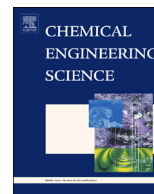




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Remote control of reaction rate by radiofrequency heating of composite catalyst pellets

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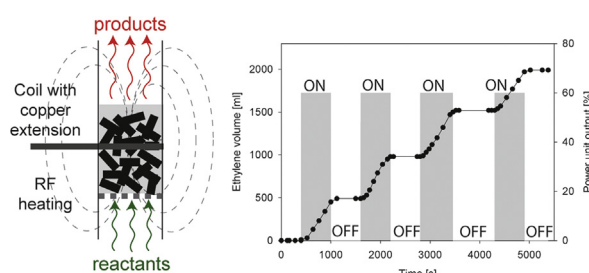
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HIGHLIGHTS

- Composite catalyst pellets susceptible to radiofrequency heating were fabricated.
- Effect of temperature and flowrate on reaction conversion was investigated.
- Transient ON/OFF control of reaction rate was achieved.
- The concept of “reaction on demand” was demonstrated.

GRAPHICAL ABSTRACT



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ABSTRACT

The vapour-phase dehydration of ethanol over alumina has been considered as a model system for demonstrating a novel concept for the “remote control” of an endothermic catalytic reaction. Control of the reaction rate was achieved by forming composite catalyst pellets consisting of porous alumina with embedded metal (Fe) microparticles. Exposing such composite catalyst pellets to alternating magnetic field in the radiofrequency range (900 kHz) resulted in localised heat generation within the catalyst pellets due to induction heating of the iron particles. The effect of the magnetic field power output and reactant flowrate on the steady-state conversion over a differential catalyst bed was systematically investigated. Transient On/Off operation of the reaction was then demonstrated by periodically changing the intensity of the magnetic field and thus controlling the local temperature of the catalyst. This concept enables the reaction product to be formed locally in short bursts, which provides opportunity for process intensification and on-demand synthesis.

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1. Introduction

The concept of chemical reaction “on demand” is quite common in living systems, where various biomolecules are produced by cells only when and where they are needed. The control mechanism is usually based either on controlling the supply of the reactants (substrates), which is regulated by trans-membrane

transport, or by controlling the presence or availability of a catalyst (enzyme), which is regulated through gene expression. Control of the reaction rate in non-living systems under laboratory or industrial conditions can be conveniently achieved by controlling the supply of energy into the system, which relies on the Arrhenius dependence of kinetic rate constants on temperature. If rapid and localised temperature cycling could be achieved, reactions could be switched on and off almost at will. However, traditional modes of heat transfer (convection, conduction, and radiation) pose limitations in this respect. Especially for endothermic catalytic reactions, heat transfer to the centre of a catalyst

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pellet or a catalyst bed can be a limiting step. Alternative means of energy supply that would facilitate rapid and uniform volumetric heating, are therefore of interest.

One possibility is the use of microwave heating (Perry et al., 2002), which was found to have several advantages over traditional methods of catalyst heating (Bhattacharya and Basak, 2013; Komorowska et al., 2009). The main advantage is the ability to supply precisely defined quantities of energy into the catalyst bed or reaction vessel at defined time intervals (Conner et al., 2004). A potential limitation is the risk of forming hot spots due to non-uniform absorption of the microwaves. Another approach is the coupling of endothermic and exothermic reactions (Frauhammer et al., 1999). In that case the endothermic and exothermic reactants are fed sequentially to the same catalyst bed (van Sint Annaland et al., 2002), which effectively acts as a recuperative heat exchanger. Such autothermal setup, however, limits the type of reaction and the purity of products.

Another method capable of heating up complex objects rapidly and uniformly is induction heating. Here the energy is provided to the system by an alternating magnetic field in the radiofrequency range (RF heating). RF heating is based on generating Foucault currents in conductors (Joule heating), or in the case of magnetic materials or nanoparticles, on the Néel and Brownian relaxation processes associated with energy dissipation (Schmid, 2010). RF heating is a scalable method proven at both laboratory and industrial conditions, for example in metallurgy. In the context of chemical engineering applications, the use of RF heating is still relatively limited. Its application for catalyst regeneration (Bhowmick et al., 2012), fluidised bed drying (Stresing et al., 2011) or catalyst testing (Latifi et al., 2014) has been reported. In our recent work, RF heating has been used for the local control of temperature-dependent processes at the small scale by forming composites with micro- or nano-particles that dissipate heat when exposed to RF magnetic field. Thus, remotely controlled release of a chemical payload from stimuli-responsive hydrogels (Zadrazil et al., 2012), composite hollow silica microspheres (Kovačik et al., 2012) or liposomes (Hanuš et al., 2013), as well as RF controlled desorption from a composite adsorbent pellet (Zadrazil and Štěpánek, 2013) were demonstrated. However, in all the above cases, the process that was controlled by RF heating was diffusion, not reaction.

