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Diana Carolina Guío-Pérez, Tobias Pröll, and Hermann Hofbauer, "Non-Intrusive Online Detection of Ferromagnetic Particles for Measurement of Bed Density and Residence Time Distribution in Circulating Fluidized Bed Systems" in "The 14th International Conference on Fluidization – From Fundamentals to Products", J.A.M. Kuipers, Eindhoven University of Technology R.F. Mudde, Delft University of Technology J.R. van Ommen, Delft University of Technology N.G. Deen, Eindhoven University of Technology Eds, ECI Symposium Series, (2013). http://dc.engconfintl.org/fluidization_xiv/118

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NON-INTRUSIVE ONLINE DETECTION OF FERROMAGNETIC PARTICLES FOR MEASUREMENT OF BED DENSITY AND RESIDENCE TIME DISTRIBUTION IN CIRCULATING FLUIDIZED BED SYSTEMS

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ABSTRACT
This work presents the progress made in the last years at the Vienna University of Technology on the design and optimization of a non-intrusive method, based on inductance changes of a coil and able to quantitatively detect fluid-dynamically similar ferromagnetic tracer particles in a fluidized bed cold flow model. The method finds application not only in RTD determination but also in the measurement of solids circulation rate and bed density in similar units.

INTRODUCTION
The use of gas-solid reactors is widely spread; more precisely, circulating beds are increasingly present in many applications and processes due to their advantages regarding mass and heat transfer and their operational flexibility. The efficiency and extent of the thermal and chemical exchange processes that take place in a fluidized bed reactor strongly depend on the contact efficiency and the contact time between the phases. These factors are particularly important in catalytic processes and in those where high reactivity is critical. Investigations on residence time distribution (RTD) as well as on distribution of solids aid in understanding the fluid-dynamics, and are essential for reactor design, plant operation and optimization of existing circulating fluidized beds.

Finding adequate tracer materials and detection methods to make these analyses is difficult. This work presents the progress lately made at the Vienna University of Technology on the design, construction and optimization of a non-intrusive online method able to detect, with high resolution, ferromagnetic tracer particles in a fluidized bed cold flow model.

IDENTIFICATION OF REQUIREMENTS
After a careful literature review on existent tracer methods used to determine residence time distribution of solids in fluidized beds (1), (2), and considering that the system was planned to be implemented in cold flow models, the requirements and expected features of the RTD measurement system were determined. An impulse-response tracer measurement was selected.

Tracer particles were expected to hold very similar fluid-dynamically-relevant properties in comparison with the bulk of bed material and not to involve health risks. With regard to the measurement system, some of the most important conditions desired were:

- Sensitivity of the measurement should be as high as possible; use of a small mass of tracer is advantageous.
- The measurement should be reproducible and repetitions should be possible within reasonable periods of time.
- Ideally, calculation of tracer concentration (and hence RTD) should be possible

without need of intricate assumptions or corrections.

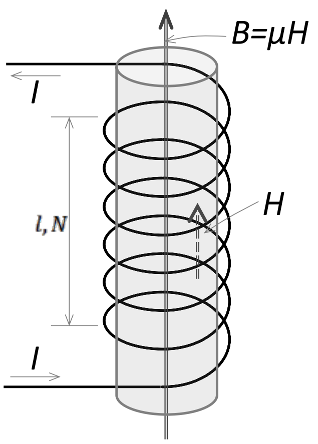
- The detection device or method should not imply any disruption of the internal flow pattern (particularly, pressure and inventory should not be effected).
- Measurement should be possible under steady state operation conditions.
- The response of the detection method should be fast enough to capture the features of RTD in fast beds, which change in very short time.

On this basis, ferromagnetic particles were chosen as good candidates to be used as tracer in a cold flow model where heavy particles, such as bronze, are used (this is to maintain the fluid-dynamic similarity with the hot unit e.g. combustors, gasifiers or chemical looping units). Ferromagnetic particles exhibit a number of advantages for their use in such tests (3), (4): they neither require special handling nor are they toxic; the density and size can be modified to fit fluid-dynamic requirements by forming a composite with a polymer; the magnetic properties of the material do not deteriorate with time or use, and temperature has only a slight influence as long as kept below certain limits (Curie temperature of Fe 770°C); the particles can be easily separated from bed material by means of magnets; and most importantly, a simple coil inductor can be used for the detection of tracer particles.

