462h Production of Hydrogen and Sulfur from Hydrogen Sulfide in a Nonthermal-Plasma Pulsed Corona Discharge Reactor

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Gas streams containing hydrogen sulfide (H_2S) are encountered in almost all fossil fuel energy extraction and processing systems. The conventional treatment method for H_2S is the Claus process, which produces sulfur and water by the net reaction: $H_2S + O_2 \rightarrow S + H_2O$. The reaction is inefficient because the valuable potential product hydrogen (H_2) is converted into water. The transformation of hydrogen in a weakly bound state in H_2S to a strongly bound state in H_2O results in the loss of a potential H_2 source. Therefore, direct dissociation of H_2S into H_2 and sulfur would be preferable.

Many methods have been investigated to dissociate H_2S into its constituent elements. These methods can be divided into two categories: thermal decomposition (either catalytic or noncatalytic) and nonthermal plasma decomposition. The direct decomposition reaction is highly endothermic with low equilibrium conversions even at high temperatures. Product removal is required to increase conversion by condensation of the sulfur and separation of the hydrogen with membranes. Nonthermal plasma has the advantage of being a partially ionized gas, which is a source of chemically active species, including radicals, excited states and ions, that can promote chemical reactions at ambient temperatures. Direct dissociation of H_2S using various plasma processing technologies, including microwave plasma, RF pulse, glow discharge, silent discharge, gliding discharge, and pulsed corona discharge, has been attempted. A large amount of work on microwave decomposition of H_2S has been carried out in former Soviet Union. Both laboratory and pilot units were used successfully for the decomposition of H_2S alone or mixed with CO_2. The limited amount of reported work on dissociation of H_2S in other types of plasmas has generally been performed at low concentrations (~3% H_2S), which are not useful for commercial application.

One of the major concerns for application of nonthermal plasma is energy consumption. Pulsed corona discharge (PCD) plasmas have been extensively investigated and used in NO_x and methane conversion. Comparison of energy efficiency of methane conversion in three kinds of nonthermal plasma reactors (PCD, microwave, and silent discharge) shows that PCD reactors are one to two orders of magnitude more energy efficient than the other two. This result prompted this investigation of H_2S conversion in a PCD reactor.

A PCD reactor has been fabricated and used to dissociate H_2S into hydrogen and sulfur. A nonthermal plasma cannot be produced in pure H_2S with our reactor geometry (even up to 30 kV) because of the high dielectric strength of pure H_2S (~2.9 times higher than air). Therefore, H_2S was diluted in another gas with lower breakdown voltage (or dielectric strength). Breakdown voltages of H_2S in four background gases (Ar, He, N_2 and H_2) have been measured at different H_2S concentrations and pressures. Breakdown voltage linearly increases with increasing pressure. H_2S conversion rates and energy efficiencies depend on the background gas and H_2S inlet concentrations. For example, H_2S conversion rates are higher in Ar than in H_2. With increasing H_2S concentrations, H_2S conversion initially increases, reaches a maximum, and then decreases. These observations can be explained by the proposed reaction mechanisms of H_2S dissociation in different background gases. The results show that nonthermal plasma technique is effective for dissociating H_2S into hydrogen and sulfur.