

In situ valorization of impurities enabling energy efficient meltless high-strength steel production from ore

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Additional Declarations: **Yes** there is potential Competing Interest. Dr. Pei Sun and Dr. Z. Zak Fang are filing a patent application related to the HRMLES process.

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Abstract

The steel industry faces immense pressure to decarbonize. Hydrogen-based direct reduction is a leading green steel alternative, but its viability is hampered by the high cost of hydrogen. Moreover, hydrogen alone cannot reduce stable oxide impurities in ores, leaving detrimental inclusions that must be removed as slag during molten steel processing. Here, we demonstrate an impurity valorization strategy that addresses this limitation. By introducing a small amount of solid carbon during sintering of H₂-reduced iron, we trigger in situ carbothermic reduction of SiO₂ at low temperatures. We provide direct evidence for a two-step solid-state pathway: carbon first dissolves into the iron matrix, and the dissolved carbon then reduces silica, converting harmful SiO₂ inclusions into beneficial dissolved silicon, a potent alloy strengthener. Using this approach, we produce an Fe-Mn-Si-C steel with an ultimate tensile strength above 800 MPa directly from ore without melting, opening a pathway for sustainable meltless metallurgy.

Introduction

Iron and steel production is one of the largest industrial sources of greenhouse gas emissions, accounting for roughly 7–8% of global anthropogenic emissions¹. This footprint is dominated by the carbon-intensive blast furnace-basic oxygen furnace (BF-BOF) route. To align with Paris Agreement goals, direct emissions from steelmaking must be reduced by ~60% by 2050². In the BF-BOF process, coke derived from coal serves as both reductant and heat source, reducing iron ore at temperatures up to 1500 °C to produce molten pig iron that is subsequently refined to steel. This route emits roughly 2.34 t CO₂ per tonne of crude steel¹. Lower-carbon routes, particularly gas-based direct reduction of iron coupled with electric arc furnaces (EAF), have therefore gained traction (1.47 t CO₂/t crude steel)¹. Several demonstration projects³ now replace fossil coke with green hydrogen to reduce iron oxides, followed by melting the direct-reduced iron (DRI) in an EAF or BOF⁴. Although hydrogen-based steelmaking can substantially lower CO₂ emissions when powered by renewable energy, it remains anchored in the energy-intensive paradigm of molten metallurgy. To truly revolutionize steel production, a fundamentally new paradigm is needed.

An emerging trend in the research community is to directly convert iron oxide with alloying oxides into alloys in the solid state⁵⁻¹⁴. These studies support a paradigm shift toward a solid state “oxides-to-steel” concept that eliminates the molten metal stage entirely. Solid-state hydrogen reduction offers a carbon-free chemical pathway at temperatures far lower than blast furnace smelting. When coupled with near-net-shape (NNS) manufacturing methods, including powder metallurgy (PM), this approach could enable oxide feedstocks to be converted into finished steel products without ever melting the metal.

Despite this promise, most prior studies of hydrogen-based direct reduction have relied on

idealized feedstocks such as reagent-grade iron oxide or synthetic oxide mixtures⁵⁻¹⁴. These high-purity precursors simplify mechanistic studies but do not capture the mineralogical complexity of mining-derived ores. In practice, even high-quality iron ore concentrates contain a few percent gangue that cannot be fully eliminated economically. According to the Ellingham diagram (Fig. 1a), these gangue impurities, containing primarily silica and oxides of alkaline metals (MgO, CaO, and Al₂O₃), are more stable than H₂O and therefore are not readily reduced by hydrogen under typical direct-reduction conditions. By relying on synthetic or highly purified oxides, prior work has bypassed this impurity challenge, leaving unresolved whether solid-state steelmaking can operate with realistic ore concentrates.

In real ores, residual gangue can influence both reduction and consolidation. Silica and other impurities in the concentrate (produced from low-grade iron ore by magnetic separation, gravity separation, or flotation after crushing and grinding) can persist after hydrogen reduction and carry over into the metallic product as non-metallic inclusions. These impurity-derived phases can affect consolidation and sintering of the powder, downstream processing, and ultimately mechanical properties. For this reason, iron powder reduced directly from iron ore is not commonly used as a feedstock for powder-metallurgy components, particularly in demanding automotive applications. Instead, most commercial iron and steel powders used for sintering are produced by atomizing molten, refined steel, often scrap-derived, from EAF or induction furnace routes^{15,16}. In addition, impurities can form low-melting point phases (e.g., silicates) that block pore networks during reduction, slowing or even stalling the conversion of oxide to metal¹⁷. Therefore, a central question is whether a solid-state metallurgy route can work with real concentrates, with their inherent impurity levels, while still producing steel with useful mechanical properties.

In conventional smelting, gangue impurities are separated as slag. In a solid-state PM route, however, they remain in the material as dispersed inclusions. These inclusions act as stress concentrators and can severely deteriorate the mechanical properties of steel products^{18,19}. Effective impurity management is therefore essential, either through upstream gangue removal, such as enhanced beneficiation or leaching, or through in situ strategies that transform these impurities. In the taconite-derived concentrate studied here, silica is the dominant residual impurity, and its conversion is the focus of this study. Because complete silica removal from iron ore is costly and often not fully effective, our Hydrogen-Reduction Meltless Steelmaking (HRMLES) process, as shown in Fig. 1b, is designed to valorize silica during processing by converting it from a detrimental oxide inclusion into a useful alloying source.

Although silica inclusions are generally detrimental, silicon dissolved in iron can provide substantial metallurgical benefits. Silicon is a well-established solid-solution strengthener in steel due to its size misfit in the iron lattice, which raises the alloy's strength²⁰. Silicon also strongly influences phase transformations by suppressing the formation of cementite (Fe₃C) and pearlite during cooling²¹. For example, advanced high-strength steels²², including dual-phase, transformation-induced plasticity, and quenching-and-partitioning grades, commonly contain ~0.45-2 wt% Si to promote microstructures with improved strength-ductility combinations. Thus,

converting silica impurities into dissolved silicon could simultaneously remove harmful inclusions, strengthen the ferritic matrix, and tailor microstructural evolution. In this way, a detrimental oxide inclusion can be valorized into a beneficial alloying element, analogous to intentional Si additions in conventional steelmaking.

Building on this insight, here we show that silica-to-silicon conversion can be achieved in HRMLES by adding a small amount of carbon during sintering of hydrogen-reduced iron powder. Under these conditions, silica can be carbothermally reduced at relatively low temperatures ($<1000\text{ }^{\circ}\text{C}$), far below those used in conventional silicon production ($1900\text{--}2100\text{ }^{\circ}\text{C}^{23}$) because the reaction is driven by low CO partial pressure and by the reduced activity of silicon once it dissolves into iron. Using thermal analysis, microstructural characterization and in situ SEM heating, we show that the reaction follows a two-step solid-state pathway in which carbon first dissolves into iron and is then transported through the iron matrix to reduce silica. We define the thermodynamic and kinetic window for this impurity-valorization route, demonstrate production of Fe-Si-C and Fe-Mn-Si-C steels with high strength directly from ore, and benchmark the process against both blast-furnace steelmaking and H_2 DRI-EAF processing.