MEASUREMENT PRINCIPLE

If a magnetic field is generated by means of a coil of N turns, a length l, a cross section area of the conductor A, and carrying a current of magnitude I, then the magnetic field strength designated by H is given by Equation 1 (5). The magnetic induction, or magnetic flux density, denoted by B, represents the magnitude of the internal field strength within a substance that is subjected to a magnetic field of strength H (Figure 1). The magnetic field strength and flux density are related according to Equation 2, where μ is the permeability. In vacuum $B_0=\mu_0 \cdot H$, where μ_0 is the permeability of a vacuum, a universal constant. The ratio of the permeability in a material to the permeability in a vacuum is as shown in Equation 3, where μ_r is called the relative permeability. For a cylindrical coil with a core different than vacuum (Figure 1) the inductance is given by Equation 4.

Thus, the inductance L will change proportionally to μ_r (6). In this way, the presence of dispersed ferromagnetic particles in the core area of a coil would influence the coil's inductance in certain proportionality with respect to the concentration of ferromagnetic material in the core volume. The chance in inductance could be analyzed in order to allow quantitative concentration measurement.



$$H=NI/l. \quad (1)$$

$$B=\mu H \quad (2)$$

$$\mu_r=\mu/\mu_0 \quad (3)$$

$$L=\mu_0\mu_r\frac{N^2A}{l} \quad (4)$$

Figure 1: Diagram of the magnetic field generated by a cylindrical coil (H) that transports the current I. Core different than vacuum. N: number of turns, l: coil length, B_0: magnetic flux density in vacuum, μ_0: permeability of a vacuum, B: magnetic flux density within a solid material, μ: permeability of the solid material.

VERIFICATION OF THE PRINCIPLE

First experiments were focused on testing the suitability of a simple coil system to reflect the proportionality between ferromagnetic particles concentration and inductance changes. The inductance of a coil was measured using a simple LC resonator.The measurement was based on the resonance frequency of the resonator (2). Tracer particles and bed material were characterized and it turns out that fluid-dynamic characteristics in terms of the Archimedes number agrees well for the chosen ferromagnetic tracer particles (Table 1).

Table 1: Properties of the bed material (bronze particles) and the tracer material (steel particles).

Parameter	Bronze	Steel	Units
Particle density (ρ_P)	8730	7579	$kg \cdot m^{-3}$
Sauter mean particle diameter (d_p)	$6.80 \cdot 10^{-5}$	$7.20 \cdot 10^{-5}$	m
Particle sphericity (φ)	1	1	--
Archimedes number (Ar)	$1.07 \cdot 10^{2}$	$1.11 \cdot 10^{2}$	--
Reynolds number (minimum fluidization, Re_{mf})	$8.03 \cdot 10^{-2}$	$8.28 \cdot 10^{-2}$	--
Minimum fluidization velocity (U_{mf})	$1.69 \cdot 10^{-2}$	$1.65 \cdot 10^{-2}$	$m \cdot s^{-1}$

The inductance sensor (coil) was installed around the pipe where particles were fluidized. The change in concentration of ferromagnetic material in the core of the coil was reflected on the inductance measured (Figure 2 A.). The results evidence the reproducibility and the accuracy of the measurements. The proportionality between the measured parameter and the concentration of ferromagnetic material in the core of the coil was found. However, because the resonance frequency was the variable measured, the time resolution of the measurement could only be improved to the detriment of the accuracy. Thus, in order for the signal from the coil to be used to determine RTD, the signal needed to be processed in an alternative way.

DESIGN OF THE RTD MEASUREMENT SYSTEM

An improved system for determination of inductance, based on an impedance circuit bridge was constructed. It consists of a specially developed multichannel carrier frequency amplifier with a very high amplification factor that enables a high sensitivity and quality of the measured signal, even with a very small percentage of ferromagnetic particles (7).

The system chosen to implement the measurement is a cold flow model of a dual circulating fluidized bed (DCFB) designed for chemical looping (8), (9). The system (Figure 3) consists of two interconnected circulating fluidized beds. In the main loop, particles are fluidized in the main reactor (air reactor, AR, internal diameter 50mm), separated in a cyclone and fed into the secondary reactor (fuel reactor, FR, internal diameter 54mm) after passing through a seal (upper loop seal, ULS). The loop is completed by a seal located at the lower end of the reactors (lower loop seal, LLS) and the flow of particles in this loop is called global circulation rate. The secondary loop includes the secondary reactor (FR) and a system for internal recirculation of particles, cyclone and seal (internal loop seal, ILS) and is of special interest regarding solids gas contact. The RTD measurement system is designed and optimized for the secondary reactor (FR) and the use of an extremely small amount of tracer, about 0.05wt.% of the total inventory.

