

An Improved External Recycle Reactor for Determining
Gas-Solid Reaction Kinetics⁺

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External recycle reactors have been used extensively to determine the rates and kinetics of gas-solid catalytic reactions. A schematic diagram of this reactor is shown in Figure 1. The advantages of this type of reactor have been noted as early as 1958 by Perkins and Rase. They pointed out that it would minimize or eliminate temperature and diffusional gradients in the catalyst bed, resulting in data which more nearly represented the kinetics of the chemical reaction. Carballs et.al. used this reactor system to study hydrogen chemisorption on supported platinum, while Schlatter and Chou measured CO oxidation rates on supported platinum and palladium in this system. Paspek et. al. and Hecker used a heating coil upstream of the reactor to insure isothermal reactor conditions and a cooling coil downstream to remove the heat and protect the pump.

A disadvantage of these systems is that at the start of a run the gas in the recycle loop must be completely purged or displaced with another gas having a different composition, so that the reaction rate of this gas can be measured with confidence. However, during the purging phase of the test, whether displacing the gas by evacuation or purging, the partial pressure of the reactants in the test gas gradually increase with time. This means that the activity of the catalyst for adsorption, surface reaction, or desorption could change during this time period. If catalyst activity is influenced by its prior history, such as the gradual increase in reactant concentrations at the start of the test, the measured rates could be significantly different from those where the catalyst is exposed to the reactant concentrations in the feed gas on first contact, or at zero time.

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The improvements in the recycle system discussed in this note effectively eliminate the initial concentration variation by two modifications shown in Figure 2. One of the modifications is a vacuum line connection to the recycle loop which permits this loop to be evacuated and then filled with the test gas mixture to slightly above atmospheric pressure.

The other modification is a bypass line across the reactor which permits the reactor to be held under vacuum while the rest of the recycle loop is filled with test gas. Note that no pre-heating coil or cooling coil is shown adjacent to the reactor. These were found unnecessary in tests at a maximum reactor temperature of 498°K and a reactor flow rate of 10 std cm³ He/min, as the gas was heated to this temperature or cooled to room temperature in stainless steel tubing only 25 cm long by 0.48 cm diameter.

The three-step procedure for bringing the feed gas mixture into contact with the catalyst at time zero is now described.

1. The feed line (between valves V1 and V2), the calibrate line (between valves V2 and V3), and the recycle loop (including the reactor and reactor bypass), are evacuated to a low pressure (below 6.5 Pa). The feed line and calibrate line are then filled with the feed gas mixture which is set by the electronic mass flow controllers (not shown). Analysis of the feed mixture is then monitored by a gas chromatograph (GC) to verify the desired reactant concentrations.

2. The feed flow is then diverted to fill the recycle loop (valve V2 is switched to the run line, valve V3 is closed) while the reactor leg is still under vacuum. When a pressure of about 0.12 MPa is reached in the recycle loop, the recycle pump is turned on, the recycle flow is adjusted, the temperature controlled oven is set for the desired temperature, and valve V3 is switched to the run line.

3. When flow and oven temperatures have stabilized, the reactor bypass valves V4 and V5 are switched to the reactor. The recycle loop pressure will immediately drop to about 0.11 MPa, at which time (time zero) the catalyst is exposed to the feed mixture, and the run is started at the desired temperature, pressure and reactant concentrations.

It is obvious from the above procedure that the recycle loop acts as a reservoir that can instantly fill the reactor and expose the catalyst with the desired composition, pressure, and temperature of the reactant gas. Thus, there is no prior history of increasing partial pressure of reactants in contact with the catalyst.

References

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Figure 1.- External Recycle Reactor

Figure 2.- External Recycle Reactor with Reactor Bypass