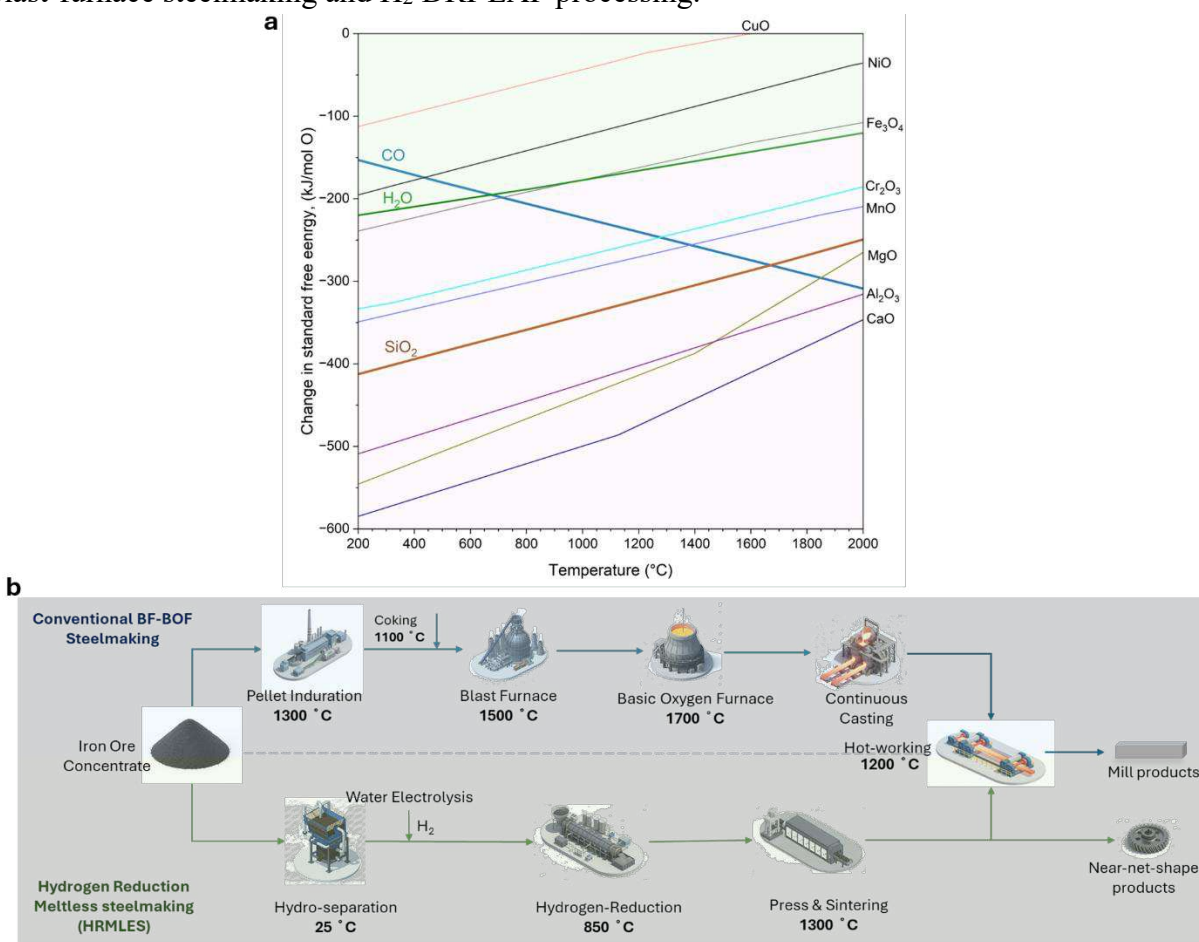


Fig. 1 | Hydrogen-Reduction Meltless Steelmaking directly from iron ore concentrate.

a, Ellingham diagram comparing oxides relevant to steelmaking, ore-gangue impurities, and the reducing agents H_2 and C . Calculations were performed using HSC Chemistry 9 with condensed phases at unit

activity and gas species referenced to standard pressure (10^5 Pa). The H_2O line corresponds to $P_{\text{H}_2\text{O}}/P_{\text{H}_2} = 1$ for the reaction $\text{H}_2(\text{g}) + 1/2 \text{O}_2(\text{g}) = \text{H}_2\text{O}(\text{g})$. Oxide lines located at higher oxygen potential correspond to less stable oxides and therefore easier reduction. **b**, Schematic of the main steps of the HRMLES process, with the maximum projected processing temperatures indicated, compared to conventional BF-BOF steelmaking.

Results

A Solid-State Process for Producing Iron Powder from Iron Ore Feedstocks

The HRMLES process, as shown schematically in Fig. 1b, integrates ore beneficiation, hydrogen reduction, and solid-state alloying into a continuous workflow, eliminating all molten-metal stages. The feedstock used here was a concentrate beneficiated from taconite mined in the Mesabi Iron Range, USA. Taconite is a low-grade iron ore containing 40-55% silica and 15-35% magnetite²⁴, and it has been the dominant iron ore mined in the USA since the 1950s. The as-received concentrate, the same material that would feed a blast furnace after pelletizing and sintering, still contains significant gangue (principally silica) despite prior magnetic separation.

As shown in Fig. 2a, the as-received concentrate (Fig. 2c) contained 1.54 wt% silicon, corresponding to an estimated 3.3 wt% SiO_2 . To lower this initial impurity burden to a manageable level for solid-state processing, we applied a physical beneficiation protocol. The concentrate was first ball-milled in water to liberate fine gangue minerals interlocked with the magnetite grains. A custom hydro-separation process was then used to exploit the density difference between the heavy iron oxides and the lighter silica-rich gangue (Supplementary Fig. 1). After hydro-separation, the concentrate consisted mainly of particles smaller than $10\text{ }\mu\text{m}$ (Fig. 2d). This physical beneficiation step reduced the silicon content by over 75%, from 1.54 to 0.38 wt%, with similar reductions in other impurities like Mg, Ca, and Al (Supplementary Table 1). Phase analysis confirmed substantial, but incomplete silica removal. Quartz peaks were no longer detectable by laboratory XRD using $\text{Cu K}\alpha$ radiation after hydro-separation (Fig. 2b), but weak low-angle quartz peaks were still detected by high-energy synchrotron XRD at 106.928 keV (Supplementary Fig. 3).

The hydro-separated concentrate was then reduced in 100% H_2 at $850\text{ }^\circ\text{C}$, producing porous, metallic iron granules (Fig. 2e). X-ray diffraction (XRD) showed that the iron oxide peaks disappeared after hydrogen reduction and that ferrite peaks dominated the reduced powder (Fig. 2b), indicating complete metallization. However, ICP analysis showed that residual impurity elements remained. Because oxygen in iron oxides was removed during reduction, the apparent Si concentration in the reduced iron increased to 0.75 wt% as a result of the overall mass loss, corresponding to $\sim 1.6\text{ wt}\%$ SiO_2 . Thus, even after physical beneficiation and hydrogen reduction, the ore-derived iron powder retained a significant silica-derived impurity burden. If this powder is consolidated, the residual silica would correspond to $\sim 4.7\text{ vol}\%$ brittle oxide inclusions in the steel matrix, motivating the in situ impurity-valorization strategy described below.

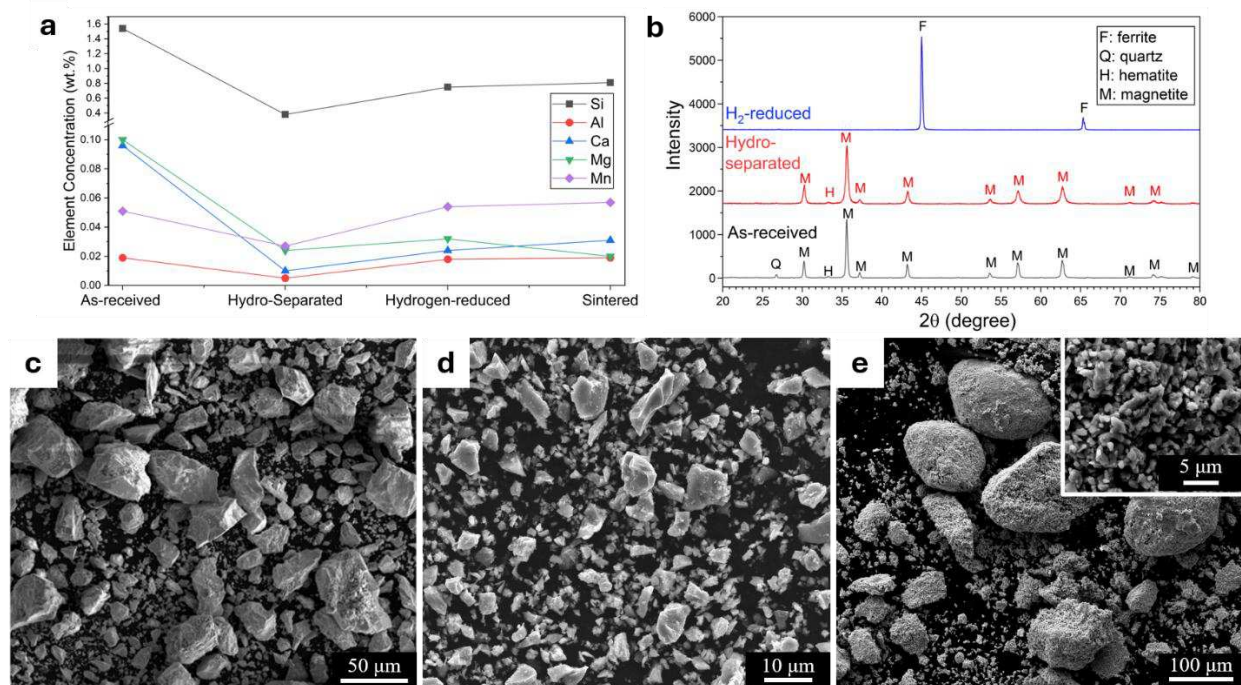


Fig. 2 | Chemical compositions, phases, and morphologies of materials at various stages of the HRMLES process.

a, Concentrations of major impurity elements determined by ICP. **b**, XRD patterns of the as-received concentrate, hydro-separated concentrate, and hydrogen-reduced iron powder. The as-received concentrate shows clear quartz (silica, labeled “Q”) peaks; after hydro-separation, no silica peaks are observed; after hydrogen reduction, only iron (ferrite, labeled “F”) peaks are observed. Hematite and magnetite phases are labeled “H” and “M”, respectively. **c**, SEM image of as-received iron ore concentrate. **d**, SEM image of hydro-separated concentrate. **e**, SEM image of hydrogen-reduced iron particles (inset shows primary particles in the granules).

Carbothermic Reduction of Silica in the Solid State

To investigate the stability and reducibility of residual silica, the H₂-reduced iron powder (containing ~1.6 wt% SiO₂) was analyzed using thermo-gravimetric analysis (TGA) and differential scanning calorimetry (DSC) under three different conditions: flowing Ar, 10% H₂/90% Ar, and flowing Ar after mixing with 1 wt% graphite.

