Production of H_2 from H_2S by plasma–catalysis coupling

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Highlights

- Non thermal plasma can produce hydrogen from H_2S
- Catalyst addition improve significantly energy efficiency

1. Introduction

Introduction Hydrogen sulfide is a gas naturally present in many geological environments and is also a by-product of numerous industrial processes. More than 10 Mt of H2S are emitted worldwide each year [1]. Today, the Claus process converts H_2S to sulfur but at the cost of a substantial loss of hydrogen as water vapor [2]. Direct decomposition of H_2S is a promising route to produce low-carbon hydrogen while valorizing sulfur. However, even at high temperature thermolysis is limited by the reaction thermodynamic equilibrium and conversions remain low (30% H_2S conversion at 1130 °C). In recent years cold plasma has emerged as a promising alternative for H_2S decomposition because it can generate active species at moderate gas temperatures. Studies by L. Zhao et al. reported that plasma discharges coupled with a catalyst (ZnS/Al_2O_3) can achieve significant conversions (100% conversion for 20% H_2S in Ar, flow 30 mL·min−1, power 4 W, EC 181 kJ·mol−1) [3]. Nevertheless, plasma chemistry remains difficult to control and the plasma–catalyst interaction is still poorly understood. The aim of this study is to provide new insights into plasma–catalysis coupling and, more specifically, to investigate the impact of various catalyst properties (physicochemical, structural, electrical) on reactivity under plasma discharge. For this purpose, MoS_2-based catalysts supported on alumina were tested under different plasma conditions.

2. Methods

Experiments were carried out in a tubular dielectric barrier discharge (DBD) reactor. The inner electrode is a stainless-steel rod (1.5 mm diameter) and the outer electrode is copper (Figure 1a). The plasma volume is 0.5 cm^3. This geometry allows insertion of a material into the plasma zone. The catalyst was employed as a powder with controlled particle size (315–600 μm). H_2S was diluted in argon at 10 vol%. The plasma was generated using a pulsed power supply, producing high, very short voltage steps that increase electron energy and the density of reactive species while limiting gas heating. Applied voltage and plasma current were measured with probes connected to an oscilloscope, enabling precise determination of the power deposited in the plasma—an essential parameter for characterizing the discharge and comparing operating conditions. Outlet gases were analyzed by micro-GC to quantify H_2S conversion and hydrogen production.

3. Results and discussion

A parametric study was conducted to evaluate the influence of operating parameters on H_2S decomposition, notably deposited power, pulse frequency, residence time and H_2S concentration. This study identified favorable conditions prior to catalyst introduction.

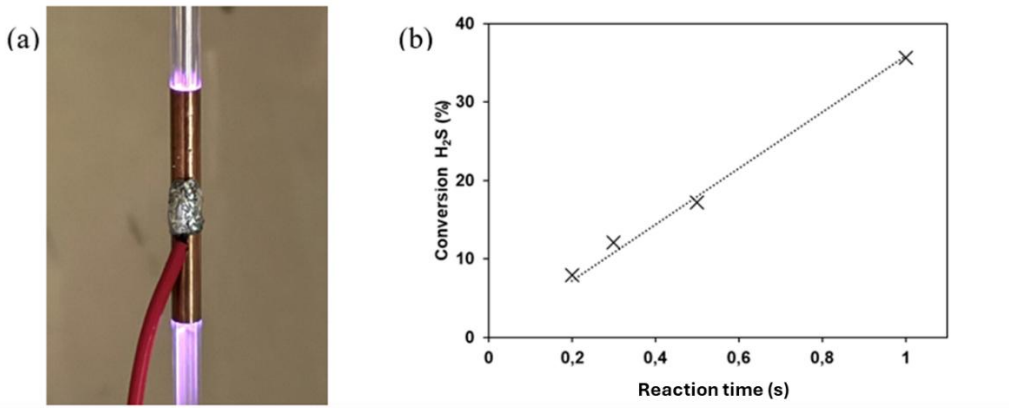

Figure 1. (a) DBD reactor under plasma, (b) H2S conversion as a function of residence time (10% H_2S in Ar, Power 2 W, frequency 5 kHz).

Figure 1b illustrates H_2S conversion versus residence time in the reactor. A marked increase in conversion is observed with increasing residence time, since the probability of collisions between H_2S molecules and plasma reactive species rises. Hydrogen selectivity is 100%: H_2S is decomposed solely to sulfur and H_2.

Table 1 compares the performance of the DBD reactor alone with that obtained in the presence of 10% MoS2/Al2O3 at identical power.

Table 1. Performance of H_2S (10 vol%) decomposition by DBD plasma alone and in the presence of a catalyst.

Catalyst	Power (W)	H_2S conversion (%)	EC (kJ/mol)
Réacteur vide	2.0 ± 0.1	12	280
10%MoS_2/Al_2O_3	2.0 ± 0.1	26	130

The 10% MoS_2/Al_2O_3 increases H_2S conversion while reducing energy consumption relative to the empty reactor, highlighting the interest of plasma–catalyst coupling. Sulfur did not cause clogging or degradation during the tests (duration: 40 min). These observations make MoS_2/Al_2O_3 a promising candidate to improve the process energy efficiency. However, these are preliminary results: further experiments are planned (higher MoS_2 loadings, different supports and longer-term stability studies) to confirm the trend and optimize catalyst formulation. In the continuation of this work, characterizations will be performed before and after plasma exposure (SEM, XRD, specific surface area, dielectric constant, etc.) to better understand material evolution and the impact of the plasma on catalyst structure and properties, and vice versa.

4. Conclusions

Plasma catalysis is a promising candidate to process endothermic reactions such as H_2S cracking, yet optimization of plasma conditions and catalyst synergies remain to be assessed.

References

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Keywords

Plasma-catalysis; H_2S cracking; hydrogen; energy efficiency