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Editorial for special issue on hydrogen metallurgy

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1. Foreword

The carbon neutral strategy is promoting the transformation and upgrading of the traditional metallurgical industry, and the high-quality and low-carbon reform of the metallurgical industry is imperative. Hydrogen, as a clean energy in the 21st century, has a potential to realize low-carbon metallurgy from the source, and "hydrogen metallurgy" has become one of the most important directions for metallurgical industry. In recent years, the traditional metallurgical processes, represented by the steel industry, have developed rapidly in terms of hydrogen-rich smelting and new hydrogen metallurgy processes, indicating that the metallurgical industry is making rapid progress on the road of low-carbon transformation.

In order to speed up the development of low-carbon metallurgical industry, a special issue of "Hydrogen Metallurgy" is published by the journal of *International Journal of Minerals, Metallurgy and Materials* (IJMMM). Experts and scholars from the world's top universities or research institutes including the Max Planck Institute in Germany and Kyushu University in Japan were invited to share their progress and prospects in "hydrogen metallurgy" research. The special issue includes a total of 12 papers, including a review of plasma hydrogen metallurgy and 11 research papers.

The papers mainly focus on three directions: hydrogen-enriched behavior in traditional ironmaking process, hydrogen-based direct ironmaking process, and hydrogen plasma metallurgy. The special issue covers a comprehensive range of topics, from the innovation of traditional metallurgical processes to the development of new hydrogen-rich metallurgy processes, involving the current hot research directions in hydrogen metallurgy.

2. Topic A: Hydrogen-enriched behavior in traditional ironmaking process

The annual CO_2 emissions of steel industry accounts for 6.7% of the global CO_2 emissions. Among them, the energy consumption and CO_2 emissions of the ironmaking account for about 70% of the total energy consumption and CO_2 emissions of the whole steel process. Therefore, ironmaking industry faces the important challenge of energy conservation and emissions reduction. The pig iron produced by blast furnace accounts for more than 90% of the total iron production. As a reductant and energy carrier that can replace carbon, hydrogen has been proved to have the capability to reduce carbon emissions from the source. Therefore, many scholars all over the world have focused on the combination of hydrogen and traditional ironmaking process.

Ohno *et al.* [1] focused on the effect of FeO content in sinter iron ore on reduction behavior in the hydrogen-enriched blast furnace. It was found that the reduction rate of the sinter iron ore with smaller amount of FeO increased faster at the initial reduction stage; the increase of H_2 content and temperature can improve the rate of reduction reaction. Besides, they also found that the reaction proceeds from the outer periphery of the sample toward the inside, and the two-interface unreacted core model is suitable for the reduction kinetic process. Zhang *et al.* [2] studied the reduction behavior of solid/liquid wustite by hydrogen and found that the thermal and kinetic conditions for the hydrogen reduction of molten phases are better than that of solid phases. Furthermore, this paper used iso-conversional method to analyze the reduction process comprehensively, which can give guidance for the hydrogen injection under different conditions of blast furnace. Zhu *et al.* [3] investigated the gasification and cogasification behaviors of iron coke and coke under simulated hydrogen-rich blast furnace condition. The results indicated the addition of iron coke also can obviously improve the CRI of coke under the simulated hydrogen-rich blast furnace condition (40vol% H_2O + 60vol% CO_2), and then keep it's the skeleton action in the hydrogen-rich blast furnace. Shatokha [4] used 1D steady-state zonal model to simulate the blast furnace operation with hydrogen injection to the conventional tuyeres. It is found that excessive hydrogen injection elongates the thermal reserve zone and might reduce chemical efficiency. When the hot blast is 1200℃, the hydrogen injection amount can be increased from 19.0 to 28.3 kg/t hot metal as the oxygen enrichment amount increases

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from 2vol% to 12vol%. Simulation results showed that coke rate can be decreased 3–4 kg/t H_2, the ironmaking productivity can be increased by up to 13.7%, with direct CO_2 emissions reducing by 10.2%–17.8%, and the decarbonization potential can reach to 9.4–9.7 t CO_2/t H_2.

Hydrogen can also replace carbon in other ironmaking processes to reduce carbon emissions. Chen *et al.* [5] investigated the phosphorus reduction behavior of high-phosphate iron ore during hydrogen-rich sintering process. This research proposed a method for phosphorus removal by gasification and the highest dephosphorization rate can be reached to 29.51%. However, the sintering process faces the problem of selective reduction of iron oxides and apatite, which will limit the dephosphorization rate during sintering process. Therefore, the authors proposed to ensure the efficient dephosphorization process by means of auxiliary heating and negative pressure. Zheng *et al.*[6] studied the effect of pre-oxidization of magnetite-based iron ore on the fluidization behavior, and the results showed that the materials with higher oxidation temperature and wider particle size range showed better fluidization behaviors, while lower oxidation temperature was more beneficial for the reduction rate, especially in the later reduction stage. The pre-oxidation degree shows no obvious influence on the fluidization and reduction behaviors. Besides, this research discussed the reduction mechanism with surface morphology and phase structure.

