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Carbon Capture and Storage by Continuous Production of CO_2 Hydrates Using a Network Mixing Technology

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Abstract : Nowadays, it is well recognized that carbon dioxide emissions, together with other greenhouse gases, are responsible for the dramatic climate changes that have been occurring over the past decades. Gas hydrates are currently seen as a promising and disruptive set of materials that can be used as a basis for developing new technologies for CO_2 capture and storage. Its potential as a clean and safe pathway for CCS is tremendous since it requires only water and gas to be mixed under favorable temperatures and mild high pressures. However, the hydrates formation process is highly exothermic; it releases about 2 MJ per kilogram of CO_2, and it only occurs in a narrow window of operational temperatures (0 - 10 °C) and pressures (15 to 40 bar). Efficient continuous hydrate production at a specific temperature range necessitates high heat transfer rates in mixing processes. Past technologies often struggled to meet this requirement, resulting in low productivity or extended mixing/contact times due to inadequate heat transfer rates, which consistently posed a limitation. Consequently, there is a need for more effective continuous hydrate production technologies in industrial applications. In this work, a network mixing continuous production technology has been shown to be viable for producing CO_2 hydrates. The structured mixer used throughout this work consists of a network of unit cells comprising mixing chambers interconnected by transport channels. These mixing features result in enhanced heat and mass transfer rates and high interfacial surface area. The mixer capacity emerges from the fact that, under proper hydrodynamic conditions, the flow inside the mixing chambers becomes fully chaotic and self-sustained oscillatory flow, inducing intense local laminar mixing. The device presents specific heat transfer rates ranging from 107 to 108 $W \cdot m^{-3} \cdot K^{-1}$. A laboratory scale pilot installation was built using a device capable of continuously capturing 1 $kg \cdot h^{-1}$ of CO_2, in an aqueous slurry of up to 20% in mass. The strong mixing intensity has proven to be sufficient to enhance dissolution and initiate hydrate crystallization without the need for external seeding mechanisms and to achieve, at the device outlet, conversions of 99% in CO_2. CO_2 dissolution experiments revealed that the overall liquid mass transfer coefficient is orders of magnitude larger than in similar devices with the same purpose, ranging from 1 000 to 12 000 h^{-1}. The present technology has shown itself to be capable of continuously producing CO_2 hydrates. Furthermore, the modular characteristics of the technology, where scalability is straightforward, underline the potential development of a modular hydrate-based CO_2 capture process for large-scale applications.

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