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^{24}Na and ^{99m}Tc Tracers Applied to the Characterization of Liquid–Solid Fluidized Bed and Hydraulic Transport Reactors

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For the characterization of fluidized bed- and hydraulic transport reactors, two radiotracer techniques have been developed permitting the study of mixing phenomena in flowing liquids as well as in beds of fluidized particulate solids. For labelling the aqueous phase, a ^{99m}Tc containing solution obtained by means of a ^{99}Mo isotopic generator has been used. The silica gel employed for fluidization has been activated by thermal neutrons to yield a ^{24}Na tracer derived from its natural sodium content.

Introduction

Chemical processes in steady-state flow systems depend not only on the mean residence time of the substances in the flow system but also on their residence time distribution resulting from channelling or dispersion phenomena, generally called backmixing. In consequence the degree of backmixing must be known for a given reaction system and reactor to assure a reliable design. The analysis of backmixing is generally performed by tracer experiments. Since the analysis of chemical tracers in reaction systems, especially those containing more than one phase, can be very difficult, radioactive tracers of short decay periods can be used for this purpose.[1]

Radioactive tracers allow continuous monitoring from outside the reaction system with an independent choice of detector position. In addition, the detectors do not distort the results by disturbing the flow[2] and the tracer concentration can be measured over the whole cross-section of the flow system. Thus disturbances due to local flow fluctuations are essentially suppressed.

The radioactive tracer technique is therefore the best means of characterizing reactors with special geometries, reactors operated under high pressure and for multi-phase reaction systems, e.g. gas–liquid[3] and liquid–solid systems.[4] Radioactive tracers have been employed in the pulp industry[5] to study the transport of cellulose fibres labelled with ^{24}Na doped glass fibres.[6] Liquid–solid fluidized beds are employed for processes like extraction, crystallization, leaching and heterogeneous reactions including the use of immobilized cells and enzymes.[7] The operation of fluidized beds however, is impaired by large backmixing and hydrodynamic instabilities when low fluid flow rates are applied in systems of large cross-section. These problems are frequently encountered in pilot and industrial scale installations when the particulate solid is of fine grain size or the density difference between the solid and liquid phases is small. To circumvent these problems, special motionless mixers have been introduced into fluidized beds. This results in a stabilization of the bed and a considerable increase in the operating range with respect to flow rate.[8] Preliminary results obtained by a tracer response method based on conductivity measurements, which cannot be carried out without disturbing the flow profile, show that backmixing may be decreased considerably by applying motionless mixers.[8]

A method based on tracer measurements employing radioisotopes with a short half-life has been worked out for a reliable characterization of both the backmixing of the liquid phase and the mixing of the solid phase in fluidized bed reactors. This was achieved by using a soluble ^{99m}Tc tracer and solids

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which could be labelled with ^{24}Na. The method has first been verified by measuring the residence time distribution of both liquid and solid tracers in an hydraulic transport reactor where the velocities of both phases are essentially equal. Then it has been applied to the characterization of fluidized bed reactors where only the liquid phase flows through the reactor whilst the solid is kept in a fluidized state. Results will be given to demonstrate the feasibility of the radioactive tracer response method. A detailed characterization of fluidized bed reactors with and without internals will be published elsewhere.

Materials and Methods

The liquid–solid phase system employed, consisted of water and an irregularly shaped porous silica gel (gel type 432, Grace, Worms) with a particle size in the range 0.1–0.16 mm, a true density of 2040 kg·m^{-3} and an apparent density in water at 50 C of 1340 kg·m^{-3}.

Tracers

The 6-h ^{99m}Tc was eluted from a ^{99}Mo generator as sodium pertechnetate with 0.9% NaCl yielding a volumetric activity of about $15 \cdot 10^8$ Bq·mL^{-1}. The counting was based on the 140.5 keV γ-ray. Since the silica gel contains about 0.1 wt% of sodium, it could be labelled with 15-h ^{24}Na by activation with thermal neutrons. Samples of 1 g silica gel were irradiated for 15 h in a flux of $1.5 \cdot 10^{13}$ n·cm^{-2}·s^{-1} at the SAPHIR reactor at the Swiss Federal Institute for Reactor Research, Würenlingen, yielding a ^{24}Na activity of about $3.7 \cdot 10^8$ Bq·g^{-1}. The activated silica gel had to be suspended in a small volume of water under low vacuum in order to evacuate the air contained in the pores prior to its use. The counting was based on the 2.75 MeV γ-ray.

