ABSTRACT

In the last few decades, greenhouse gas emission has increased very rapidly due to a surge in energy demand which results in a rise in global temperature. Hydrogen is an alternative source of energy because it is a non-polluting, eco-friendly, and regenerating nature which makes it a most attractive fuel source. Thermochemical decomposition of water in the Iodine-Sulfur (I-S) cycle for hydrogen production is the most reliable and propitious because of its higher efficacy of 56 %. I-S process involves mainly three reactions namely, Bunsen reaction, sulfuric acid decomposition, and hydro iodic cycle decomposition**.** Among them, thermal decomposition of sulfuric acid is highly corrosive and very energy-intensive usually requires a high temperature of approximately 973-1173 K and exhibits a large kinetic barrier (73-150 kJ/mol). Finding ways to circumvent the high energy consumption may render a path towards enabling a more efficient, and more economically viable low-temperature process.

This research is focused on the estimation of thermodynamic properties including phase behavior and chemical speciation, finding a suitable catalyst, and reactor design for high energy demanding sulfuric acid decomposition for hydrogen production. In this work, metal oxides and spinels are supported over mesoporous silica and pretreated silicon carbide. These catalysts were characterized through XRD, FESEM, HRTEM, and BET following activity and 100 h long-run stability testing. Iron oxide and spinels dispersed on a silica surface grown on a SiC support material are found to be the best catalyst for an endothermic high temperature and highly corrosive reaction condition such as sulfuric acid decomposition. The combined theoretical and experimental findings not only allow us to design cheaper and stable catalysts but also provide an atomistic insight into the mechanism of the metal-support interaction. Consequently, the successful tuning of the latter helps in the enhancement of overall activity and stability. The high activity and stability are ascribed to the presence of oxygen deficit sites in the catalyst developed for the model reaction under consideration.

After that, the reaction kinetics of sulfuric acid decomposition over highly active and thermally stable Fe_2O_3/SiC-Pretrt catalyst is investigated in a fixed-bed quartz reactor. To scale-up the process and improve the heat transfer and integration, the influences of the different sizes and shapes of the catalyst pellet on the reaction performance and effectiveness factor are estimated.

Further, both decomposition and recuperation can be achieved simultaneously through effective reactor structure optimization