

## Abstract

Observed signals from a transient response analysis developed by combining a broadened pulse with a step change or an isotopic pulse. The combined method provides information about the activation of the catalyst and the catalytic reaction mechanism. The method applied to the CO<sub>2</sub> reforming of methane over NiO/SiO<sub>2</sub> at 8000C. The response to the broadened CH<sub>4</sub>/CO<sub>2</sub> (1:1) pulse over fresh NiO/SiO<sub>2</sub>, exhibited an induction time during which no reaction between CH<sub>4</sub> and CO<sub>2</sub> occurred, indicating that NiO/SiO<sub>2</sub> had no activity. As the NiO was gradually reduced by CH<sub>4</sub>, the reforming reaction took place, indicating that Ni<sup>0</sup> is the active site for this reaction. During the transient response to the step change to He in the broadened pulse of CH<sub>4</sub>/CO<sub>2</sub> over reduced NiO/SiO<sub>2</sub> catalyst at 8000C, a long CO tail was observed during the experiment; however, no such tail was noted in the response after a step change to He in the broadened pure Co pulse at 8000C. This indicates that the CO desorption is rapid and that the surface reaction between C and O species constitutes the rate-determining step of the reforming reaction. The transient response to a sharp <sup>18</sup>O<sub>2</sub> (isotope) pulse introduced into the broadened CH<sub>4</sub>/C<sup>16</sup>O<sub>2</sub> pulse indicated that, over a reduced NiO/SiO<sub>2</sub> catalyst, <sup>18</sup>O<sub>2</sub> replaced some C<sup>16</sup>O<sub>2</sub> in the reaction with CH<sub>4</sub> to generate CO<sup>18</sup> and that some Ni<sup>0</sup> was oxidized to Ni<sup>18</sup>O. The long tail of CO<sup>18</sup> indicates that carbon species reduced this Ni<sup>18</sup>O to Ni<sup>0</sup>.

.Keywords: Rate Determining Step, Transient Response, broadened Pulse, Induction time, Isotopic Pulse.