Efficient and Reversible Catalysis of Formic Acid-Carbon Dioxide Cycle Using Carbamate-Substituted Ruthenium-Dithiolate Complexes

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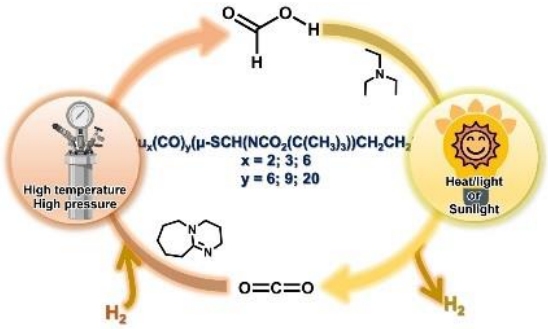
It is important to develop viable technologies for efficient hydrogen (H_2) production and carbon dioxide (CO_2) conversion to realize the future supply of clean energy and reduction of global CO_2 concentration. Herein, we report a series of carbamate-substituted ruthenium-dithiolate complexes **1**–**3**, [$Ru_x(CO)_y$(μ-SCH($NCO_2(C(CH_3)_3)$)CH_2CH_2S)] (x = 2, 3, 6 and y = 6, 9, 20), which can effectively catalyze the reversible formic acid/carbon dioxide (FA-CO_2/H_2) cycle that can be used to convert CO_2 and produce H_2. Complex **2** effectively dehydrogenates FA to produce H_2 with an unprecedented turnover frequency (TOF) of 1.15×10^6 h^{-1} under conditions of sunlight irradiation. Under conditions of high temperature and pressure, complex **3** hydrogenates CO_2 to FA with a high initial TOF of 1.02×10^6 h^{-1}, resulting in an efficient FA-CO_2/H_2 cycle. These results can potentially help provide the platform for the development of technologies that can be used in future to produce clean energy and control environmental threats.



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