Equipment

The installation is shown schematically in Fig. 1. For fluidized bed experiments, a reactor has been employed with a height of 1.6 m and an i.d. of 40 mm. The reactor consisted of a jacketed glass column to assure temperature stability.

It was equipped with motionless mixers, type SMX (Sulzer, Winterthur) with an hydraulic diameter of 11 mm. The entrance for the main flow of water was designed as a cone filled with 1 mm glass beads, kept in place between stainless steel screens, to provide an essentially equal liquid distribution over the whole cross-section at the reactor entrance.

For analysing the backmixing in the liquid phase, a secondary flow was fed at the rate attaining about 4% of the main flow and was introduced into the reactor just above the upper screen of the cone. This secondary flow could be labelled with the ^{99m}Tc tracer by means of an HPLC injection valve. The lower detector was located 0.3 m, and the upper detector 1.3 m, above the entrance.

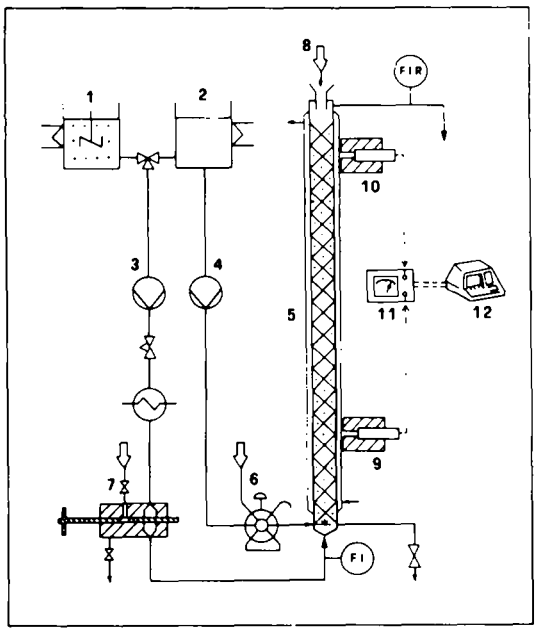

Fig. 1. Experimental installation. 1- Silica gel suspension; 2—water; 3– main flow; 4– secondary flow; 5- jacketed column; 6- liquid tracer injection system; 7- solid tracer injection system; 8—solid tracer introduction for fluidized beds; 9— lower NaI(Tl) detector (LD); 10—upper NaI(Tl) detector (HD); 11 – ratemeter; 12—Plessey Micro II minicomputer; FI—flow indication; FIR- flow indication and recording.

For the analysis of the solid phase mixing, the ^{24}Na labelled silica gel tracer was introduced to the top of the fluidized bed. During these experiments the fluidized bed had to expand to the upper mixing elements (1.5 m), so that the amount of silica gel had to be adjusted when the flow rate was changed. The detectors were placed at 0.5 m and 1 m above the reactor entrance.

In case of the hydraulic transport reactor, a stainless steel column, with a height of 1.6 m and an i.d. of 32 mm, has been used. The reactor contained motionless mixers of the type SMXL (Sulzer, Winterthur) with a hydraulic diameter of 16 mm. The reactor was fed with an aqueous suspension of silica gel at room temperature. An empty cone at the bottom of the reactor served to provide an undisturbed distribution of the suspension. The soluble ^{99m}Tc tracer was introduced as described for the fluidized bed experiments. The solid tracer was injected into the main flow just before the reactor entrance by means of a piston provided with a chamber which contained the tracer. The detectors were located at a distance of 0.3 and 1.3 m above the reactor entrance.

Detection

Two shielded NaI(Tl) detectors 44.5 × 44.5 mm were mounted normal to the reactor axis and could be adjusted to any height parallel to the axis. For ^{99m}Tc a front shielding of 50 mm was sufficient,

whereas for ^{24}Na a front shielding of 100 mm was needed. The distance between the detector and the external surface of the reactor was equal to the thickness of the front shielding. For both tracers the radial shielding had a thickness of 40 mm. In the case of ^{24}Na the radial shielding was fortified parallel to the reactor axis by lead blocks of 150 mm in each direction to suppress effectively the angular response. Since the local tracer concentration has to be analysed, the front shieldings contained a rectangular aperture with a height of 3 mm. For ^{24}Na experiments, the aperture height was 5 mm. The aperture width was always 60 mm to assure an analysis over the whole cross-section of the reactor. A window of 70–270 keV was used for ^{99m}Tc and of 250–3100 keV for ^{24}Na. The outputs of the ratemeters were connected to a dual channel recorder and a minicomputer for data storage and treatment.

