Chemical Engineering Journal 189–190 (2012) 383–392

Contents lists available at SciVerse ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Transient distributions of composition and temperature in a gas–solid packed bed reactor by near-infrared tomography

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ARTICLE INFO

Article history:
Received 20 August 2011
Received in revised form 11 February 2012
Accepted 14 February 2012

Keywords:
Near-infrared tomography
Visualization
Isotopic exchange
Packed bed
Porous media
Transport processes

ABSTRACT

Near-infrared diffuse tomography was used in order to observe dynamic behaviour of flowing gases by measuring the 3D distributions of composition and temperature in a weakly scattering packed bed reactor, subject to wall effects and non-isothermal conditions. The technique was applied to the vapour phase hydrogen isotopic exchange reaction in a hydrophobic packing of low aspect ratio made of platinum on styrene divinyl benzene sulphonate copolymer resin. The results of tomography revealed uneven temperature and composition maps of water and deuterated water vapours in the core-packed bed and in the vicinity of the wall owing to flow maldistribution. The dynamic lag between the near-wall water vapour and deuterated water vapour compositions were observed suggesting that the convective transfer which was significant near the wall at the start, owing to high porosity, was also effective at large conversions.

1. Introduction

The design of gas–solid catalytic reactors requires a detailed knowledge of local dynamics of compositions and temperature which, unfortunately, have not always been easy to measure experimentally. Flow dispersion, channelling and maldistribution are commonly measured by multiple experiments in which invasive probes are placed at different locations [1–4]. However, recent developments in spatially resolved techniques allowed magnetic resonance imaging [5], X-ray [6], neutron [7], and laser spectroscopy [8], to measure anisotropic flow in liquid–solid reactors, achieving rapid access to scalar data of chemical kinetics and transports in a single experiment. These techniques however, have not been sufficiently adaptable to be applied to a wide spectrum of gases, owing to the inherently weak signals from the gaseous phase. Being sensitive to chemical species and boosted by rapid developments in tunable lasers and 2D array detectors, optical techniques are achieving promising spatiotemporal resolutions. Near-infrared (NIR) tomography has been widely used in low scattering media, such as combustion imaging for simultaneous measurements of gas composition by absorption spectroscopy [9] and two-line thermometry [10]. Other examples are the spatial distributions of hydrocarbons at the exit of a combustion chamber [11,12], the spatial breakthrough of water vapour in a packed bed adsorber [13], and the 3D distributions of temperature and concentrations inside and at the exit of a silica gel packed bed adsorber [14]. Notwithstanding a spatial resolution of millimetres inside gas–solid packed beds of small aspect ratios, NIR tomography revealed uneven distributions of temperature and compositions caused by the flow maldistribution, channelling and intra-particle rate limitations [14].

The major limitation of laser-based tomography is commonly related to scattering of the source intensity in non-transparent media, which can introduce substantial errors into the tomographic measurements. Under weak scattering operation, analytical methods to measure light propagation such as the pseudo- Beer law model is attracting increasing popularity than the statistical methods [15–17].

$$\mathrm{Tr} = \frac{I_{\mathrm{exit}}}{I_{\mathrm{inlet}}} = e^{-\mathrm{OD}} \quad (1)$$

where Tr is transmittance, I_{exit} and I_{inlet} are light intensities at the exit and the inlet of the packed bed [15]. The OD is the optical depth which provides an estimation of the average number of times that

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1385-8947/$ – see front matter
doi:10.1016/j.cej.2012.02.037

