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Heterogeneous Catalysis: Effect of an Alternating Electric Field

Abstract. Oxidation of carbon monoxide was studied in a static reactor at $123^\circ \pm 1^\circ\text{C}$, with NiO as catalyst. An alternating electric field of about 22,000 volts per centimeter, peak to peak, was applied normal to the catalytic surfaces. The rate of oxidation was thereby enhanced by a factor of 6 at 100 cycles per second, with maximum enhancement at between 100 and 200 cycles per second.

I studied the oxidation of carbon monoxide in a static reactor containing 20 plates of grade-A nickel, 5.08 cm in diameter, oxidized by controlled oxidation to a thickness of about $1\ \mu$ NiO (1). The plates were separated by ceramic spacers—1-mm-thick rings. An a-c electric field of 22,000 volts/cm, peak to peak, was applied between the plates. At frequencies ranging from 50 to 300 cy/sec, the field consisted of a smooth sine wave (Fig. 1). The reactor was totally enclosed in a thermostat controlled at $123^\circ \pm 1^\circ\text{C}$. The re-

actant gases, air and CO, had been dried by passage through drying tubes containing 5A molecular sieve of 30 mesh; they were mixed before entering the reactor. The feed contained 0.028 percent CO₂ and 5.2 percent CO (both by volume); the balance was air. After purge of the reactor for 4 hours at a flow rate of 96 cm/min, the reactant mixture was trapped in the reactor at a pressure of 12.5 cm water above atmospheric.

Compositions of CO and air were determined by measurements of flow rates with two calibrated Matheson-610 flow meters. After a reaction period of from 19 to 68 hours, the resultant gaseous mixture was analyzed for CO₂ content by a gas chromatograph using a silica-gel column; the area under the gas-chromatographic curve was determined with a planimeter, with an estimated possible error of up to 0.01 percent (by volume) CO₂. Table 1 summarizes the data.

Net conversion of CO to CO₂ was the difference between CO₂ concentrations in the gaseous mixture before and after the reaction (Fig. 2). For the runs without electric field (runs 4/5, 4/6, and 4/12; Table 1), the relation was linear between the amount of CO converted to CO₂ and the resident time of the reactive mixture; this fact may suggest a zero-order reaction. However, since the change in CO concentration was relatively small, the order of the reaction cannot be asserted with certainty.

The net conversion to CO₂ during 19 hours in the reactor is plotted in Fig. 2 as a function of the a-c field frequency at a constant peak-to-peak field of 22,000 volts/cm. Apart from high-frequency distortions at 400 cy/sec, the field was a smooth sine wave. The enhancement of the rate of oxidation of CO does not reflect the heating effects of the a-c circuit because the capacitor

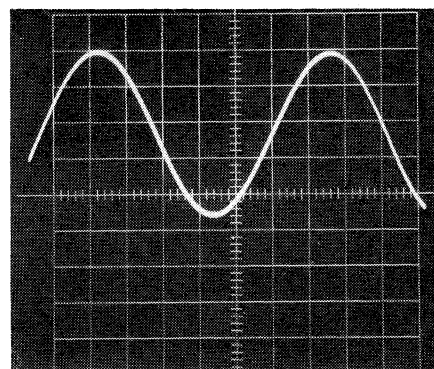


Fig. 1. Oscilloscope display of the sinusoidal electric field applied between the NiO catalytic surfaces. Scales: horizontal, 0.5 millisecond per division; vertical, 500 volts per division.

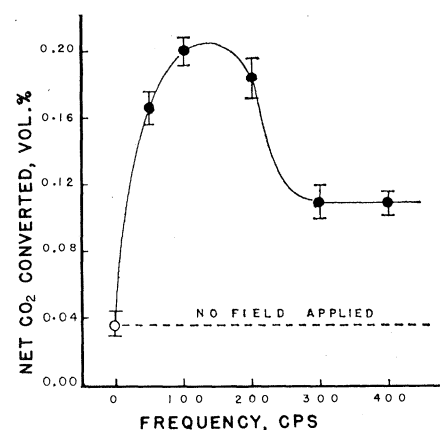


Fig. 2. Net increment (percentage by volume) of CO₂ after 19 hours in the reactor at $123^\circ \pm 1^\circ\text{C}$ and at a pressure of 12.5 cm water above atmospheric. Open circle, with no field applied on the NiO catalyst; solid circles, with sinusoidal electric field applied. Cps, cycles per second.

plates, serving as catalyst, were the only part of the circuit in the reactor. *A fortiori*, the temperature of the reactor was automatically controlled at $123^\circ \pm 1^\circ\text{C}$ during all experiments.

The fact that the rate of reaction was maximum at between 100 and 200 cy/sec may imply that the resident time of the adsorbed species is of the order of 5 to 10 msec. The nature of the adsorbate and the reaction mechanism remain to be determined.

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References and Notes

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2. I thank R. G. Schultz, M. A. Gilleo, and G. E. Barker of the Monsanto Co., St. Louis, Missouri, for their participation.

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Table 1. Oxidations of CO to CO₂ during eight runs. PP, peak to peak.

Run	Duration (hr)	Field, PP (volts/cm)	Frequency (cy/sec)	CO ₂ (vol %)	
				In residue	Net conversion
4/5	67	none		0.168	0.140
4/6	19	none		.060	.032
4/7	19	22,000	50	.195	.167
4/8	19	22,000	100	.230	.202
4/9	19	22,000	200	.215	.186
4/12	68	none		.175	.147
4/13	19	22,000	300	.138	.110
4/14	19	22,000	400	.138*	.110

* Wave form contained high-frequency distortions.