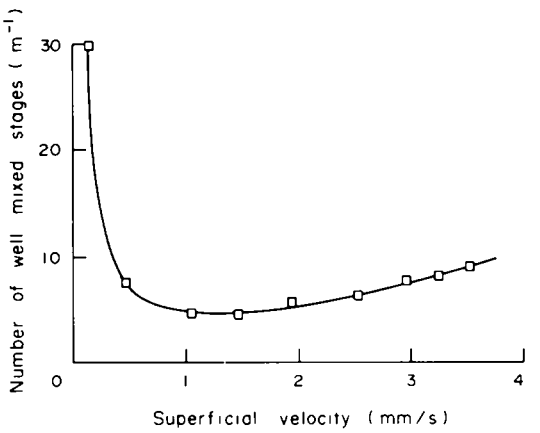

Fig. 3. Axial backmixing of the liquid phase in fluidized beds equipped with internals of the type SMX. Expansion: 1.1–3.0; temperature: 50 C.

Results and Discussion

Backmixing of the liquid phase

The evolution of the tracer concentrations as a function of time was measured at two sites along the reactor after injection of 0.5 mL aqueous solution containing $7.4 \cdot 10^7$ Bq ^{99m}Tc.

Since only small amounts of ^{99m}Tc have been applied, it was necessary to demonstrate that the tracer does not adsorb on silica gel. This has been proven by applying pulses of tracer to the entrance of a fluidized bed reactor and analysing each response in the outlet pipe (closed reactor). The mean residence times evaluated by means of the momentum method were equal to the hydraulic residence times. Adsorption phenomena would have increased the mean residence time of the tracer.

Figure 2 shows the normalized response of the lower detector (LD) and the upper detector (HD) for a fluidized bed experiment. The areas under the distribution functions have been normalized to unity after deduction of the background radiation. The statistical noise level does not exceed 10% of the signal. For the characterization of backmixing the convolution integral method has been applied in conjunction with the parameter estimation algorithm of Marquardt. The sum of the squares of the deviations between the second response measured and that calculated, applying common models for real reactors, has been minimized.

The axial backmixing as a function of the liquid superficial velocity is shown in Fig. 3 for a fluidized bed equipped with internals of the type SMX. The degree of backmixing is given according to the tanks-in-series model as the number of equivalent well mixed stages per meter reactor length. The liquid linear velocities applied correspond to bed expansions in the range of 1.1–3. At very low flow rates the transition from a fixed bed to a fluidized bed has been observed. Therefore, the number of tanks as a function of the fluid velocity decreases steeply at low fluid velocities due to the sudden increase of backmixing. When the flow rate is further increased, the reactor

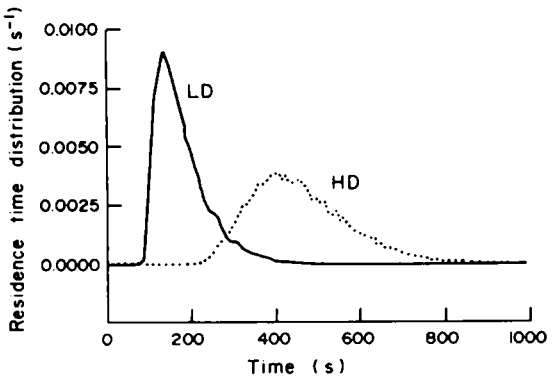

Fig. 2. Residence time distribution of the liquid phase in a fluidized bed reactor equipped with internals of the type SMX. Expansion: 2.73; superficial velocity: 3.27 mm·s^{-1}; temperature: 50 C.

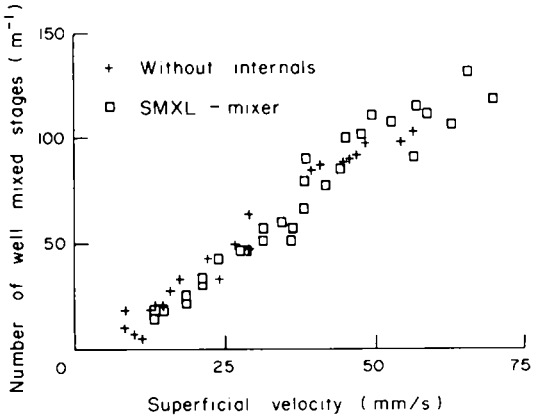

Fig. 4. Axial backmixing of the liquid phase in hydraulic transport reactors with and without internals of the type SMXL. Solid phase concentration: 5%; temperature 25 C.