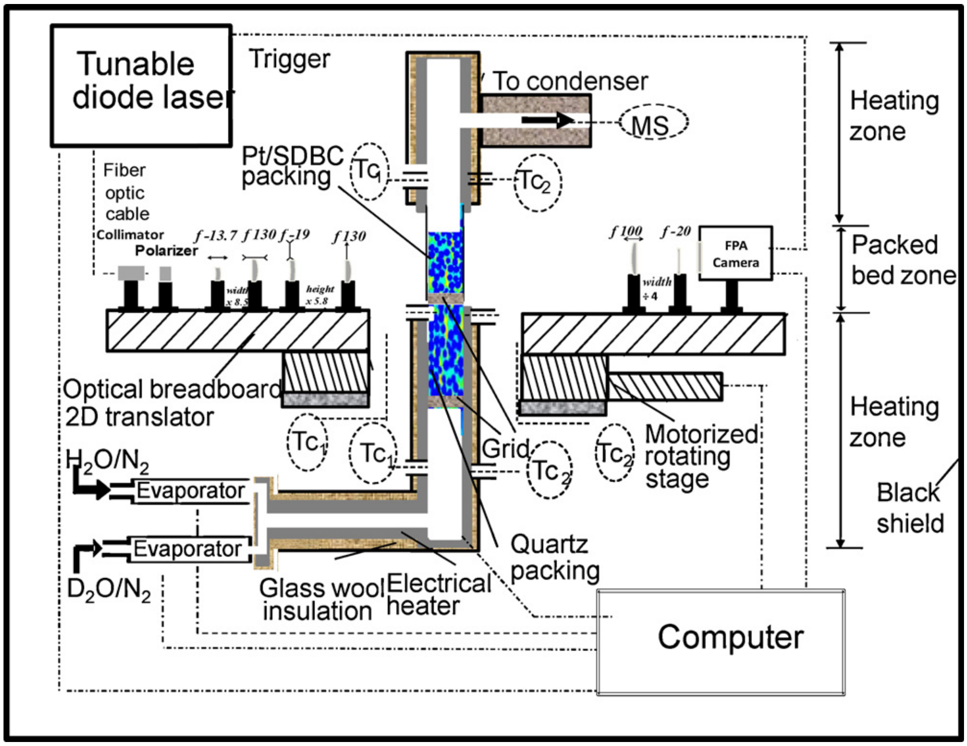

Fig. 1. Scheme of the whole tomography apparatus: evaporator (Bronkhorst) = mass flow controller (N_2), air-actuated switching valve, distilled water bath; H: humidity sensor; TC_1 = thermocouples (monitoring), TC_2: thermocouples connected to programmable temperature controllers; MS: mass spectrometer; optics = details of optics are shown in Fig. 1; tuneable diode laser: lock-in monomode connected to FPA camera; quartz packing: pre-packed bed mixer, Pt/SDBC packed bed = height:14 cm; visible aperture by NIR camera: $1.20 \times 1.82\,cm^2$. Monomode fibre-optic cable connected to a collimator with a top-hat beam shaper (size: 5 mm); polarizer, two couple of cylindrical lenses of focal points *f* (mm): *f*-13.7, *f*-130 and *f*-19, *f*-130, trans-illuminated backed bed and couple of lens (*f*-100, *f*-20) and intercepted beam size by FPA detector.

photons interact with solid particles, via absorption and scattering, prior to exiting a medium of length *l*.

$$OD = l\mu_e$$

where μ_e is the extinction coefficient. The imaging is, therefore, constructed by photons which travel through the sample straight (ballistic photons), slightly scattered (OD < 1), scattered or diffusively (multiple scattering, OD > 1) [15–17]. In this work, NIR tomography is used to visualize the spatial distributions of compositions and temperature in a gas–solid packed bed reactor, subject to wall effects, intra-particle rate resistance and non-isothermal conditions. The technique's capability is extended to the vapour phase of hydrogen isotopic exchange (HIE) reaction between deuterated water and hydrogen, where the spatial distributions of both water vapour (H_2O_v) and deuterated water vapour (HDO_v) are obtained simultaneously in a packed bed reactor, filled with platinum catalyst, supported on hydrophobic styrene–divinyl-benzene copolymer (Pt/DSBC) resin. Unlike adsorption process where the heat produced interfere with the thermal exchange from the heating wall [13,14], the HIE will not show heat exchange from the process as it is thermally neutral. Owing to its high separation factors and near-ambient operating conditions, the HIE reaction is very attractive for processing active wastes generated in heavy water moderated-nuclear reactors, and future fusion machines [18,19]. Similar to silica gel adsorber [14], the Pt/DSBC resin reactor is expected to show flow maldistribution caused by the small aspect ratio of the packed bed, used in conjunction with the intra-particle diffusion in Pt/DSBC catalyst as reported by Izawa et al. [20] and Kawakami et al. [21]. Since the kinetic behaviour of the vapour phase HIE reaction is inhibited significantly by the adsorbed waters, hydrophobic supports are preferred to achieve high exchange rates at the highest relative humidity (RH), i.e. RH of 90% with Teflon-carbon mixtures and saturated H_2O_v with Pt/SDBC resin, respectively [22]. The overall mechanism is considered to include steps of the reaction and the mass transfer between the vapour and the liquid phases.

$$H_2O_v + HD\,(g) \Leftrightarrow HDO_v + H_2\,(g) \tag{2}$$

$$H_2O\,(l) + HDO_v \Leftrightarrow H_2O_v + HDO\,(l) \tag{3}$$

The first reaction occurs only at catalytic sites of the hydrophobic catalyst and the second is driven by the vapour–liquid equilibrium of water and HDO_v mixture, where rates of HDO_v absorption and H_2O evaporation are commonly accelerated by adding hydrophilic supports. As a result of flow channelling and liquid distribution around the catalytic particles, an arrangement of mixed hydrophobic and hydrophilic packing revealed lower exchange efficiency compared with a unmixed arrangement, where the vapour phase HIE and phase equilibrium reactions are functionally separated from each other [23]. In this work, the derived transient changes of H_2O_v, HDO_v and temperature of the vapour phase HIE reaction, between heavy water vapour (D_2O_v) and hydrogen gas, are described by 3D distributions inside and at the exit of a packed bed reactor of low aspect ratio, by using NIR tomography. The main design employs a continuous wavelength (CW) tunable diode laser, focal planar array (FPA) detector and Pt/SDBC resin packing of low scattering effect of NIR light.

2. Material and methods

The NIR tomography set-up was designed by collecting data from the 2D projection of the full packed bed. 3D distributions of temperature and composition inside and above the packing were obtained simultaneously. The reactor and optical set-ups are shown in Fig. 1. The tube was made of fused quartz of 9 mm I.D. and filled