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Coal-derived syngas purification and hydrogen separation in a supersonic swirl tube

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ABSTRACT

Coal gasification is a means to produce synthesis gas (or syngas) which may then be processed further to produce hydrogen of desired purity. Ultimately separation of the H_2 and CO_2 from the process stream is required. Membrane technologies are currently applied to such separation processes. Here a theoretical approach derived from aerospace technologies is shown to have promise as a robust method for extracting H_2 from syngas. It is based on a swirling expansion process in a specially designed supersonic flow nozzle which combines both condensation and segregation of the unwanted species. The quasi-one-dimensional equations for swirling compressible flow with rate controlled condensation are derived, the dynamics of the separation of condensed species is described, and preliminary solutions for cases of practical interest are presented. This theoretical foundation is shown to support the feasibility of an industrial-scale device incorporating this effect and lays the groundwork for future experimental investigations.

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1. Background and process description

Coal gasification is a means of producing synthesis gas (or syngas) which may be processed to produce hydrogen of desired purity. After an initial cleaning step to remove solid particulates including SO₂, the remaining coal-derived syngas is primarily H₂, CO, CO₂, N₂ and H₂O with trace amounts of CH₄, COS, H₂S, NH₃ and HCN. The water gas shift (WGS) reaction is used to reform the remaining CO with the H₂O to produce more H₂ (and CO₂). A twostage WGS approach is often used to maximize H₂ and minimize CO yields. These processes require catalysts with the first (high temperature) WGS reaction occurring at an input temperature of 315 °C-370 °C. This exothermic process results in a gas flow of about 425 °C which requires cooling for the second WGS reaction which occurs at approximately 200 °C. Significant research is being devoted to improving the performance of WGS catalysts and minimizing susceptibility to "poisoning" or deactivation from exposure to undesired compounds such as NH₃, COS and H₂S. However, subsequent separation of the H₂ and CO₂ from the process stream is still necessary - the CO₂ being sequestered and the H₂ stored for transportation or fed directly to a local power generator (e.g. turbine).

Thus far membrane technologies have been the primary focus of attention for the required separation process [1]. Polymer and metal substrates with selective catalysts allow, for example, only H_2 to permeate. Challenges remain for the membrane-based separation approach including mass flux limitations, catalyst poisoning and agglomeration of residue, effectively clogging the process. Industrial applications seek "process intensification" whereby multiple processes are integrated into one, thereby reducing steps, improving efficiency, and reducing cost. A new scheme is proposed here, one derived from aerospace technologies, which promises to provide such integration in a robust method for extracting H_2 from syngas. This unique aero-thermodynamic inertial separation device is best described sequentially, as follows:

- 1. Coal-derived syngas at gasifier exit conditions is directed to a converging-diverging nozzle and expanded to supersonic speed.
- 2. Supersonic expansion results in a rapid decrease of temperature and pressure leading to the sequential condensation (and/ or solidification) of most of the undesirable constituents including H₂O, CO₂, H₂S, NH₃, HCN, and COS (the H₂O is recovered in a subsequent step).
- 3. The high-density liquid and solid droplets and particulates are inertially separated by centrifugal forces instigated by either stationary turning vanes or an energy-extracting turbine in the



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