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Urea electrosynthesis at industrially viable level on Cd-Fe_2O_3 via a functional design

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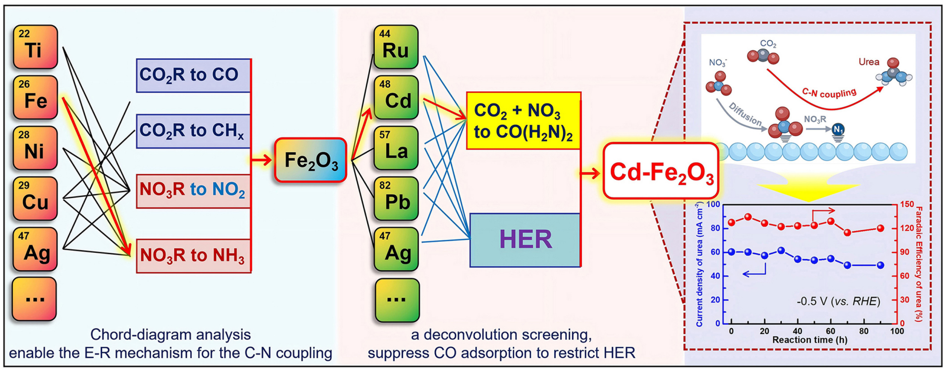

Urea is one of the most important feedstocks in the field of agriculture, chemical industry, *etc*., with a global market above 115 billion by 2031[1]. It is industrially produced by a chemical reaction between liquid NH_3 and liquid CO_2 in a vertical reactor under harsh conditions (e.g., > 10 MPa, > 350 °C[2]), causing > 2% of global annual energy consumption and over 200 million tons of CO_2 releases every year. Developing alternative strategies for the industrial synthesis of urea with low energy consumption and negligible CO_2 emission is a pressing demand.

Electrocatalysis for the synthesis of urea with NO_x^- ions as nitrogen sources is a highly desired strategy[3], owing to its an easily accessible nitrogen source (e.g., waste water[4]), a facile electroreducibility of NO_x^-, a green and sustainable catalytic process at ambient condition, *etc.* It performs a co-reduction of CO_2 and NO_x^- species on the cathode catalyst for the formation of C-N coupling, which is the critical step in urea

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electrosynthesis [$(H_2N)_2$-CO][5]. However, plenty of inevitable side-reactions occur on the surface of the catalyst with a variation of the cathodic potential, e.g., the hydrogen evolution reaction (HER), the CO_2 electrocatalytic reduction (CO_2R), the over electroreduction of NO_x^-. Such competitive side-reactions strongly limit the activity and selectivity of the catalyst toward urea (i.e., $FE_{urea} < 30\%$[6]), revealing the significance of designing suitable catalysts for urea electrosynthesis. Although extensive research has been focused on developing efficient catalysts[7] and some of them could achieve ~50% FE_{urea}[8], but the partial current densities for urea electrosynthesis were merely below a few tens of $mA \cdot cm^{-2}$, indicating a very low urea productivity. Seriously, elevating the current density by an increase of overpotential was found to cause intensive side-reactions, rather than promoting the urea productivity[9]. Currently, figuring out the threshold (i.e., the urea partial current density, the FE_{urea} and the long-term stability) of urea electrosynthesis for the industrialization, with developing an efficient electrocatalyst to reach such a level, has not yet been achieved, hindering the industrial accessibility of urea electrosynthesis.

Hu *et al.* recently solved some of these puzzles[10]. Briefly, they clarified that the threshold of urea electrosynthesis for reaching the industrial large-scale urea productivity is ~95 $mA \cdot cm^{-2}$ at a 50% FE_{urea} by a techno-economic calculation. Importantly, they proposed a rational principle for designing efficient catalyst to achieve the industrial threshold, i.e., a suitable catalyst should have both a high nitrate reduction activity and a negligible CO_2 reduction, promoting the urea electrosynthesis via the Eley-Rideal mechanism. Following this principle, a potential cadmium-modified iron oxide (Cd-Fe_2O_3) catalyst was discovered. The Cd-Fe_2O_3 catalyst shows an excellent urea electrosynthesis performance, with the urea partial current density of 140 $mA \cdot cm^{-2}$ at an appreciable FE_{urea} of 52% for a long-term stability above 100 h at -0.5 V [*vs.* reversible hydrogen electrode (RHE)]. It allows the urea electrosynthesis to reach the industrially viable production rate. Moreover, according to the techno-economic calculation, the cost of electrosynthetic urea is ~550 dollars per ton, which is much lower than that of urea market price (i.e., 616.24 dollars per ton[11]), indicating a profitable urea from the electrosynthesis.

