

Carberry J. J. Designing laboratory catalytic reactors. *Ind. Eng. Chem.* 56:39-46, 1964.
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To achieve the goal of direct-rate procurement for fluid-solid catalyzed reactions, a unique gradientless spinning-basket reactor was designed and demonstrated. Solid catalysts (or any solid coreactants) are contained in four gas-permeable baskets attached to a shaft and rotated through the well-mixed, continuously fed/withdrawn fluid phase. [The SCI® indicates that this paper has been cited in over 115 publications.]

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Since Sir Humphrey Davy's discovery in 1815 of oxidation catalyzed by metals, one of the many challenges posed by heterogeneous catalysis was establishing reaction velocity (kinetics) as a function of reactant-products concentrations and temperature. These data, when rationalized and elevated to a mathematical form (a reaction kinetic model), would then provide one basis for the elucidation of reaction mechanisms and a basis for the design and scale-up of a catalytic reactor.

Until the 1960s, solid-catalyzed kinetic modeling rested upon treatment of species conversion-contact time data, which yielded rate *per se* indirectly (i.e., the raw data had to be carefully differentiated to secure the rate [velocity] functionality). While in principle, laboratory reactors operated at quite small conversion levels give a direct approximation of rate, the so-called differential (conversion) reactor is plagued by errors associated with low (differential) conversion.

Although sophistication in data reduction techniques rendered the commonly used integral reactor more potent, problems of undetected long- and short-range gradients in concentrations and temperatures cast doubt upon derived catalytic kinetic models. These models should reflect chemical dispositions of the catalysts free of nonchemical diffusional disguises.

Homogeneous kinetic studies of K.G. Denbigh¹ and L.P. Hammett² had taught the merits of the continuous-flow stirred-tank reactor (CSTR) in providing reaction rate (R) directly by a simple material balance about the CSTR:

$$(C = \text{concentration, } \theta = \text{holding time})$$
$$R = \frac{C_{\text{feed}} - C_{\text{exit}}}{\theta}$$

This obviated risky data reduction steps that plagued integral reactor utilization. All well and good for homogeneous reactions, but what of fluid-solid catalyst systems? How would one elevate the CSTR concept to reality for heterogeneously catalyzed reactions?

The notion of the spinning basket catalytic CSTR (now celebrated as the "Carberry Reactor" or, as I would prefer, the "Notre Dame Spinning Basket Reactor"), in which solid catalyst particles are swirled through the uniform, continuously fed/withdrawn fluid phase, was inspired by—of all unscientific things—the common Christmas tree. In 1962 I had envisaged the glittering balls gracing the tree's gentle branches as catalyst pellets. A spinning Christmas tree laden with solid catalysts came to mind. Thus was born the concept of the spinning basket catalytic CSTR.

"The children all snuggled asleep in their beds, while visions of catalysts danced in Dad's head."

In those pioneering, largely pre-Federal Acronym Agency days, monies were scarce. Happily, our then-chairman at Notre Dame, Julius Banchoer, found some unfettered funds. I found a recent graduate of Northwestern University, Dan Tajbl, and persuaded him to pursue his PhD with me (cautiously mumbling to Dan some vague generalities about a spinning catalytic Christmas tree). George Battles, our gifted machinist, quickly fashioned the prototype at the astounding cost of \$47.85. Happily, a Petroleum Research Fund grant then sustained the research and Dan's PhD thesis.³

The Notre Dame Spinning Basket Catalytic CSTR is now a conventional laboratory device permitting direct measurement of catalytic reaction rates and *a priori* detection and corrections for inter-intra-phase concentration and temperature gradients.^{4,5}

The conception and design, no doubt, inspired colleagues to award me the Yale Science and Engineering Association Award for Advancement of Pure and Applied Sciences in 1968, and in 1976, the American Institute of Chemical Engineers' R.H. Wilhelm Award in Chemical Reaction Engineering.

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2. Crowell T I & Hammett L P. Kinetics of the reactions of thiosulfate ion with ethyl, propyl and isopropyl bromides. *J. Amer. Chem. Soc.* 70:3444-50, 1948.
3. Tajbl D G, Simons J B & Carberry J J. Heterogeneous catalysis in a continuous stirred tank reactor. *Ind. Eng. Chem. Fundam.* 5:171-5, 1966. (Cited 90 times.)
4. Carberry J J. *Chemical and catalytic reaction engineering*. New York: McGraw-Hill, 1976. 642 p. (Cited 165 times.)
5. ———. Total oxidation catalysis over supported metals. *Account. Chem. Res.* 18:358-65, 1985.