In the first control experiment, the powder was heated under flowing argon. The material proved thermally stable, with the TGA curve (Fig. 3a, green line) showing no significant weight change. The corresponding DSC curve (Fig. 3b, green line) revealed only the expected endothermic events for the iron matrix: the Curie transformation at 771 °C and the α -ferrite (BCC) to γ -austenite (FCC) phase transformation at 934°C. This experiment serves as a critical baseline, establishing 934°C as the reference transformation temperature for the unalloyed iron at the testing heating rate (20 °C/min). The sample was cooled to 500 °C after holding at 1200 °C for 30 minutes, and then reheated to 1200 °C. The transformation temperature remained unchanged on reheating,

establishing 934 °C as the reference $\alpha \rightarrow \gamma$ transformation temperature for the reduced iron powder at this heating rate. Any deviation from 934 °C in subsequent tests provides direct evidence of possible solid-solution alloying. SEM-EDS mapping after the test showed residual Si-O-rich particles (Fig. 3c), confirming that silica remained stable under Ar.

In the second experiment, the powder was heated under a 10% H₂/90% Ar atmosphere using the same temperature-time profile. This condition was included because the reduced powder may partially reoxidize during handling and crushing. The sample showed a small initial mass loss of 0.16 wt%, attributed to reduction of surface iron oxides introduced during handling, followed by a slow total mass loss of ~0.60 wt% by 1200 °C (Fig. 3a, red line). These data indicate that hydrogen can partially reduce residual silica under the present conditions, but the extent of reduction is limited. Consistent with this, EDS mapping of the post-test sample (Fig. 3d) showed that, despite some evidence of partial reduction, distinct silica particles remained prevalent. The Ellingham diagram (Fig. 1a) suggested that the reduction of SiO₂ with hydrogen is thermodynamically unfavorable under H₂O/H₂=1. The sluggish reduction observed here is likely enabled by the combination of a lowered H₂O partial pressure and a decrease in silicon activity as silicon dissolves into iron. A slight increase in the α to γ transformation temperature on the second heating ramp (to 942 °C, red line in Fig. 3b) indicates a small amount of silicon dissolved into the iron, consistent with the report that dissolved silicon in iron stabilizes ferrite and thus increases the austenitizing temperature²⁵.

In the third experiment, the powder was mixed with 1 wt% graphite and heated under Ar. In contrast to the two control conditions, this sample showed a pronounced two-stage mass loss (Fig. 3a, blue line). The first stage began at ~721 °C and corresponded to a mass loss of 0.28 wt%, consistent with reduction of residual iron oxides by carbon. The second stage began at ~830 °C and produced a much larger mass loss of 1.79 wt%, accompanied by a broad endothermic peak in the first heating cycle (Fig. 3b, blue line). This reaction, absent in the Ar-only and H₂-containing control experiments, is the carbothermic reduction of SiO₂.

These results show that solid carbon enables silica reduction at temperatures far below those used in conventional silicon production (1900–2100 °C) and below the temperatures typical of blast-furnace reactions. Furthermore, the heat-flow curve of the second heating showed a strong exothermic peak starting at ~756 °C, corresponding to the α -ferrite + cementite $\rightarrow$ γ -austenite eutectoid transformation. The appearance of this peak, combined with the weakening of the α to γ transformation at higher temperatures, indicates that both the added carbon and the newly reduced silicon have dissolved into the iron matrix, transforming the material into a Fe-Si-C alloy. EDS mapping of the tested sample (Fig. 3e) supported the TGA and DSC results. No obvious silicon-rich areas were detected, indicating the silica particles had been fully reduced and their constituent silicon dissolved into the iron.

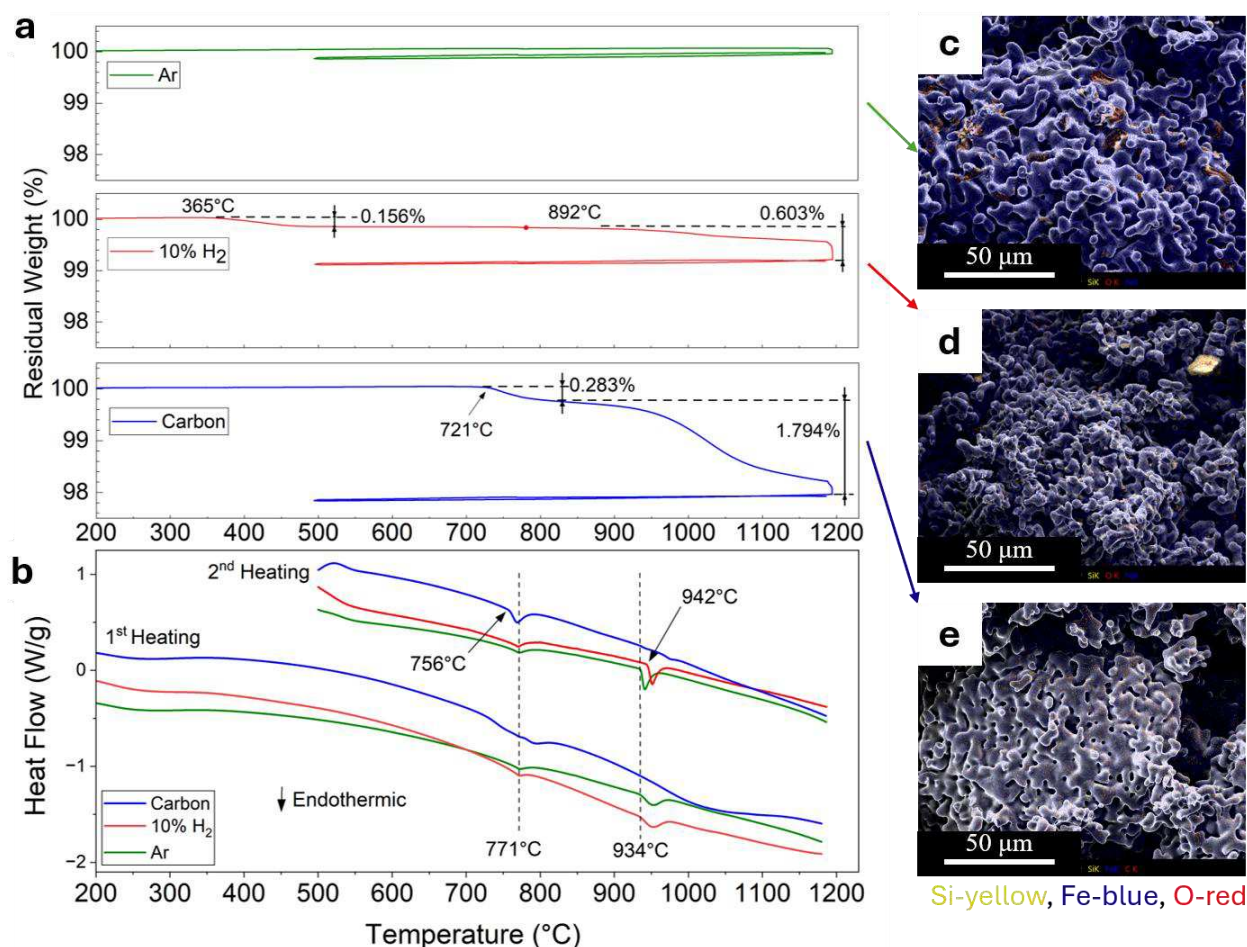


Fig. 3 | Heating hydrogen-reduced iron powder under different conditions.

a, TGA curves and **b**, DSC curves for the sample heated in pure Ar (green curves), the sample heated in 10% H₂ (red curves), and the sample with 1 wt% carbon heated under Ar (blue curves). **c**, SEM-EDS map of sample after heating in Ar, showing residual silica particles (yellow). **d**, SEM-EDS after heating in 10% H₂, showing partial SiO₂ reduction at particle surfaces. **e**, SEM-EDS after heating with carbon, showing no remaining silica particles (silicon signal is uniformly distributed in the iron matrix). All three samples were heated from room temperature to 1200 °C and held for 30 minutes. After cooling to 500 °C, they were reheated to 1200 °C.

Direct Observation of the In Situ Alloying Pathway

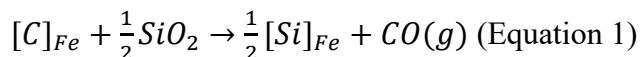
In conventional blast furnace ironmaking, silica is present in both the iron ore pellets and coke. At temperatures up to 1500 °C, a fraction of this silica is reduced by carbon and dissolves into the molten iron, producing liquid iron containing 0.3-0.9 wt% Si and 3-5 wt% C²⁶, while most silica is separated into the slag. Thermodynamically, the direct reduction $\text{SiO}_2 + 2\text{C} = \text{Si} + 2\text{CO(g)}$ is unfavorable below 1650 °C under standard conditions (Fig. 1a). Conventional high-temperature pathways are thus believed to involve gaseous intermediates or slag-metal reactions. Proposed mechanisms include²⁶: (1) the formation of silicon monoxide gas, which then either

disproportionates or is further reduced to dissolve silicon into the iron, $\text{SiO}_2 + \text{C} = \text{SiO}(\text{g}) + \text{CO}(\text{g})$; or (2) the direct reduction of SiO_2 from the slag by carbon dissolved in the liquid iron, $\text{SiO}_2 (\text{slag}) + 2[\text{C}]_{\text{Fe}} (\text{l}) = [\text{Si}]_{\text{Fe}} (\text{l}) + 2\text{CO}(\text{g})$.