A key innovation of this work is to propose an alternative way for exploring desired catalysts, rather than the classic prediction by a theoretical calculation[12]. In detail, they proposed a four-step design strategy under the guidance of functional requirement [Figure 1]: (1) a techno-economic analysis figures out the industrially promising threshold of urea electrosynthesis and confirms the targeted performance (i.e., 96 $mA \cdot cm^{-2}$ at a 50% FE_{urea}) of desired catalysts [Figure 1A and B]; (2) a reaction procedure analysis proposes a suitable reaction mechanism (i.e., the C-N coupling via the Eley-Rideal mechanism) and determines the critical characteristic of ideal catalysts for the followed catalyst design (i.e., a high nitrate reduction activity and a negligible CO_2R in Figure 1C); (3) a chord-diagram analysis of representative candidates (i.e., Ti, Fe, Cu, *etc.*) for screening out the potential catalyst (i.e., Fe_2O_3) [Figure 1D]; (4) an experimental verification with a precise optimization, which finally confirms Cd-modified Fe_2O_3 as the desired catalyst [Figure 1E and F]. Compared to the theoretical prediction that is performed based on an ideal situation (i.e., ideal crystal surface, ideal coordination number of active sites, *etc.*)[12], this function oriented strategy strongly relies on the existing database from relevant studies, indicating its high reliability for developing catalysts, but with a potential risk of database shortage and additional catalyst optimization on both the catalytic selectivity and stability.

In addition, an efficient Cd-modified Fe_2O_3 catalyst was developed to allow urea electrosynthesis reach the industrial productivity level. Specifically, a preliminary investigation on the Fe_2O_3 candidate shows a superior urea electrocatalytic selectivity (i.e., 71.5% FE_{urea}) and an excellent CO_2R limitation (i.e., < 1% FE_{CO2R}) at 0 V (*vs.* RHE), verifying the effectiveness of the design principle. However, with an increase of cathodic potential to -0.5 V (*vs.* RHE), even though the FE_{CO2R} remains below 1%, the dramatic decline of FE_{urea} to ~20% with an intensive HER (i.e., ~70%) indicates the inferior urea selectivity of Fe_2O_3 at high cathodic potential, as the

*CO-species from CO_2 strongly covers the Fe_2O_3 surface and restricts the NO_x^- activation. The shortage of Fe_2O_3 was then optimized by introducing cadmium additives, which is selected by a rapid deconvolution screening method. The obtained Cd-Fe_2O_3 catalyst shows a urea partial current density of 140 mA·cm^{-2} and an appreciable FE_{urea} of 52% at -0.5 V (*vs.* RHE), much higher than that of unmodified Fe_2O_3 (i.e., ~20% FE_{urea}) and superior to the industrial threshold (i.e., ~95 mA·cm^{-2} at a 50% FE_{urea}). A long-term stability on Cd-Fe_2O_3 for over 100 h was performed [Figure 1G and H]. The relatively high catalytic stability of Cd-Fe_2O_3 in laboratory indicates its promise for the industrial exploration. An *in-situ* monitor by the attenuated total reflectance Fourier-transform infrared spectroscopy (ATR-FTIR) measurement identifies that the protonation of *CO_2NH_2 is the rate-determining step, which is enhanced by the Cd incorporation that alters the electronic structure of Fe_2O_3 for a weak CO adsorption and a suppressed HER.

