quantifying the X-ray spectrum as acquired by the EDX detector that our TEM is equipped with. For most accurate

results, a cluster of several tens (if coverage allowed) representative NPs were probed by the focused 200kV electron beam. The X-ray spectrum was acquired for as long as was necessary to obtain at least several thousand counts of the relevant spectral peaks, which usually took around two minutes, while continuously correcting for drift. The raw data was quantified by using the element K line with the Cliff-Lorimer without absorbance method, carried out by the NSS software V3.1 from Thermo Fisher Scientific. The quantified result of the above spectrum is shown in the following table.

Element line	Counts	Counts std. dev.	at%	at% std. dev.
C K	86991	475	96.038	0.524
O K	6293	88	1.086	0.015
Mg K	20907	167	1.092	0.009
Si L	0	41	-	-
Si K	3508	88	0.132	0.003
Ti L	292	367	-	-
Ti K	9502	150	0.206	0.003
V L	0	274	-	-
V K	21	53	0	0.001
Cu K	69250	462	1.445	0.01
Cu L	1752	62	-	-

The relative composition of Mg and Ti is obtained by dividing the Mg or Ti signal with their sum signal, and the uncertainty is corrected accordingly. For instance, $100 \times 1.092 / (1.092 + 0.206) = 84.129$ at% Mg, with uncertainty $0.009 \times (84.129 / 1.092) = 0.693$ at%. Hence, individual quantified results determine the Mg and Ti composition with a standard deviation of less than 1 at%, despite the large contribution (>90 at%) of carbon from the holey carbon substrate. This data acquisition and quantification procedure was carried out at least twice, or multiple times when necessary (e.g. in the case of very low coverage), to improve the

measurement precision. Typically the standard deviation of multiple individual measurements is less than 1 at%, but occasionally is higher when for instance the coverage or total counts is low.



Figure S6. Showing different types of section target arrangement used for the production of nanoparticles.

Information for generic geometrical model.

Furthermore it is assumed that the NPs are spherical i.e. aspect ratio of unity, with a pure Ti core and a fixed MgO thickness of 2 nm. These assumptions can be justified because the influence of the aspect ratio is negligible, and the Ti density in Ti/TiC_x/TiH_x is relative similar (within 10% of each other) such that the error is falls within the experimental uncertainty. Therefore, the composition is the leading term that determines the dimensions of the NPs.