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## ABSTRACT:

### Mechanochemical Hydrogenation Of CO<sub>2</sub> and Carbonates from Solid Materials under Ambient Conditions

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From the perspectives of both atmospheric CO<sub>2</sub> mitigation and carbon utilization, the hydrogenation of CO<sub>2</sub> into useful fuels and chemicals has attracted considerable attention. Among the various approaches, CO<sub>2</sub> methanation is particularly attractive because it enables the conversion of CO<sub>2</sub> into CH<sub>4</sub>, a widely used fuel. Moreover, since the CO<sub>2</sub> released upon the combustion of CH<sub>4</sub> can in principle be recycled back into CH<sub>4</sub>, this process can be regarded as part of a carbon cycle. Based on this concept, many studies have been reported in recent years<sup>1</sup>. However, conventional CO<sub>2</sub> methanation generally requires elevated temperatures above 200 °C for kinetical reasons and involves challenges associated with the handling of high-pressure H<sub>2</sub> gas. In contrast, from a thermodynamic standpoint, CO<sub>2</sub> methanation is more favorable at lower temperatures. In this presentation, we report mechanochemical CO<sub>2</sub> methanation as an alternative approach to lowering the reaction temperature and simplifying the reaction system<sup>2</sup>. In particular, for CO<sub>2</sub> methanation using LaNi-based hydrogen storage alloys, we developed a gas-flow type mechanochemical reactor and demonstrated in-situ monitoring of the reacted gas. As a result, CH<sub>4</sub> generation was confirmed without external heating. Furthermore, we found that hydrogen (H) dissolved in the alloy accelerates CO<sub>2</sub> methanation. Post-reaction analysis revealed that LaNi<sub>5</sub> underwent disproportionation into Ni, La-carbonate, La-hydroxides. These results further suggest that the carbonates act as the carbon source for CH<sub>4</sub> formation and that hydrogen evolution from hydroxides also occurs<sup>3</sup>.

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[2] K. Sawahara, K. Yatagai, T. Boll, A. Pundt and R. Gemma, Int. J. Hydrogen Energy, 47, 19051 (2022).

[3] K. Sawahara, K. Asano, R. Sato, R. Kanega, T. Sakai and R. Gemma, Manuscript under review (2025).