**Supplementary materials**

**An iron ore-based catalyst for producing hydrogen and metallurgical carbon via catalytic methane pyrolysis for decarbonization of the steel industry**

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XRD analysis

A Bruker D8 Discover in transmission geometry with monochromated cobalt Kα1 radiation and a position sensitive detector was used to obtain XRD patterns. The cobalt source is monochromated with Kα = 1.789Å, operating at 35 kV and 40 mA. The data sets were acquired in continuous scanning mode over the 2θ range 20° – 80°, using a step size of 0.00750404° per second. The raw data was peak matched using EVA software and PDF-1997 database.

GC analysis

The exhaust gases were collected in foil, gas bags every 30 minutes for 5 hours. The gases were then analysed on an Agilent 8860 GC system equipped with a packed column (Agilent ShinCarbon ST) and a Thermal Conductivity Detector (TCD). Argon was the carrier gas, and the oven temperature was maintained at 150 °C for each analysis. The Clarity version 8.5 software was used to develop and control methods, run single or multiple samples, and view and modify run results. The hydrogen and methane concentrations were obtained using a calibration curve, that was made beforehand with known concentrations of hydrogen and methane.

FT-IR analysis



Figure S1. FT-IR spectrum of the unmilled iron ore

Raman analysis – iron oxide standards

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1. (ii)



(iii) (iv)

Figure S2. Raman spectra of four iron oxide standards: (i) FeO, (ii) Fe3O4, (iii) γ-Fe2O3, (iv) α-Fe2O3

SEM images – iron ore

A picture containing text, wall, black, nature

Description automatically generated A close-up of a leaf

Description automatically generated with medium confidence A picture containing rock, stone

Description automatically generated

(i) (ii) (iii)

A close-up of some rocks

Description automatically generated with low confidence A picture containing barnacle

Description automatically generated A close-up of some rocks

Description automatically generated with low confidence

(iv) (v) (vi)

Figure S3. SEM images (i) – (vi) showing changes in the morphology of the iron ore at milling times of 0, 60, 120, 180, 240 and 300 minutes respectively.

XRD analysis – iron ore



Figure S4. XRD patterns showing changes in the crystallinity of the iron ore at milling times of 0, 60, 120, 180, 240 and 300 minutes

Raman analysis – milled iron ore

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Figure S5. Raman spectra showing changes in the iron oxide phases of the iron ore at milling times of 60, 120, 180, 240 and 300 minutes.

SEM images – carbonaceous material

 A picture containing text, old

Description automatically generated

(i) (ii)

A picture containing old

Description automatically generated A picture containing black

Description automatically generated

(iii) (iv)

Figure S6. SEM images of the carbonaceous material formed during methane pyrolysis

XRD analysis – transmission medium



Figure . XRD pattern of the transmission medium

SEM-EDS – used catalyst

A picture containing dark, outdoor object, clouds

Description automatically generatedA large group of lights in the night sky

Description automatically generated with low confidenceA picture containing purple, dark, star, night sky

Description automatically generatedA picture containing grass, green, night sky

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A picture containing green, dark

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A picture containing graphical user interface

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Figure S8. SEM-EDS elemental analysis of used catalyst