Therefore, the aim of the present work is to explore the potential to control local reaction rate by means of RF induction heating for the energy supply directly to the interior of individual catalyst pellets. As a model endothermic reaction, the catalytic dehydration of ethanol to ethylene over alumina was chosen (Zhang and Yu, 2013). This process requires temperatures in the range of 300–500 °C (Sundaram et al., 2010) and is strongly temperature-dependent. The present work demonstrates that flexible On/Off modulation of the reaction rate via RF catalyst heating enables on-demand product formation.

2. Materials and methods

2.1. Materials

The following chemicals were used. Ethanol for uv/vis spectroscopy (Penta, Czech Republic), aluminium hydroxide (Lach-ner, Czech Republic), LUDOX AM-30 colloidal silica – 30 wt% suspension in water (Sigma Aldrich) and iron (Fe) microparticles (mean particle diameter 25 μm). All chemicals were used as received.

2.2. Composite catalyst fabrication

The catalytic pellets were prepared by mixing aluminium hydroxide (10.0 g) with Fe microparticles (2.5 g) and adding a colloidal silica dispersion (2.5 ml) as a binder. After homogenisation, the suspension was poured into rectangular moulds (fabricated from ABS using a 3D printer) with internal dimensions 4 × 4 × 57 mm and placed into an oven for 3 h at 120 °C. In the next step the dried blocks were removed from the moulds and broken into smaller pellets approximately 4 × 4 × 5 mm, slowly heated in an oven to 500 °C (5 °C/min, air atmosphere) and kept at the final temperature for another 6 h in order to dehydrate the aluminium hydroxide and form aluminium oxide with a porous structure. To increase the mechanical stability of alumina composite pellets, the pellets were soaked for a short time (1 s) in a 10% w/w solution of colloidal silica and dried again. The porous structure of the pellets was analysed by scanning electron microscopy (SEM, Jeol JCM-5700).

2.3. Radiofrequency heating set-up

The catalyst pellets were placed into a glass tube (internal diameter 22 mm) to form a loosely packed bed, which was kept in position by a support grid. The height of the packed bed was 50 mm and the total mass of pellets used was 18.75 g. The glass tube containing the packed bed was covered by a knitted fibreglass insulation sleeve and placed into the axis of an induction coil as shown in Fig. 1. In order to achieve a uniform magnetic field in the entire catalyst bed (22 × 50 mm) a special induction head consisting of single-turn, water-cooled copper coil (26 mm ID) and a cylindrical copper extension (CEIA, Italy) was used. The induction head was connected to a 900 kHz RF generator (PowerCube 64/900, CEIA, Italy).

2.4. Characterisation of the catalyst bed

Temperature of the catalyst during RF heating was monitored by a fibre optic thermometer with a protective Teflon cup, which enabled direct temperature measurement up to approximately

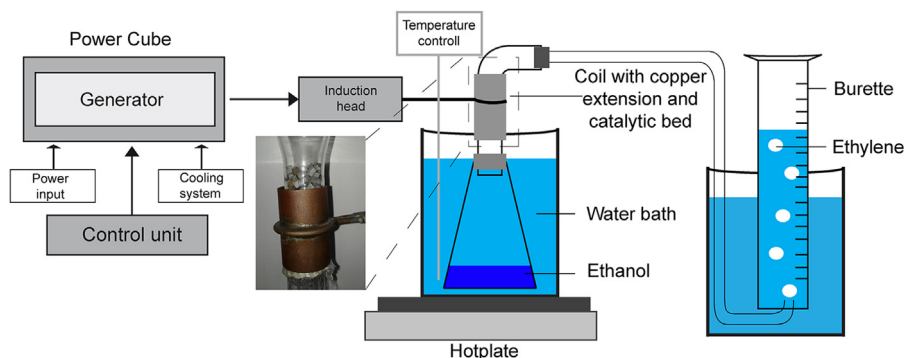


Fig. 1. The apparatus for endothermic catalytic reaction controlled by induction heating.

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