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Syngas Production: Diverse H_2/CO Range by Regulating Carbonates Electrolyte Composition from CO_2/H_2O via Co-Electrolysis in Eutectic Molten Salts

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Results and discussion - Supporting Information

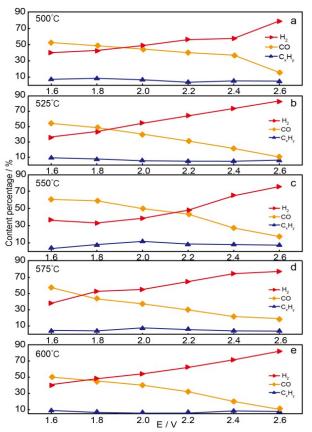


Fig. S1: Compositions of the electrolysis gaseous products in the operating voltage range from 1.6 V to 2.6 V at temperatures of 500

°C, 525 °C, 550 °C, 575 °C, and 600°C in the electrolyte of the $Li_{1.51}K_{0.49}CO_3$ -0.1LiOH system.

The products trend in Li_{1.51}K_{0.49}CO₃-0.1LiOH electrolyte is observed from **Fig. S1**. The CO content at all temperatures decreases significantly with voltage changes in the range of 1.6-2.6 V, as evident by Fig. S1. For example, at 550 °C, the CO content decreases from 52% at 1.6 V to 21% at 2.6 V, and the result shows that at the same temperature, an increase of electrolysis is not conducive to the generation of CO. Specifically, 1.6 V results in the best electrolysis of the Li-K system at each temperature.

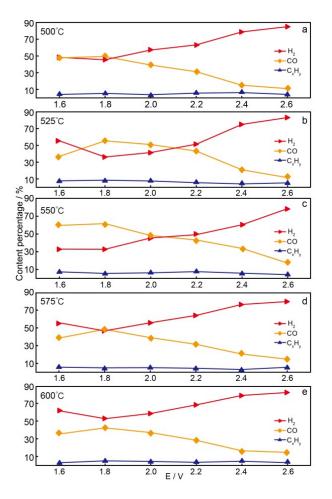
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Fig. S2: Compositions of the electrolysis gaseous products in the operating voltage range from 1.6 V to 2.6 V at temperatures of 500 °C, 525 °C, 550 °C, 575 °C, and 600°C in the electrolyte of the $Li_{1.43}Na_{0.36}K_{0.21}CO_3$ -0.1LiOH system.

Fig. S2 shows the H₂, CO and C_xH_y content in the Li-Na-K ternary system with various electrolytic conditions. When the temperature change is in the range of 500-550 °C, at each temperature, there is a similar trend among the content of each product. The yield of CO increases with an increasing applied voltage (1.6-1.8 V), and when the voltage continued to increase (1.8-2.6 V), the CO content begins to decrease. When electrolysis with 1.8 V voltage, the optimum CO fraction is obtained.

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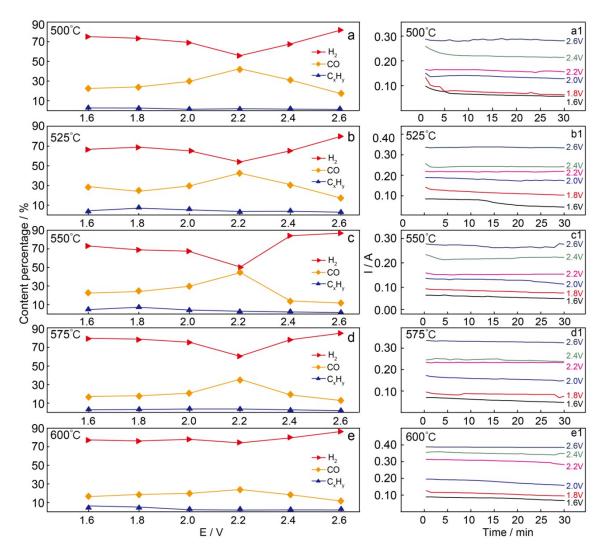


Fig. S3: Compositions of the electrolysis gaseous products and I-t curves of the electrolytic system in the operating voltage range from 1.6 V to 2.6 V at temperatures of 500 °C, 525 °C, 550 °C, 575 °C, and 600°C in the electrolyte of the Li_{0.85}Na_{0.61}K_{0.54}CO₃-0.1LiOH system.

Fig. S3 presents the yield of each gaseous product in the Li_2CO_3 - $Na_2CO_3-K_2CO_3$ system with a mass ratio of 31:32:37. When the temperature is in the range of 500-600 °C, a low yield of CO is obtained at 1.6 V. This observation can be explained because 1.6 V is an insufficient voltage for CO generation. When the cell voltage increases to 1.8 V, the enlarged voltage leads to an enhanced current density. At 2.2 V, the current is elevated to approximately 0.16 A at 550 °C, which can guarantee the electrochemical reactions proceeded facilely. At 2.4 V and 2.6 V, the steady larger current results in a relatively lower current efficiency, as shown in **Fig. 10d**. This is due to the increasing current of the electrolyte at the voltage of 2.4-2.6 V, which increases the power consumption in the same

time, resulting in a decrease in current efficiency. Therefore, 2.2 V is always the best electrolysis voltage in the temperature range of 500-600 $^{\circ}$ C.