The configuration avoids invasive procedures or elements in both injection of tracer and sensing of concentration. For the injection, instead of using the common air pulse injection devices, a system was built to collect the tracer at a certain point in the unit in order to use the same particles for the next experiment. The position of the collecting point was selected such that it serves directly as an injector, i.e. close enough to the input signal measuring point.

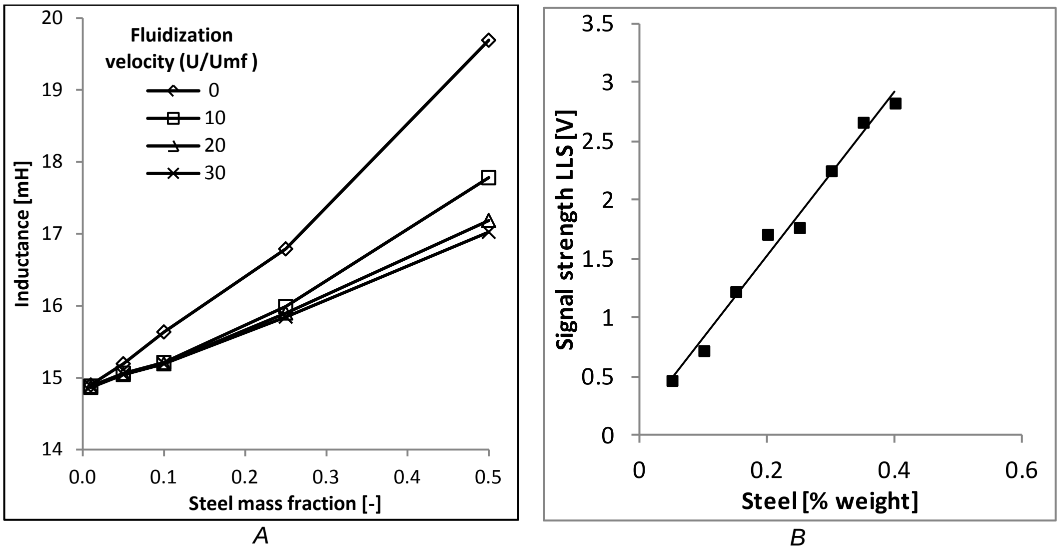


Figure 2: A: Inductance measured with LC resonator at fixed bed and fluidized bed conditions for different compositions of bronze/steel mixtures. B: Signal strength in volts measured by impedance method for different concentration of ferromagnetic particles.

The collection/injection system was placed at the lowest end of the AR cyclone body (along the whole perimeter) and consists of a number of strong permanent magnets in a row that collect at the wall the ferromagnetic particles passing through the cyclone. The injection is done by pneumatically removing the magnets from the wall; the particles fall immediately and flow together with the stream of bronze powder. With such a device a series of measurements can be carried out with the same inventory alternating periods of collecting and measuring. The operating conditions can be set before the injection is done and, therefore, the measurement is made during stable operation. No additional process is needed to clean or regenerate the bed material. The magnets arrangement is able to separate the ferromagnetic particles from the solids stream even for very low concentration (few grams of tracer).

To monitor the concentration of ferromagnetic particles in the stream, three coils were placed on the sensing points. The ducts did not need to be modified in any way for the installation of the coils. The input signal was measured at the only solids input to the FR, namely at the ULS. The closeness between injection and input measurement point guarantees a high quality impulse-type input signal. Since the measurement is done in the outlet duct of the loop seal, the residence time in the seal itself is not taken into consideration. Two points are taken for output signal measurement, namely one in each of the solids outlets of the FR, namely, one in the LLS for the global loop and one in the ILS for the internal loop.The loop seals were selected for the concentration measurements since they provide the zones with the highest particle density available, a fact that enhances the resolution and accuracy of the measurement. A diluted bed would increase the sensitivity requirement.

The proportionality between ferromagnetic particles concentration and signal strength was proven with the impedance system as well; the correlation is presented in Figure 2 B. The linear proportionality allows a direct calculation of RTD.