The unusually low reduction temperature observed in our TGA-DSC analysis suggests a different, solid-state reaction mechanism. To directly observe this pathway, we performed in situ SEM heating on a compacted granule consisting of HRMLES particles and graphite particles. Before heating, EDS was used to identify SiO_2 , graphite, and iron particles. Backscattered-electron imaging was then used to track the same region during heating to 1200 °C and subsequent holding (the full video sequence is provided in the Supplementary Information).

At room temperature, the granule is a compacted mechanical mixture of iron, graphite, and silica particles (Fig. 4a and b). Two microscale silica particles (rich in Si and O, marked by the green ellipse) are identified, spatially separated from distinct graphite particles (rich in C, marked by the red ellipse). During heating to 1200 °C (Fig. 4c and d), the graphite particles in the red ellipse begin to disappear as they dissolve into the surrounding iron particles (Fig. 4d). After holding at 1200 °C for 23 seconds, all graphite particles had disappeared (Fig. 4e). In contrast, before graphite dissolution is complete, the silica particles in the green ellipse remain at the same location and retain a similar morphology, while adjacent iron particles begin to sinter together. For example, the small iron particles marked by the yellow circle coalesce with surrounding particles, leaving a rounded pore (Fig. 4c,d). This sequence provides direct morphological evidence for the first step of the reaction, i.e., solid-state dissolution of carbon into iron $\text{C}(\text{s}) + \text{Fe}(\text{s}) \rightarrow [\text{C}]_{\text{Fe}} (\text{s})$, before silica reduction occurs. After holding at 1200 °C for 1,564 seconds, the previously visible silica particles are no longer observed (Fig. 4f), consistent with reduction of silica and the subsequent dissolution/homogenization of silicon within the surrounding iron matrix. The tracked region also becomes more densified, indicating simultaneous sintering and in situ alloying.

The in situ SEM experiment shows a higher apparent reduction temperature than the TGA–DSC experiment, likely because of thermal lag between the heating element and the porous powder compact during in situ heating. Nevertheless, the experiment directly reveals the sequence of events underlying the solid-state alloying pathway. First, solid carbon dissolves into the iron (forming $[\text{C}]_{\text{Fe}}$). Second, this dissolved carbon is transported through the iron matrix to react with the solid silica particle, reducing it. This reaction pathway was also confirmed by the interrupted sintering study (Supplementary Fig. 4). The gaseous CO product is removed by the vacuum system or flowing inert gas, driving the reaction forward. The iron matrix itself acts as both a solvent for carbon and silicon and a transport medium for carbon. The overall reaction is:



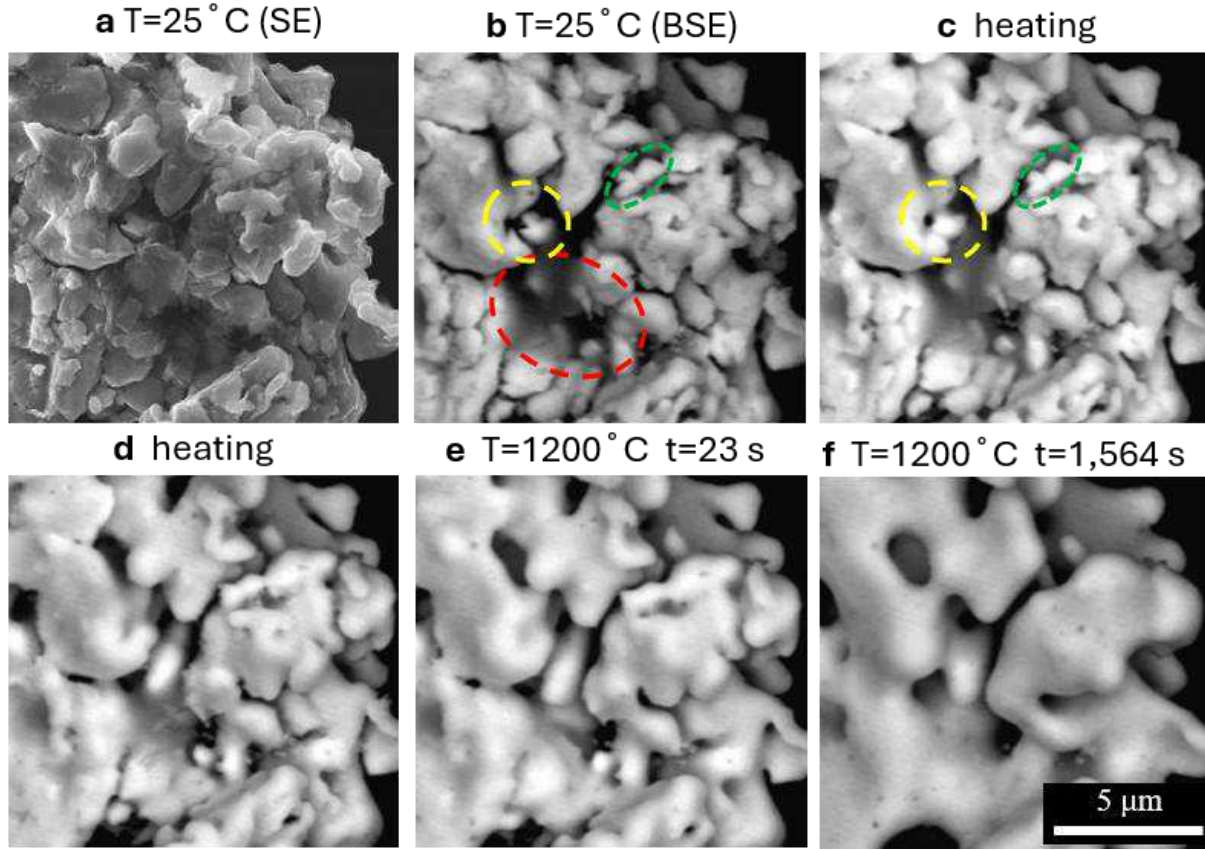
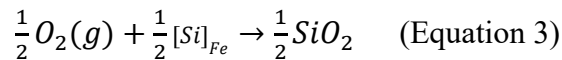
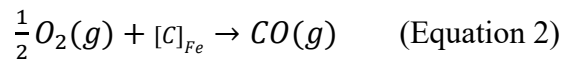


Fig. 4 | In situ SEM investigation showing the stepwise in situ reduction of silica particles by dissolved carbon.

a, Secondary electron image and b, backscattered-electron image of a compacted granule (die-pressed powder with 1 wt% graphite and crushed to granules) before heating: dark particles in the red ellipse are graphite; the particles in the green ellipse are silica identified by EDS; and the bright particles in the yellow circle are iron particles. c and d, backscattered-electron images of the granule during heating to 1200 °C, and after holding at 1200 °C for (e) 23 s and (f) 1,564 s.

Thermodynamic modeling of the reduction of SiO₂ with C dissolved in Fe

The ability to reduce silica at a moderate temperature can be explained based on the thermodynamics of the reaction between the dissolved carbon in Fe and SiO₂. To explore this, we performed thermodynamic calculations to predict the conditions under which carbon can reduce SiO₂ when both carbon and silicon are dissolved in γ -Fe (FCC) at 800–1200 °C. The overall reaction (Equation 1) can be conceptually split into two reactions of oxide formation:



Specifically, we consider the oxidation of dissolved carbon to CO (Equation 2) and the oxidation

of silicon to SiO₂ (Equation 3). The oxygen potential for each reaction as a function of the activities of carbon and silicon dissolved in γ -Fe and CO partial pressure can be expressed as:

$$RT \ln p_{O_2(g)}^{1/2} = G_{CO(g)}^0 - \left(\frac{1}{2} G_{O_2(g)}^0 + G_C^0 \right) + RT \ln \left(\frac{p_{CO(g)}}{a_C} \right) \quad (\text{Equation 4})$$

$$RT \ln p_{O_2(g)}^{1/2} = \frac{1}{2} G_{SiO_2}^0 - \left(\frac{1}{2} G_{O_2(g)}^0 + \frac{1}{2} G_{Si}^0 \right) + \frac{1}{2} RT \ln \left(\frac{1}{a_{Si}} \right) \quad (\text{Equation 5})$$

In Equations 4 and 5, the left-hand side represents the oxygen chemical potential normalized to one mole of O, G_i^0 is the Gibbs energy of the species i , and a_i is the activity of the species. The standard Gibbs energy functions for the pure elements were taken from the SGTE pure substances database²⁷, using the solid graphite phase for C and the diamond-cubic solid phase for Si as reference states. Gibbs energy functions for SiO₂ (tridymite) and CO were obtained from the HSC Chemistry 9 database. The Gibbs energy expressions on the right side of the equation simplify to the Gibbs energy of formation for the oxides, reducing the expression for the oxygen potentials to:

$$RT \ln p_{O_2(g)}^{1/2} = \Delta G_{f,CO(g)}^0 + RT \ln \left(\frac{p_{CO(g)}}{a_C} \right) \quad (\text{Equation 6})$$

$$RT \ln p_{O_2(g)}^{1/2} = \frac{1}{2} \Delta G_{f,SiO_2}^0 + \frac{1}{2} RT \ln \left(\frac{1}{a_{Si}} \right) \quad (\text{Equation 7})$$

The Gibbs free energy changes for the formation of SiO₂ and CO (from Si and C dissolved in γ -Fe) were then calculated and are presented in Fig. 5. As shown in Fig. 5a, the oxygen potential (free energy of oxide formation) of 1 wt% C dissolved in γ -Fe decreases significantly as the partial pressure of CO is lowered. In other words, the dissolved carbon becomes a stronger reducing agent when the CO partial pressure is decreased. For silicon, dissolving Si in γ -Fe dramatically increases the oxygen potential of SiO₂, thus destabilizing SiO₂ in the iron matrix. The combined effect of lowering the CO partial pressure and dissolving Si in γ -Fe is that even 2.14 wt% silica (i.e., 1 wt% Si in Fe) can be thermodynamically reduced by 1 wt% C in γ -Fe at relatively low temperatures (>900 °C) and at CO partial pressures below ~100 Pa.