3. Topic B: Hydrogen-based direction ironmaking process

In 2021, the world's total DRI (direct reduction iron) production is 113 million tons, an increase of 50% compared to 2015, indicating that all countries in the world have a tendency to develop direct reduction process. Since the direct reduction process does not require high pollution processes such as sintering and coking, it is clean and environmentally, and the product quality is also very high. Therefore, the direct reduction process also has great potential in the use of hydrogen.

First of all, Mao *et al.* [7] investigated the reduction kinetic mechanism of iron ore powder reduced by H_2/CO gas mixture. The whole reduction process can be divided into two steps, of which the first step is a double-step reduction ($Fe_2O_3 \rightarrow Fe_3O_4 \rightarrow FeO$) and the second step is a single-step ($FeO \rightarrow Fe$), which is the rate-determining step. It is also found that hydrogen can significantly improve the reduction rate and the reduction rate with pure hydrogen is 3–4 times higher than that with pure CO gas. The swelling behavior of pellets during the reduction process has a direct impact on the entire smelting process. Zhao *et al.* [8] studied the direct reduction swelling behavior of pellets in the hydrogen-based direct reduction process. It is found that the crystalline transformation of iron oxides is the primary reason of pellets swelling. They proposed that controlling a reasonable temperature and increasing the H_2 proportion can decrease the swelling behavior of pellets effectively.

The reducing gas used in the current direct reduction process is mainly a mixture of CO, H_2, CH_4, etc., and the direction reduction of pure hydrogen cannot reach the level of industrial application. Ma *et al.* [9] focused on the influence of spatial gradients, morphology, and internal microstructures of ore pellets on reduction efficiency and metallization during hydrogen direct reduction process. This research found that since the remaining wustite is encapsulated by iron, the outbound diffusion of oxygen is substantially delayed not only in the center but also in the sub-surface zones. The findings can provide guidance for optimizing the particle size, porosity, and microstructure of the microspheres. Metolina *et al.* [10] further investigated the effect of pellet diameter, porosity, and temperature on the reduction process with pure H_2. There is an interactive relationship between temperature and pellet size, reducting pellet size at high temperature favors the reduction reaction, but not at low temperature. This research suggests that the structure and characteristics of the pellets used in the traditional direct reduction process are very different from those used in pure hydrogen direct reduction, so the pellets used in the pure hydrogen atmosphere may require further design. Wang *et al.* [11] used theoretical analysis and calculation methods to discuss the application prospect of pure hydrogen and the key issues in basic theory and application. The hydrogen recution reactor and precess are designed with reference to the industrialized hydrogen-rich reduction process, and a method to appropriately increase the iron reduction temperature and pressure is proposed. The research put forward technical ideas for the development of new temperature-resistant materials and metal coating materials, and provide guidance for the selection of heater and reactor materials. The key factors affecting the smooth operation of the hydrogen reduction process in engineering applications are analyzed to provide a reference for the industrial application of the pure hydrogen reduction process.

4. Topic C: Hydrogen plasma metallurgy

Hydrogen plasma is a further upgrade of hydrogen gas. In terms of thermodynamics and kinetics, hydrogen plasma is superior to H_2. In addition, the range of gas sources can be supplemented by adding reducing gases such as CO and CH_4 to the hydrogen plasma, thereby achieving the effect of complementary advantages. Therefore, hydrogen plasma metallurgy may not only reduce CO_2 emissions from the source, but also achieve the faster production.

Sabat [12] concluded the theory and practice of hematite reduction by hydrogen plasma and investigated the reduction process of solid-state and liquid-state hematite by hydrogen plasma. The in-flight hematite reduction by hydrogen plasma has been identified as a potentially promising alternative to carbothermic reduction. However, there are also many problems, such as excessively high temperatures in thermal hydrogen plasma and considerable vacuum costs in non-thermal hydrogen plasma, need to be further addressed.

5. Perspective

This "Hydrogen Metallurgy" special issue introduces some latest development in metallurgy engineering, which is believed to provide theoretical and technical support for the development of low-carbon metallurgy process. Under carbon neutrality plan, hydrogen metallurgy is bound to become a research hotspot in metallurgy in the coming decades. We hope this special issue can provide more research ideas for future research. We sincerely thank all authors and all reviewers for their efforts. We would like to thank the editorial team of IJMMM for their hard work.

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