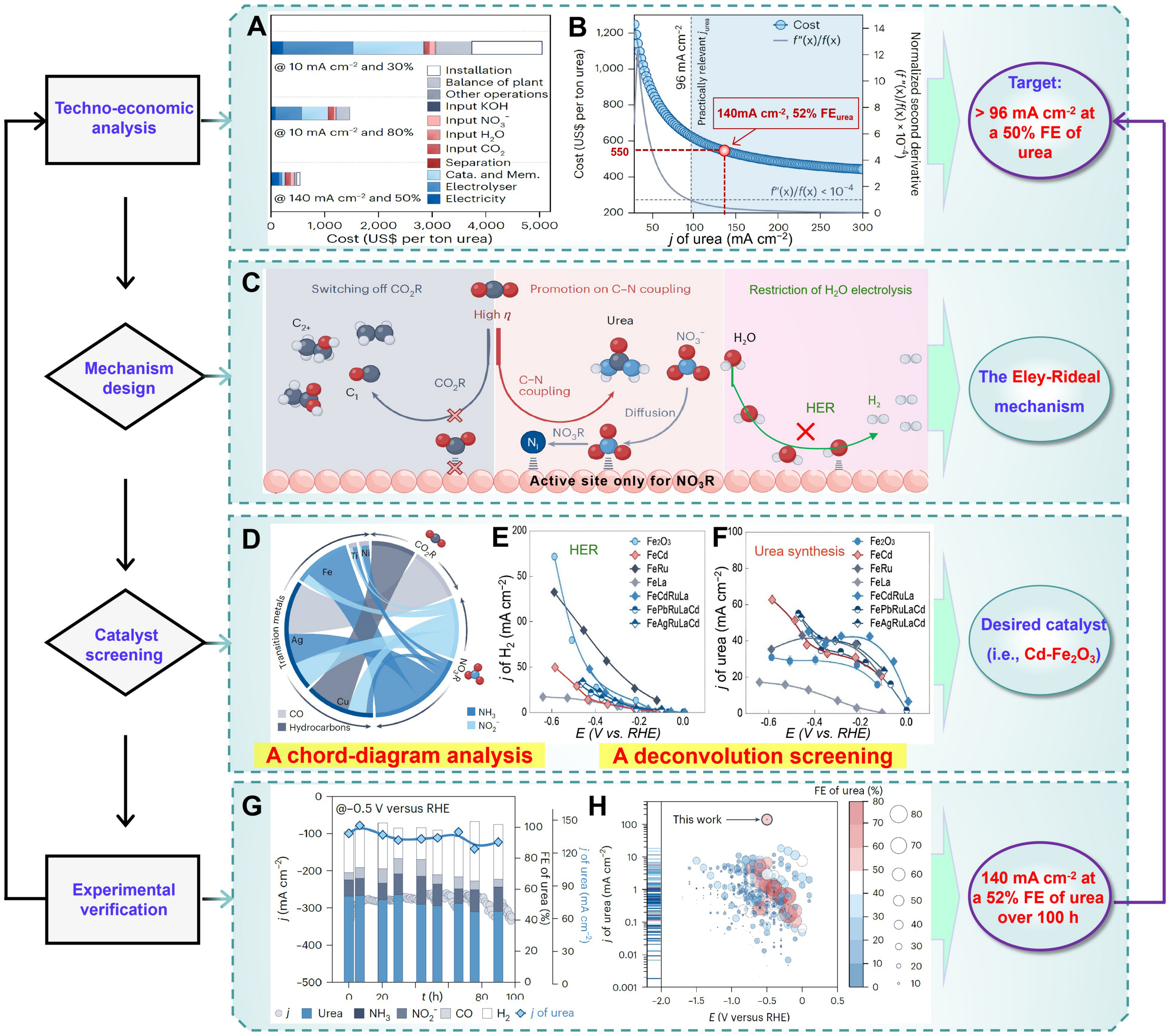

Figure 1. (A) The levelized cost of urea by various electrosynthesis methods; (B) Correlation of urea cost on urea partial current density at 50% urea FE; (C) Reaction pathway of CO_2R, HER and C-N coupling; (D) Chord diagram describing the selectivity of typical transition metals towards CO_2R and NO_3R; Partial current densities of (E) H_2 and (F) urea on Fe-based catalyst; (G) Stability test of urea electrosynthesis on Cd-Fe_2O_3; (H) Summary of the reported urea electrosynthesis performance. (A-H) Adapted with permission from Ref.[10]. Copyright 2026 Spring Nature. FE: Faradaic efficiency; CO_2R: CO_2 electrocatalytic reduction; HER: hydrogen evolution reaction; RHE: reversible hydrogen electrode.

As a conclusion and perspective, Hu *et al.* developed an efficient Cd-Fe_2O_3 catalyst by following an innovative catalyst-design strategy[10]. The catalyst enables urea electrosynthesis to reach an industrially accessible level. Besides that, several questions remain open: (1) A slight deactivation of catalyst can be clearly observed in Figure 1G (i.e., the urea partial current density decreases from 140 to ~120 mA·cm^{-2} during 100 h of reaction). The long-term stability issue has to be solved before reaching an industrialization; (2) Cd is a poisonous metal. Exploration on the Cd-alternative catalytic additives is desired, but with a great challenge as Cd is already the optimized choice; (3) The unclear state of Cd additives on Fe_2O_3, the practical active site for the $^*CO_2NH_2$ protonation, the missing threshold of stability, *etc.*; (4) Only the electric energy at the cathode is utilized (i.e., the urea electrosynthesis), resulting in a waste of energy at the anode. Further oxidation reaction could be explored on the anode; (5) Optimizing the mass transfer efficiency within reactor components (e.g., membrane electrodes) may promote the catalytic system to meet industrial application standards. All these questions are required to be addressed in future studies in order to realize the industrial synthesis of urea by the electrocatalytic way.

DECLARATIONS

Authors' contributions

Made substantial contributions to conception and design of the study and performed data analysis and interpretation: Li, W. X.; Wang, Z.; Su, B. L.

Performed data acquisition and provided administrative, technical, and material support: Wang, Z.; Li, Y. C.; Li, Z. D.

Availability of data and materials

Not applicable.

AI and AI-assisted tools statement

Not applicable.

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Conflicts of interest

Su, B. L. is Editor-in-Chief of the journal *Chemical Synthesis*. Su, B. L. was not involved in any steps of the editorial process, notably including reviewers' selection, manuscript handling, or decision-making. The other authors declare that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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