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Current Opinion in Chemical Engineering

Integrated design of microwave and photocatalytic reactors. Where are we now?

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Despite many potential advantages, catalytic reactors utilizing electromagnetic energy, such as light or microwaves, lack significant commercial applications. One of the main reasons for this situation is the fact that current microwave and photocatalytic reactor designs suffer from a poor definition of the field geometry and intensity, as well as its interaction with the catalytic material or the processed medium. The present opinion paper discuses those elements of the reactor design, which have not been sufficiently addressed so far. The development of novel integrated reactor concepts should be in continuous dialog with reliable control and design models. Both activities should be supported by instrumentation development to measure electromagnetic field under real process conditions, at relevant spatial and temporal scales.

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Current Opinion in Chemical Engineering 2014, **5**:37–41

This review comes from a themed issue on **Reaction engineering and catalysis**

Edited by **Marc-Olivier Coppens** and **Theodore T Tsotsis**

http://dx.doi.org/10.1016/j.coche.2014.04.002

2211-3398/Published by Elsevier Ltd.

Introduction

Intensification of chemical reactors by application of electromagnetic fields (microwave to UV range) has been extensively discussed in original research papers, position papers and reviews (e.g. [1,2,3•,4•]). *Photocatalytic reactors* can initiate or accelerate redox reactions at low, or even ambient, process temperature implying energy savings compared to thermal reactors [5]. Besides, they can potentially enable chemistry activation directly by sunlight and thus truly sustainable processes. On the low energy part of the spectrum, *microwaves* can provide tailored and selective heating at macroscale and microscale. In several cases, significant process enhancement under microwave heating, compared to conventional heating, is reported. The enhancement usually concerns reduction in process time, improved selectivity and enablement of reactions that would otherwise be impossible [6••]. For certain systems, positive results obtained can be attributed with a fair degree of certainty to the specific microwave heating characteristics, such as fast volumetric heat transfer, superheating of polar molecules above the boiling point and selective heating of catalytic sites. In other cases, the existence of specific or 'non-thermal' microwave effects is claimed [7]. Further, microwave irradiation can facilitate the photocatalytic degradation of pollutants [8–11]. A good review on the subject of microwave photochemistry and photocatalysis is given in [12].

Notwithstanding their process intensification potential, microwave and photocatalytic reactors have found only a very limited range of commercial applications so far. Broadly speaking, there are major *materials* and *reactor engineering* challenges to be addressed. The former concern design of photo-activated catalysts with optimal band gap configuration, energy surface and chemisorption properties as well as design of microwave-activated catalysts with optimal dielectric properties for a given application. The latter concern design, modeling and control aspects of the integrated reactor system as a whole comprising the electromagnetic energy source, the reactor configuration and the catalytic materials involved. Herein, we present our opinion on the necessary developments to advance microwave and photocatalytic reactor design.

Integrated design of microwave reactors

Despite the undisputed distinct microwave heating characteristics, there is still a fair amount of uncertainty as to firstly, which chemical systems can be intensified by the technology; secondly, under what process conditions, and finally, through which thermochemical mechanisms. To a large extent, the reason behind these issues is the poor definition and controllability of the microwave field in commonly employed microwave cavities, whether monomode or multimode ones, for chemical processing.

Poor field controllability results in uncertain process conditions and hinders:

(a) fundamental studies on microwave-chemistry/catalyst interactions under precisely defined electromagnetic and temperature conditions;
(b) process optimization;
(c) reliable scale-up/scale-out.

