Supplementary Information

Tuning Structural Motifs and Alloying of Bulk Immiscible Mo-Cu Bimetallic Nanoparticles by Gas-Phase Synthesis.

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A. Target configuration

Figure 1. Showing the section target of Mo-Cu used for the production of nanoparticles.

B. Nanoparticle deposition source

Figure 2. Showing the schematic of the nanoparticle production set up (Mantis deposition Ltd).
C. Nanoparticle Synthesis Condition

<table>
<thead>
<tr>
<th>Current (A)</th>
<th>GasFlow Rate(sccm)</th>
<th>shape</th>
<th>Structure</th>
<th>Aperture Size (mm)</th>
<th>Figures in the Manuscript</th>
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</thead>
<tbody>
<tr>
<td>0.250</td>
<td>40</td>
<td>Cube</td>
<td>Alloy</td>
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<td>Figure 1</td>
</tr>
<tr>
<td>0.250</td>
<td>70</td>
<td>Spherical</td>
<td>Alloy</td>
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<tr>
<td>0.350</td>
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<td>Cube+Spherical</td>
<td>Core-Shell</td>
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<tr>
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<td>Onion/Multishell</td>
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<tr>
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<td>Core-Shell</td>
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<td>Figure 6</td>
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<tr>
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<td>150</td>
<td>Spherical</td>
<td>Janus/Dumbbell</td>
<td>5.5</td>
<td>Figure 7</td>
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</table>

**Low gas flow** 0-40 sccm  
**Medium gas flow** 40-80 sccm  
**High gas flow** >80 sccm.

**Table 1.** The table (upper) shows the various parameters involved in the synthesis of various structural motifs of Mo-Cu bimetallic nanoparticles. (b) The table (bottom) shows the gas flow rates stated in the manuscript.
Figure 3. Shows the bright field TEM image of the Mo-Cu alloy NPs. (a) Mo-Cu alloy NPs with a size distribution of 5±3 nm. (b) Mo-Cu alloy nanoparticles produced with an average composition of Mo 75.6±1 at% and Cu 24.4±1 at% showing the NP size distribution of 20±2 nm. (c) Mo-Cu alloy nanoparticles produced with an average composition of 70±1 at.% Mo and 30±1 at.% Cu showing nanoparticle size distribution of 16 ±2 nm.
Figure 4. Showing Mo/Cu core/shell nanoparticles in two different magnifications with the same composition and size range of the Mo–Cu alloy nanoparticles shown in figure 1 of the manuscript.
**Figure 5.** (a) HRTEM image of the Mo-Cu-Mo onion structure (b) FFT of the image showing the presence of pure Cu along with pure Mo.
Figure 6. (a) Bright field image of the Mo-Cu NPs in the intermediate stage of forming a completely phase separated janus structure. Inset shows the NP at high magnification.  (b) HRTEM image showing the lattice spacing of Cu d (111) and Mo d (110) as a separated Janus structure.