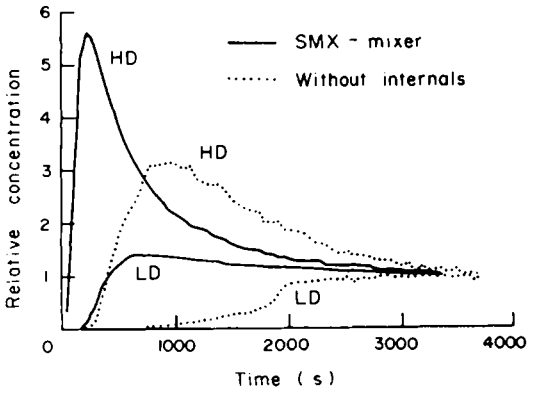

Fig. 5. Axial mixing of the solid phase in a fluidized bed with and without internals of the type SMX. Expansion: 2; temperature: 50°C.

behaviour is improved by intensification of the turbulence. When the internals were removed, flow instabilities caused the fluidized bed reactor to exhibit irreproducible variations in backmixing.

The backmixing behaviour of the liquid phase in the hydraulic transport reactor is shown in Fig. 4 for a reactor without internals and one with internals of the type SMXL. An aqueous suspension of 5% silica gel has been applied. At these high fluid velocities the internals have essentially no influence on the degree of backmixing.

Solid phase mixing in fluidized beds

The tracer technique for ^{24}Na labelled silica gel has been applied to measure the backmixing of the solid phase in the hydraulic transport reactor. But the main interest has been directed towards the mixing phenomena of particles in fluidized beds. The internals employed are able to stabilize the fluid flow profile and increase the apparent final velocity of solid particles by a factor of 2.3.[8]

Figure 5 shows the relative tracer concentrations in fluidized beds with, as well as without internals at two different sites along the reactor, as a function of time. The fluidized bed has been operated in both cases at a bed expansion of 2. The tracer concentration was normalized by fixing the final concentration of the evenly distributed tracer to unity.

The experiments show that the mixing rate of the solid phase is enhanced by the use of motionless mixers, of the type SMX, more than 3-fold. The obvious difference between the maximum tracer concentrations reported in Fig. 5 seems to indicate that internals may entirely alter the mixing mechanism. The extremely high maximum value measured by the upper detector (HD) for the fluidized bed with internals can be explained by the assumption that the solid phase is segregated in one solid-rich and another solid-poor phase with a rapid interchange of particles.

Conclusion

The characterization of reactors, with respect to backmixing of liquids as well as particulate solids, has been achieved by the development of tracer techniques for the labelling of both phases. These methods have been applied to the study of fluidized bed and hydraulic transport reactors. The main advantage of these techniques, besides the obvious difficulty of applying other continuous analytical methods to such systems, is that the movement of media in the reactor is not disturbed by the detectors.

The neutron activation of sodium traces within the silica gel has proved to be a very versatile method for the production of particulate tracers, since it conserves the physical properties of the original material. This is extremely important for mixing studies of solids in the fluidized state.

Radiocontamination problems are avoided by the use of small amounts of radioactive material of short half-life.

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References

1. Vogg H. *Chem. Ing. Tech.* **50**, 729 (1978).
2. Brewster B. S. and Seader J. D. *A.I.Ch.E. Journal* **26**, 325 (1980).
3. Evans G. V., Robertson J. M. and Spackman R. *UKAEA Report* No. AERE-R6753 (1971).
4. Toda M., Ishikawa T., Saito S. and Maeda S. *J. Chem. Eng. Jpn* **6**, 140 (1973).
5. Ljunggren K. In *Radioisotopes in the Pulp and Paper Industry*, Panel Proceeding Series. (IAEA, Vienna, 1968).
6. Moreira R. M. and Felder R. M. *A.I.Ch.E. Journal* **25**, 131 (1979).
7. Renken A., Flaschel E. and Fauquex P. F. *Enzyme Eng.* **6**, 309 (1982).
8. Flaschel E., Fauquex P. F. and Renken A. *Chem. Ing. Tech.* **54**, 54 (1982).