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Facile Fabrication of Magnetically Recyclable Metal-Organic Framework Nanocomposites for Highly Efficient and Selective Catalytic Oxidation of Benzylic C–H Bonds

Oxidation of Benzylic C–H Bonds with HKUST-1@Fe₃O₄

**Preparation of HKUST-1@Fe₃O₄:**

\[
\begin{align*}
\text{Fe₃O₄-CO₂H} & \quad \text{Cu(OAc)}_2 \cdot \text{H₂O} \\
\text{HKUST-1@Fe₃O₄} & \quad \text{BTC} \quad \text{Cu(OAc)}_2 \cdot \text{H₂O}
\end{align*}
\]

**Oxidation of benzylic hydrocarbons:**

\[
\begin{align*}
\text{HKUST-1@Fe₃O₄ (25 mg)} & \quad \text{TBHP (2.5 mol equiv), benzonitrile (0.5 mL)} \\
{} & \quad 80 ^\circ \text{C, 14 h}
\end{align*}
\]

94.7% conversion  95.2% selectivity  >99% conversion  >99% selectivity  >99% conversion  >99% selectivity  >99% conversion  93.3% selectivity  >99% conversion  97.1% selectivity

**Significance:** The magnetic core–shell nanocomposites HKUST-1@Fe₃O₄ were prepared from Fe₃O₄-CO₂H (Φ 20 nm), polyvinylpyrrolidone (PVP), Cu(OAc)_2, and trimesic acid (BTC), in which the iron-based nanoparticles were encapsulated by the resulting HKUST-1 shell [for the copper-organic framework of Cu(OAc)_2 and trimesic acid, see: Chui et al. Science *1999*, 283, 1148]. The oxidation of benzylic C–H bonds was carried out with HKUST-1@Fe₃O₄ and TBHP to give the corresponding desired carbonyl products in up to >99% conversion and >99% selectivity.

**Comment:** The catalyst was characterized by SEM, HR-TEM, PXRD, BET, and FT-IR analyses. Elemental analysis revealed a ratio of copper and iron of 19.34% and 28.63%. The catalyst was recovered by an external magnet and reused twice without significant loss of the catalytic activity.