To address the problem, research has to focus on development of

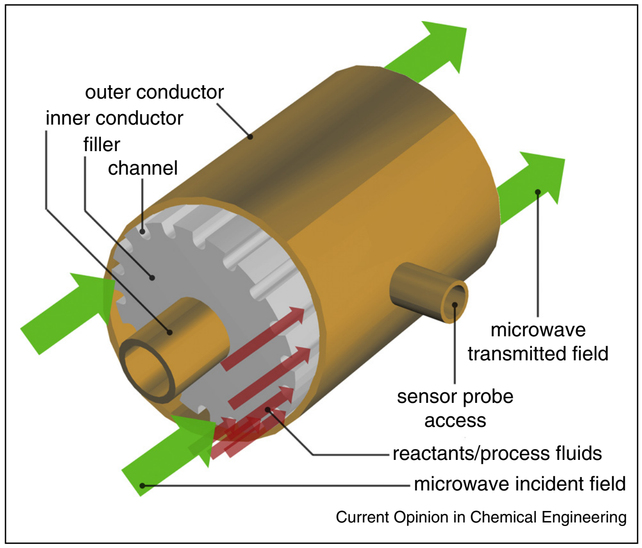
(a) novel integrated microwave-reactor systems;
(b) instrumentation to measure dielectric properties under real process conditions, and *in situ* high temperatures (>300°C) in a microwave field;
(c) suitable models for process control and design.

Integrated microwave-reactor systems

What is lacking is a knowledge base of conceptual options to merge chemical processing and electromagnetics in one system. Although the bulk of research on microwave-assisted processing is conducted using 'traditional cavities', several alternative options already exist. One of them is the range of integrated reactor and microwave transmission systems, under the generic name Labotron™ [13] by Sairem SAS, which is especially designed to carry out batch or continuous flow microwave-assisted synthesis and extraction processes. This range of products employs an internal transmission line to effectively introduce energy into the processing volume. Another relevant example is radio frequency heating, which uses electromagnetic fields of much lower frequencies and electrode applicator systems [14]. The microwave spectrum is defined to range from vacuum wavelengths of one meter to one millimeter. There is no reason to assume that the physics of electromagnetic fields in spectra alongside the microwave spectrum deviate considerably from microwave fields save for their wavelength and frequency. The design principles may thus be extended to the shorter wavelength terahertz or far infrared fields, or to the longer wavelength radio frequency fields. Radio frequency heating is widely applied in industry; a few examples of its applications are: adhesive heating in wood gluing presses, heat treatment of steel and welding of plastics. Because of the longer wavelengths, standing wave patterns would take much more space to emerge in and therefore, spatial field non-uniformities are less pronounced compared to microwave cavities. Additionally, there is much potential to manipulate the electromagnetic field via shaping of the electrodes, but due to the lower frequency, the heating rate is more limited.

There is ample room for novel concepts development. Very recently, in ref. [15•], the concept of traveling wave reactor (TWR) was presented (Figure 1). Traveling electromagnetic waves do not occur in confined spaces. Rather, they travel through a medium in one direction without being reflected by reflective surfaces, such as cavity walls. As they travel, part of the electromagnetic energy is dissipated to heat, according to the dielectric properties of the medium, and the remainder of the energy is transported downstream. Due to the absence of wave interference, standing wave patterns arising from superposition of interfering waves, as in microwave cavities, do not occur. Simulations show that TWRs, firstly, can be optimized for different objectives, such as a uniform heating rate distribution; secondly, can deliver practically useful heating rates, and finally, avoid complex dynamics of resonating electromagnetic fields. The TWR concept may not be the best solution for all microwave-assisted chemistry applications, nor has it been proven in practice, but it does give strong indications that highly controlled and optimized microwave conditions can be realized in chemical processes [15•].

Figure 1



Multichannel traveling wave reactor (TWR) concept. More information can be found in Ref. [15•].

There is one additional dimension to this framework, which is the frequency or, equally, wavelength of the electromagnetic field. As long as the system geometries can practically be scaled to the same ratio as the wavelengths, then findings at a particular frequency also hold at other frequencies, setting aside changes in medium properties that may occur under frequency variation. It would be difficult at present to judge the outcome of explorations in the frequency domain, since — to the best of our knowledge — all relevant studies have been conducted at one frequency only, mostly in the conveniently available 2.45 GHz band. If frequency dependent interactions could be evinced, then this may lead to new ways of controlling chemical processes. However, the first major challenge in such studies would be to develop the required equipment.

Instrumentation

Very frequently, microwave irradiation is applied to catalytic reactors aiming at selective microwave-catalyst interaction. Dielectric properties (real and complex permittivity) of the catalytic material are key to predicting