We calculated the thermodynamic properties of our experimental system, and the results are plotted in Fig. 5b. Fixing $P_{CO} = 5$ Pa (the vacuum level in sintering) at 1200 °C, we find that CO in equilibrium with only ~0.01 wt% C is more stable than SiO₂ in equilibrium with 0.8 wt% Si in γ -Fe. In other words, at 1200 °C under 5 Pa vacuum, after the silica is reduced (yielding ~0.8 wt% Si in the iron), the reaction remains thermodynamically favorable as long as a stoichiometric excess of carbon is present to capture the oxygen as CO. Practically, all of the 1.7 wt% SiO₂ in the iron can be fully reduced by dissolved carbon, provided that sufficient carbon is available to bind the oxygen into CO. This result is consistent with Thermo-Calc equilibrium predictions (Supplementary Fig. 8) made with the TCFE14 database (Thermo-Calc 2025a).

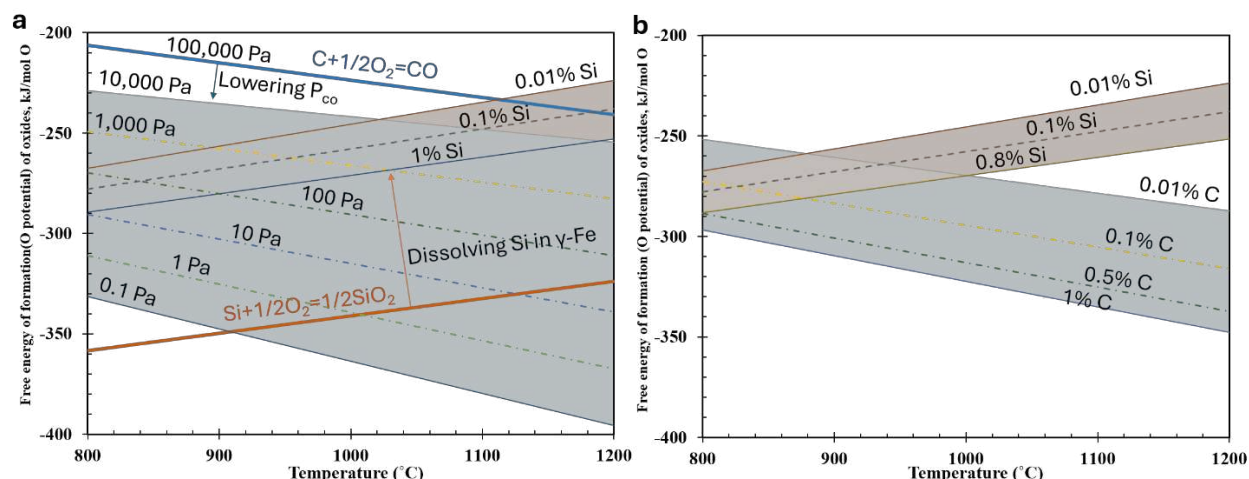


Fig. 5 | Thermodynamic calculations for silica reduction.

a, Ellingham diagram illustrating the oxidation of 1 wt% C (dissolved in γ -Fe) to CO under varying CO partial pressures, and the oxidation of Si (dissolved in γ -Fe) to SiO_2 for different dissolved Si contents. For comparison, the lines for oxidation of pure carbon at 10^5 Pa CO and pure Si are also shown. **b**, Ellingham diagram showing the effect of dissolved carbon content on the oxygen potential of CO at a fixed CO partial pressure of 5 Pa, compared to the oxygen potential of SiO_2 for different Si contents dissolved in γ -Fe. These plots illustrate that lowering the CO partial pressure and dissolving Si in iron both shift the thermodynamics in favor of silica reduction.

A Thermo-Kinetic Conundrum: The Role of Pore Closure

The effect of carbon content on this new reduction and sintering process was also experimentally investigated. However, it revealed an unexpected trend. As shown in Fig. 6a (Supplementary Table 2), increasing the initial graphite addition from 0 to 1.2% behaves as expected: the final oxygen content steadily drops (from 1.20% to 0.106%) as oxides are reduced, and the final sintered density concurrently increases, reaching a peak of 7.27 g/cm³. This behavior is consistent with the increased activity of dissolved carbon, which promotes the reduction of residual oxides and improves densification.

However, a clear anomaly occurs at carbon additions above 1.2%. While the final oxygen content remains minimal (e.g., 0.098% O at 1.6% C), the final density paradoxically decreases, falling from its peak of 7.27 g/cm³ back down to 7.14 g/cm³. This is also visualized in the final microstructures (Supplementary Fig. 5), where the porosity visibly increases in the 1.4% and 1.6% C samples compared to the 1.2% C sample. This trend is counterintuitive from a purely thermodynamic perspective: higher carbon activity should further promote oxide reduction and, by removing oxide barriers, assist densification. The density decrease at excess carbon addition therefore indicates that the process is not controlled by equilibrium thermodynamics alone.

The residual oxygen in Fig. 6a indicates that oxide reduction remains incomplete in some regions. This is confirmed by transmission electron microscopy (TEM) and EDS analysis of the final

sintered samples (Fig. 6b and Supplementary Fig. 7), which reveals the persistence of unreacted, microscale and nanoscale inclusions, identified as both quartz (SiO_2) and magnesium silicate (MgSiO_3). Thus, even when sufficient carbon is available and the overall reduction is thermodynamically favorable under low external P_{CO} , some impurity oxides remain unreduced. Moreover, the excess carbon appears to hinder densification. The observation of MgSiO_3 also indicates that the residual inclusions are not limited to pure silica. ICP analysis detected trace Ca and Al (Fig. 2a), and because CaO and Al_2O_3 are more stable than SiO_2 , other complex silicates, such as calcium- or aluminum-containing silicates, may also persist under the present sintering conditions.

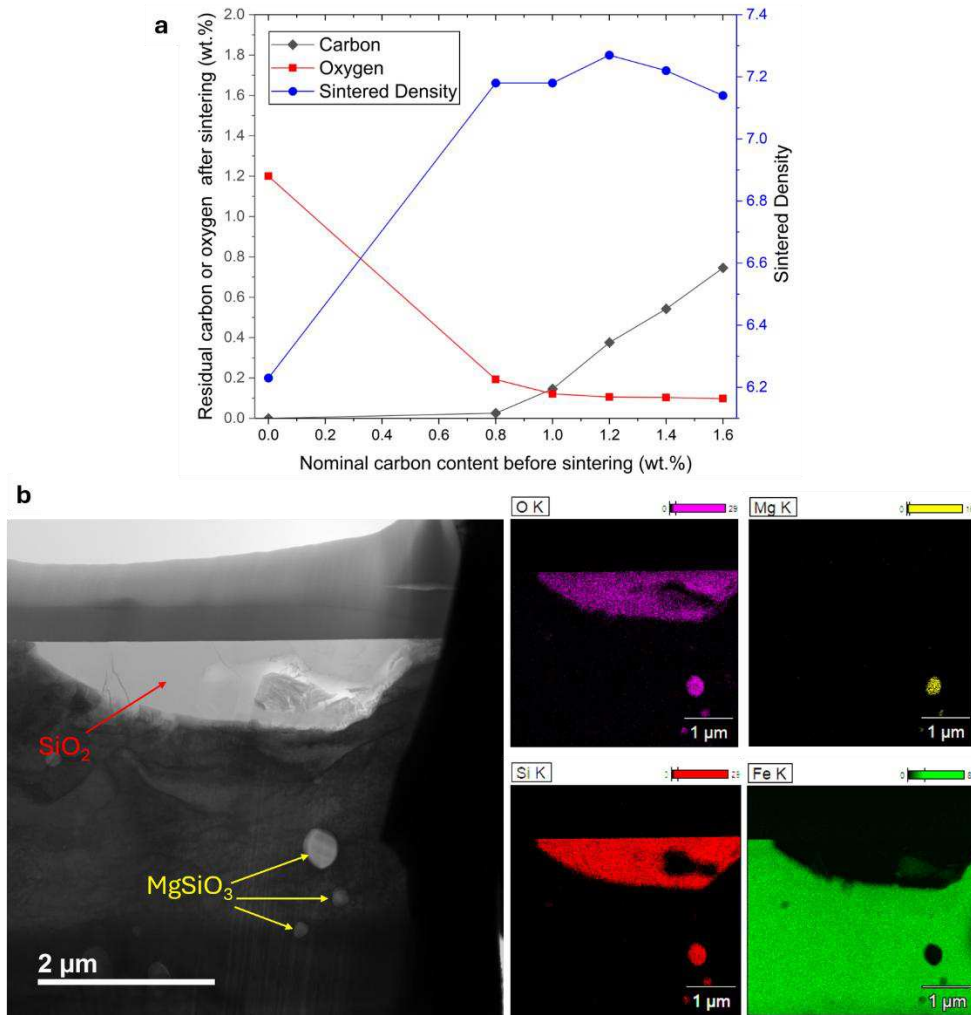


Fig. 6 | Effect of carbon addition on oxygen removal, densification, and residual inclusions after sintering.

a, Residual carbon content, oxygen content, and sintered density as a function of the nominal carbon content added before sintering. Increasing carbon addition reduces the final oxygen content and increases the sintered density up to an optimum carbon level, beyond which the density decreases despite the low residual oxygen content. **b**, STEM-EDS elemental maps (Fe, Si, O, Mg) of a sintered sample with 1.2 wt% carbon addition, showing persistent inclusions of SiO_2 (quartz) and MgSiO_3 (magnesium silicate) trapped in the iron matrix.

