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Haemoglobin as a bio-derived precursor for FeNx single-site catalysts

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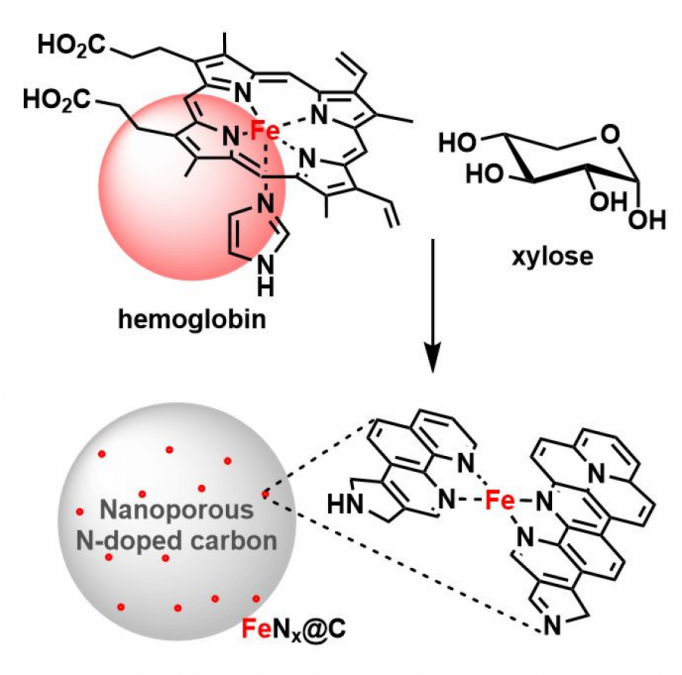
PURPOSE OF THE ABSTRACT

Single-site metal catalysts combine high activity and recyclability. Although their synthesis and characterisation are still challenging, they appear to be ?ideal' catalysts as they are 100% metal atom-efficient. Individual Fe atoms chelated to nitrogen atoms supported on carbon materials (FeNx@C) have been developed for organic chemistry for various transformations, but they are currently made from synthetic products (such as Fe-phenantroline complexes and carbon black). [1]

Here we used haemoglobin, a waste from the meat industry that contains these so-called FeNx sites. Every year just in UK, approximately 100 kt of blood (representing 20 kt of proteins) is generated, driving the need to valorise such amounts of waste. We transformed haemoglobin into the desired material by thermal treatment, combining it with xylose as a bio-based carbon precursor. The Fe single-site nature was confirmed by X-ray absorption technique (EXAFS) and high-resolution microscopy (STEM-HAADF).

With their catalytic versatility in hand, we tested FeNx@C for both the reduction and N-formylation of nitroarenes. In both transformations the catalyst was fully selective to nitro groups and recyclable without degradation or aggregation of the FeNx sites, showing the viability of hemoglobin as a catalyst precursor.

FIGURES



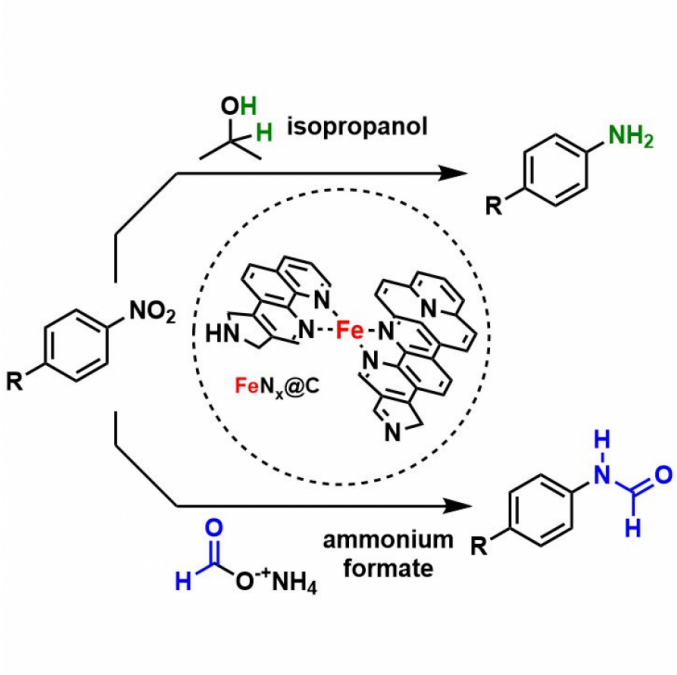


FIGURE 1

FeN@C catalyst preparation from hemoglobin

Xylose was used as a carbon source. The material was prepared in a simple two-step method: Hydrothermal carbonisation (HTC, 220°C, 12h) followed by pyrolysis at 1000°C, 2h under 6% O2/N2

FIGURE 2

Catalytic activity of FeNx@C

Two reactions have been explored: nitroarene transfer hydrogenation (using isopropanol and potassium formate) and one-pot reduction/N-formylation (using ammonium formate)

KEYWORDS

Single-site catalysts | Nitroarene reduction | Nitroarene N-formylation | Biowaste

BIBLIOGRAPHY

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