

Introductory Remarks

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Introductory remarks

Chemical instabilities involve discontinuous changes in qualitative behaviour in response to a gradual change in the experimental conditions. Common examples are the birth and extinction of oscillations or chaos, the onset of explosive behaviour, the propagation of a wave of chemical reaction or the development of patterns. Such changes may occur in laboratory experiments, on an industrial scale or in living systems. They are not pathological curiosities but a natural consequence of rather common features in chemical kinetics: nonlinearity and feedback. Nonlinearities arise whenever a reaction rate depends on concentration in anything other than strict first-order fashion under strictly isothermal circumstances. Feedback may arise through the chemical mechanism (autocatalysis, chain-branching or auto-inhibition) or through thermal mechanisms. The phenomena are widespread through chemical science, and our understanding of such responses can be generalized to many other systems.

The nine papers that constitute this theme begin with gas-phase reactions (Baulch et al. and Proudler et al.) and with gas-solid interactions (Capsaskis & Kenney). In two of these cases, whereas the reactions are exothermic, thermal feedback is as much a consequence as a cause of the remarkable basic behaviour. In butane oxidation, however, as with most hydrocarbon combustion processes, both chemical and thermal feedback routes interact crucially to produce thermokinetic oscillations.

Travelling waves of isothermal chemical reaction were examined as early as 1908 but virtually forgotten until 1939, when Semenov's laboratory applied the then recent work of Kolmogorov and Fisher to cubic autocatalysis. Current interest in chemical waves is extremely active. The paper by Stedman presents experimental results for a new system, while Gray et al. and Needham & Merkin consider some general themes based on the prototype models of such Fisher-Kolmogorov waves. The analytical study of model schemes has played a key role in the development of this subject, and Gasper et al. consider both simple and more realistic schemes to expose the origins of 'canards' or false bifurcations of oscillatory responses. Larter & Steinmetz also study a model, appropriate to the oscillatory and chaotic behaviour exhibited by the horseradish peroxidase system, in which the bifurcations leading to periodic-chaotic sequences involving mixed-mode oscillations are revealed most clearly. Perhaps the most famous of all models for biological systems is the Michaelis-Menten treatment of the rate law for simple enzyme-catalysed reactions. Typically this is justified by considering a situation in which the total enzyme concentration is very small compared with the initial substrate concentration. Maini et al. consider a different scenario, where spatial propagation of substrate into an immobilized enzyme does not satisfy this requirement, but show that the Michaelis-Menten form may still arise if the Michaelis constant is large.

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