We attribute this discrepancy to a thermo-kinetic trap created by the competition between the gas-producing silica-reduction reaction (Equation 1) and the kinetics of pore closure during sintering. An ex situ sintering study reveals the critical microstructural and pore change. The green compact (Fig. 7a) consists of densified granules with small intra-granular pores, separated by large inter-granular voids. After heating to 800 °C (Fig. 7b), initial neck formation between iron particles begins, but the overall pore network remains open and continuous. At 1000 °C (Fig. 7c), two distinct pore environments emerge: some larger pores remain connected to the external atmosphere (indicated by the red dashed outlines), whereas smaller pores become isolated within the densifying iron matrix (indicated by the blue dashed outlines). This distinction persists after heating to 1200 °C (Fig. 7d), where open-pore and closed-pore regions coexist.

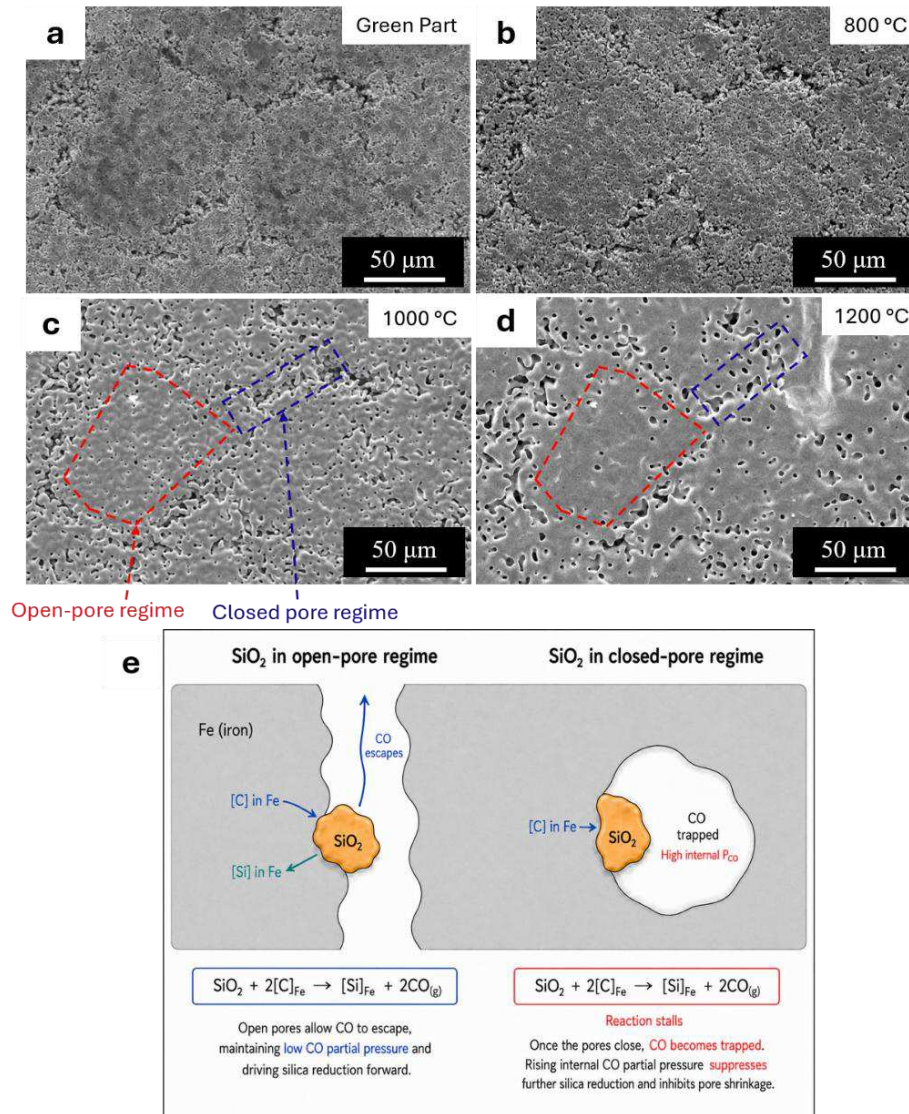


Fig. 7 | The role of pore closure in the reduction of SiO₂.

a–d, SEM images showing the evolution of pore morphology during sintering of the compact containing 1 wt% graphite: **a**, green compact before sintering; **b**, after heat treatment at 800 °C; **c**, after heat treatment

at 1000 °C; and d, after heat treatment at 1200 °C. Large inter-granular pores remain connected to the external atmosphere up to ~1000 °C, whereas smaller intra-granular pores become sealed during sintering, forming isolated gas pockets. Blue and red dashed outlines in **c** and **d** indicate representative open-pore and closed-pore regimes, respectively. **e**, Schematic illustrating the effect of pore closure on solid-state carbothermic reduction of SiO₂. In the open-pore regime, SiO₂ in contact with iron containing dissolved carbon is reduced while CO escapes through connected pores, maintaining a low CO partial pressure and driving the reaction forward. In the closed-pore regime, CO generated by SiO₂ reduction becomes trapped in isolated pores, increasing the internal CO partial pressure, suppressing further reduction, and inhibiting pore shrinkage.

This pore evolution creates two distinct reaction regimes, schematically illustrated in Fig. 7e. In the open-pore regime, SiO₂ particles that are in physical contact with iron containing dissolved carbon can be reduced according to Equation 1. The Si produced by the reaction dissolves into the surrounding Fe matrix, while the CO product escapes through connected pores. Continuous CO removal maintains a low local P_{CO}, preserving the thermodynamic driving force for silica reduction. In the closed-pore regime, SiO₂ may still contact the surrounding Fe matrix and react initially with dissolved carbon, but the CO product becomes trapped once the pore is sealed. As reduction proceeds, the internal P_{CO} rises. Our thermodynamic calculations show this reaction is extremely sensitive to P_{CO}: as the internal pressure approaches ~10³-10⁴ Pa, the Gibbs free energy becomes positive ($\Delta G > 0$), and the silica reduction stops.

The same pore-closure mechanism also explains the decrease in density at carbon additions above 1.2 wt% (Fig. 6a and Supplementary Table 2). Higher carbon content increases the activity of dissolved carbon and accelerates CO generation. In open pores, this promotes oxide reduction because CO can escape. In closed pores, however, the generated CO accumulates, raising the internal gas pressure. This pressure opposes pore shrinkage and can inhibit densification, leading to the observed density loss at high carbon additions. The competition between open-pore CO removal, closed-pore CO trapping, and sintering-driven pore closure therefore defines a critical processing window for maximizing impurity reduction while avoiding density loss.

Tensile Properties of HRMLES Steels

We next evaluate whether impurity valorization can produce mechanically useful steels directly from ore-derived powder. With 1.2 wt% graphite addition, HRMLES produced an Fe-0.8Si-0.4C alloy. In the as-sintered state, this alloy exhibited a ferrite-pearlite microstructure (Fig. 8a) and achieved an ultimate tensile strength (UTS) of 516 MPa, a yield strength of 204 MPa, and 14% elongation. To demonstrate the versatility of the HRMLES process for in situ alloying, we also added 3.1% MnO₂ powder along with 1.74 wt% graphite in another experiment. The MnO₂ was reduced during sintering and distributed uniformly in the steel, as shown in Supplementary Fig. 6, producing a final alloy of Fe-2Mn-0.8Si-0.2C. In its as-sintered state, this alloy showed a refined ferrite-pearlite structure (Fig. 8b) with a UTS of 560 MPa, a yield strength of 346 MPa, and 16.7% elongation. After water-quenching from 780 °C, it was converted to a ferrite-martensite dual-phase microstructure (Fig. 8c), increasing the UTS to 831 MPa and the yield strength to 426 MPa while

retaining 6.4% elongation.

The tensile properties of the HRMLES steels compare favorably with representative property ranges for conventional PM steels and wrought carbon structural steels (Fig. 8d and Supplementary Table 3). In particular, the as-sintered Fe-0.8Si-0.4C and Fe-2Mn-0.8Si-0.2C alloys show substantially higher elongation than two widely used PM steel grades, Fe-2Cu-0.8C (FC-0208) and Fe-2Ni-0.5C (FN-0205), while also achieving comparable or higher strength. The as-sintered Fe-2Mn-0.8Si-0.2C alloy falls within the tensile property range of widely used high-strength C-Mn structural steels (defined as steels with yield strengths greater than 275 MPa²⁸). After quenching, the same alloy reaches a strength level above 800 MPa, demonstrating that conventional heat treatment can further enhance the mechanical performance of HRMLES steels. These results show that HRMLES can directly produce high-strength steels from impurity-containing iron concentrate without melting. They also confirm that silica-derived silicon and externally added MnO₂ can both be incorporated through solid-state alloying.

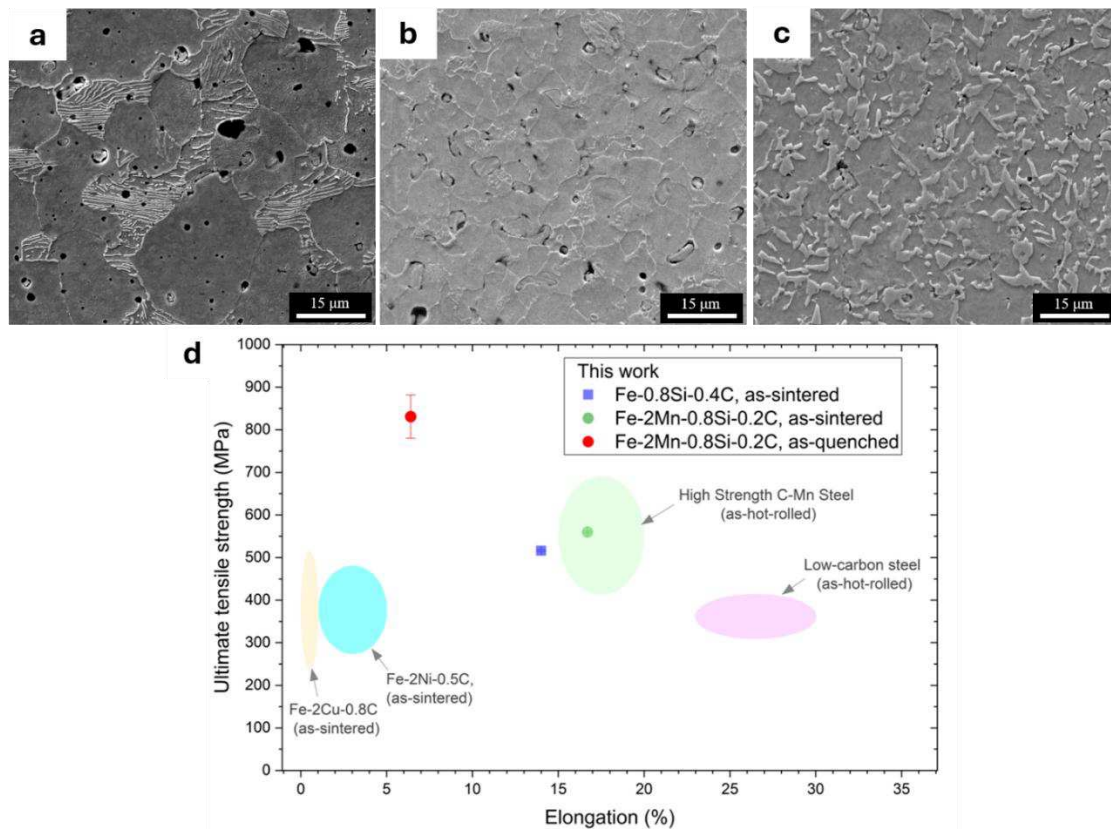


Fig. 8 | Microstructures and tensile properties of HRMLES steels.

a-c, SEM micrographs of HRMLES steels: **a**, as-sintered Fe-0.8Si-0.4C; **b**, as-sintered Fe-2Mn-0.8Si-0.2C; and **c**, Fe-2Mn-0.8Si-0.2C after quenching, showing a dual-phase ferrite-martensite microstructure. Scale bars, 15 μm. **d**, Ultimate tensile strength as a function of elongation for HRMLES steels, compared with representative property ranges of as-hot-rolled low-carbon steels²⁸, as-hot-rolled high-strength C-Mn steels²⁸, and commonly used powder-metallurgy steels FC-0208 and FN-0205²⁹. Error bars indicate the

standard deviation of tensile tests where shown. (For the as-sintered HRMLES conditions, the standard deviations in ultimate tensile strength are only 2 MPa and are therefore smaller than the symbols in the plot.) Exact tensile properties are summarized in Supplementary Table 3.

Discussion

The mechanical properties reported here are those of sintered materials without post-sintering mechanical working, and therefore reflect the influence of residual porosity. Such porosity can limit the mechanical potential of steels, particularly ductility, toughness, and fatigue resistance. At the same time, the powder-based HRMLES route is compatible with near-net-shape manufacturing, enabling complex geometries and high material utilization while reducing the material losses associated with conventional deformation and machining. The as-sintered Fe-Mn-Si-C alloy already reaches tensile properties comparable to representative hot-rolled C-Mn steels in strength and elongation. For more demanding applications, secondary thermomechanical processing such as powder forging, hot rolling or extrusion could be used to close residual porosity, increase density, and further improve mechanical performance. Thus, the present results should be viewed as a baseline for meltless ore-to-steel production, with additional opportunities for property optimization through densification and heat treatment.

In HRMLES, the primary reduction of iron ore is conducted using hydrogen. If this hydrogen and the required electricity are supplied from renewable sources, the direct process emissions are dominated by the small amount of carbon consumed during sintering to reduce residual oxides. We can quantify this for the Fe-2Mn-0.8Si-0.2C alloy: to chemically reduce the oxides required to produce 0.8 wt% Si and 2.0 wt% Mn, approximately 1.3 moles of carbon are consumed per 1 kg of steel. This reaction generates CO, equivalent to only 0.057 tonne CO₂ per tonne of steel. This value is far below the direct emissions associated with conventional BF-BOF steelmaking, although a complete life-cycle assessment must also account for hydrogen production, electricity supply, ore preparation, and downstream processing.

We also estimated the total energy consumption for manufacturing a representative product (automotive connecting rod, a typical product that is produced by wrought and PM routes^{30,31}) using the HRMLES route versus a conventional route (machining drop-forged BF-BOF steel). As shown in Fig. 9, approximately 18.3 GJ of energy is required to produce 1 tonne of connecting rods via HRMLES, which is roughly 46% lower than the energy required using the conventional BF-BOF route, and 48% lower than that for the H₂ DRI-EAF route. It is noteworthy that more than half of this energy (~55%) in the HRMLES route is attributable to hydrogen production via water electrolysis (assuming renewable-powered electrolysis). This highlights the importance of low-cost, low-carbon hydrogen for realizing the energy and emissions benefits of the process. Because HRMLES combines lower processing temperatures with near-net-shape material utilization, it may offer cost advantages for selected components; however, a full techno-economic analysis is required to quantify capital costs, electricity and hydrogen prices, equipment utilization, yield

losses, and product-specific processing costs.

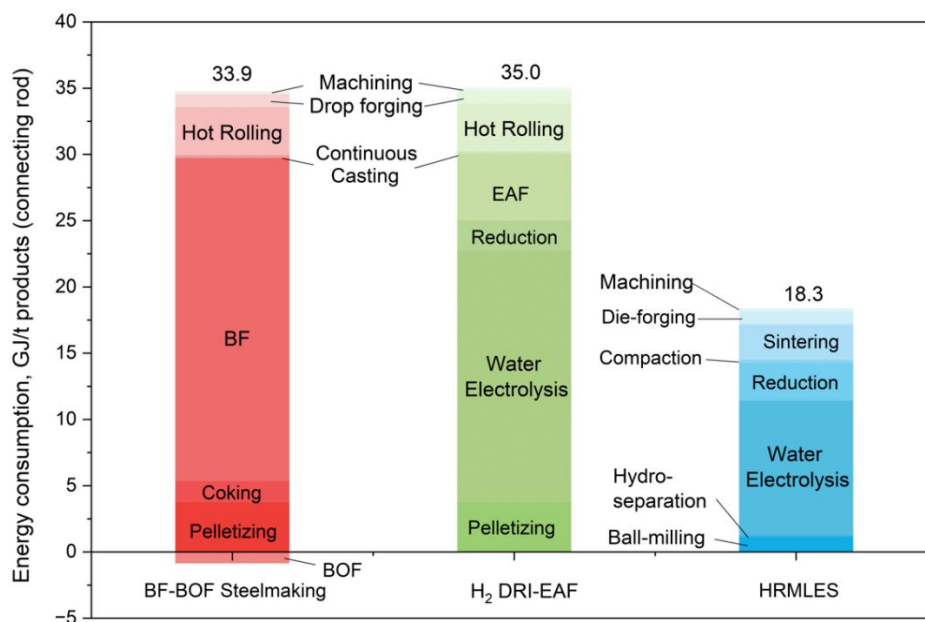


Fig. 9 | Estimated energy consumption for manufacturing connecting rods using the HRMLES process, compared to machining drop-forged BF–BOF steel and H₂ DRI-EAF preforms.

The analysis indicates a ~46% energy saving by using HRMLES with hydrogen from water electrolysis. Energy consumption data for BF-BOF steelmaking were collected from Ref³². Energy consumption data for H₂ DRI and EAF were collected from Ref³³. The material utilization rates are 50% for BF-BOF and H₂ DRI (HYBRIT)-EAF and 90%³⁰ for HRMLES, due to its near-net-shape compatibility.

In summary, this work presents a solid-state steelmaking strategy that addresses a key impurity challenge in powder metallurgy routes based on ore-derived iron powder. We have identified and demonstrated a low-temperature chemical pathway for the carbothermic reduction of solid silica, a ubiquitous and problematic impurity in iron ores. The core of this discovery is a two-step mechanism, confirmed by direct observation, where the solid iron matrix itself acts as a solvent and diffusion medium, first dissolving solid carbon and then using that dissolved carbon to reduce solid silica. We have also defined the critical thermo-kinetic boundaries of this process, where the competition between pore closure and the CO-producing reduction reaction dictates the final density and purity. Because residual inclusions and pores can affect fatigue crack initiation and other damage processes, future work should focus on optimizing the processing window to promote silica reduction while avoiding premature pore closure. The influence of residual silicates and porosity on final mechanical properties, particularly fatigue and fracture behavior, also warrants systematic investigation. Because this study uses a silica-dominant taconite-derived concentrate, further work is needed to evaluate the feasibility of HRMLES for other iron ores with different gangue chemistries and mineralogies, particularly those containing higher levels of more stable oxides such as Al₂O₃, CaO or MgO.

More broadly, this study suggests that selected gangue impurities can be converted from processing liabilities into useful alloying sources under controlled solid-state conditions. In the HRMLES process, silica serves as an in situ source of silicon, coupling the reduction of a detrimental inclusion with the addition of a beneficial strengthening element. The approach therefore expands the range of reaction and alloying pathways available for primary steel production without requiring melting. By demonstrating that iron ore can be converted into steel entirely in the solid phase, we challenge the long-held notion that a molten phase is indispensable. The ability to bypass melting could not only reduce emissions and energy demand but also grant access to previously unviable chemical reactions and alloying strategies. These findings have broad implications. Using a primary metal matrix as a medium to drive impurity-to-alloy transformations could be applied beyond iron, opening a pathway for sustainable metallurgy in other alloy systems.

Methods

The iron ore concentrate was further purified using a customized hydro-separator and reduced in 100% H₂ at 850 °C. The evaluation of the overflow material and the purified concentrate after hydro-separation are shown in Supplementary Fig. 2. The reduced iron powder was mixed with graphite (0–1.6 wt%) and die-pressed. Sintering was conducted in a ceramic-tube furnace under vacuum (1–10 Pa). Thermal analysis was performed using TGA-DSC. Microstructural and chemical analyses were conducted using SEM, STEM, XRD, and ICP-MS. Carbon and oxygen contents were determined by combustion/inert gas fusion. Mechanical testing was performed on sintered rods.

Materials

Iron ore concentrate provided by ArcelorMittal was used as the feed material for this study. It was beneficiated from taconite mined in Mesabi Iron Range, USA. Graphite powder purchased from ALFA Chemistry (purity >99%) and MnO₂ purchased from Thermo Fisher Scientific (>99.9% purity, -325 mesh) were used in this study.

Hydro-separation Process

The as-received concentrate, with a median particle size of 29.2 μm, was first ball-milled in an attritor mill with water to fine particles with a median particle size of 4.0 μm. This milling liberates the fine gangue particles from the iron ore. The milled slurry, containing 60 g of solids, was fed to a 1-liter flask (shown schematically in Supplementary Fig.1). A magnetic stir bar was placed inside and set to a speed of 1000 rpm. Water was introduced for 1 hour, and a vertical outlet tube was attached at the top neck for removal of the low-density overflow. The hydro-separated particles were pressed and crushed to make -100 mesh granules after drying.

Hydrogen Reduction

The ore granules were reduced in a tube furnace with 100% hydrogen flow at 850 °C for 30 minutes with a hydrogen flow of 4 L/min. After hydrogen reduction, the iron granules were slightly sintered together, so they were crushed with a mortar and pestle to recover the granules to powder form.

Sintering and Alloying

The reduced iron granules were mixed with different amounts of graphite powder (0–1.6 wt%) for 30 minutes. For the carbon-variation experiments, the powder mixtures were die-pressed into disk-shaped green compacts with a diameter of 15 mm and a thickness of ~6 mm under a pressure of 440 MPa. The green compacts were then loaded into a ceramic-tube furnace and sintered at 1200 °C for 2 h under vacuum (1–10 Pa), generated using a rotary vane pump. For Mn-containing alloys, MnO₂ powder and graphite were premixed with the hydrogen-reduced iron powder before compaction and sintering, as described below for the tensile specimens.

Characterization

After sintering, samples were sectioned and polished for microstructural characterization. The density of as-sintered disks was determined using the geometric method. Microstructures and elemental distributions were characterized using an FEI Quanta 500 SEM equipped with EDS and a JEOL JEM 2800 STEM equipped with EDS. Phase identification was performed using a Rigaku MiniFlex 600 X-ray diffractometer with Cu K α radiation. High-energy synchrotron X-ray diffraction of the hydro-separated concentrate was performed at beamline 11-ID-C, Advanced Photon Source, Argonne National Laboratory, using an X-ray energy of 106.928 keV. Two-dimensional diffraction patterns were collected and integrated to obtain one-dimensional diffraction profiles.

The Si, Al, Ca, and Mg contents in the materials were determined using ICP-MS (Agilent 8900), which required complete digestion of the materials using HF. The carbon contents in powder and sintered samples were determined by a combustion method (LECO C/S 230). Oxygen contents were determined by the inert gas fusion method (TCH 600). In the oxygen determinator, (~0.1 g) iron or steel samples react with high-purity graphite powder and crucibles at a temperature of ~2150 °C to release CO and CO₂. CO and CO₂ gases were carried by ultra-high purity helium gas to infrared gas detectors to determine the amount of oxygen released from the sample. This method is commonly used to determine dissolved oxygen in metals and alloys. The accuracy of the oxygen determination is 0.025 ppm or 0.5% relative standard deviation.

To track particle morphology changes during heating, a green compact (1 wt% graphite) was die-pressed and then crushed into granules for the in situ SEM study. In situ SEM was conducted using a JEOL JSM-7800F equipped with a Protochips heating e-chip (E-FHDS-VO-10) with a maximum temperature of 1200 °C. The guaranteed thermal uniformity region of the chip consists of a 3 × 3 grid of 8 μ m holes spanning an area of 40 × 40 μ m. The temperature was ramped to 1200 °C and then held for 30 min. The chip was factory-calibrated to apply the correct voltage for accurate temperature control. However, the actual temperature of the iron sample may be lower than the

recorded temperature because the porous structure of the sample has low thermal conductivity.

For the interrupted sintering experiment shown in Fig. 7 and Supplementary Fig. 4, a die-pressed disk was first examined by SEM-EDS (FEI Quanta 600) in the green state. The sample was then heated in a tube furnace to 800 °C for 2 h under vacuum. After cooling to room temperature, the same region was re-examined by SEM-EDS. This procedure was repeated after sequential heat treatments at 1000 °C for 2 h and 1200 °C for 2 h to track pore and particle evolution at the same location.

TGA-DSC Analysis

Approximately 60–70 mg of hydrogen-reduced iron powder (either alone or premixed with 1 wt% graphite) was tested in a thermogravimetric analyzer/differential scanning calorimeter (TGA–DSC, TA Instruments SDT Q600). The samples were tested under two different atmospheres: flowing argon gas (200 mL/min) or a reducing 10% H₂ in Ar mixture (20 mL/min H₂ + 180 mL/min Ar). The temperature program for TGA–DSC was as follows: ramp from room temperature to 1200 °C at 20 °C/min, hold at 1200 °C for 30 min, cool down to 500 °C, then ramp up again to 1200 °C at 20 °C/min (to observe any changes in phase transformation temperatures upon reheating). After TGA–DSC testing, the reacted samples were examined by SEM-EDS to analyze any remaining inclusions.

Mechanical Testing

Cylindrical rod specimens (for tensile testing) were prepared by cold isostatic pressing of hydrogen-reduced iron powder mixed with 1.2 wt% graphite, followed by vacuum sintering under the same conditions (1200 °C, 2 h). For the alloyed steel, 3.1% MnO₂ powder and 1.74 wt% graphite powder were premixed with the iron powder and sintered to make samples with the target composition of Fe-2Mn-0.8Si-0.2C. After sintering, some samples of the alloyed steel were quench-hardened by a water quench from 780 °C to achieve a dual-phase microstructure. The sintered rods were machined into standard round tensile bars (with a gauge length 16.0 mm and diameter 4.0 mm). Tensile testing was performed on three specimens for each condition by an Nadcap- and ISO- accredited testing laboratory (Westmoreland Mechanical Testing & Research, Inc.) in accordance with ASTM E8. Results are reported as mean ± standard deviation.

Data Availability

The data supporting the findings of this study are available within the paper, its Supplementary Information, and the accompanying Source Data file.

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