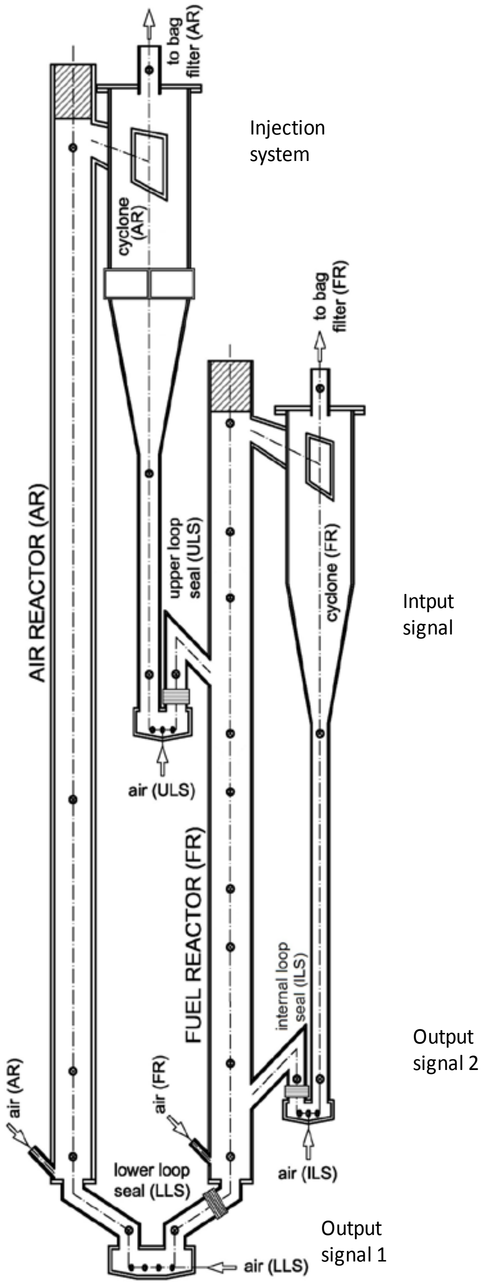


Figure 3: Cold flow model scheme indicating the main parts and the location of measuring coils and of the injection system. AR: 50mm internal diameter and 1.3m height, FR: 54mm internal diameter and 1m height.

RTD CALCULATIONS AND MODELING

In this work, the **E** curve that relates the experimental input and output signals is found on the basis of a previously proposed ideal system, which should approach the conditions expected to be found in the reactor for the corresponding experiment. This proposed transfer function is used to calculate a simulated output signal based on the experimental input signal and according to Equation 5, where $C_{out}(t)$ and $C_{in}(t)$ are the normalized out- and input curves. The mean residence time, τ, is defined as the first moment of the distribution curve if there are no dead, or stagnant zones within the reactor (Equation 6).

$$C_{out}(t) = \int_0^t C_{in}(t')E\,(t-t')dt' \qquad (5)$$

$$\tau = \int_0^\infty t\,E(t)dt \qquad (6)$$

The model consists of a combination of ideal reactor models for which the **E** functions are known. The simulated output is then compared to the experimental output, and the divergence of the simulated signal is minimized by means of a least squares fitting routine. The ideal reactor models considered, for which the residence time distribution is well-known are the following, e.g. from Levenspiel (10):

Plug flow:

$$\boldsymbol{E}(t) = \delta(t-\tau) \qquad (7)$$

Ideally mixed stirred tank:

$$\boldsymbol{E}(t) = \frac{1}{\tau}e^{-t/\tau} \qquad (8)$$

Plug flow with dispersion:

$$\boldsymbol{E}(t) = \sqrt{\frac{u^3}{4\pi \boldsymbol{D}L}}\; e^{\left[-\frac{(L-ut)^2}{4\boldsymbol{D}L/u}\right]} \qquad (9)$$

Ideally mixed stirred tanks in series:

$$\boldsymbol{E}(t) = \left(\frac{t}{\bar{t}_i}\right)^{n-1} \frac{1}{(n-1)!}\; e^{-t/\bar{t}_i} \qquad (10)$$

RESULTS

The proposed model looks for a real representation of the actual unit and its operation principles (Figure 4). Three main zones are identified:

(a) The internal loop that includes the upper part of the fuel reactor, the FR-cyclone, the FR down-comer and the internal loop seal. This section is considered to have flow of particles in only one direction; this flow would be equivalent to the internal circulation rate.

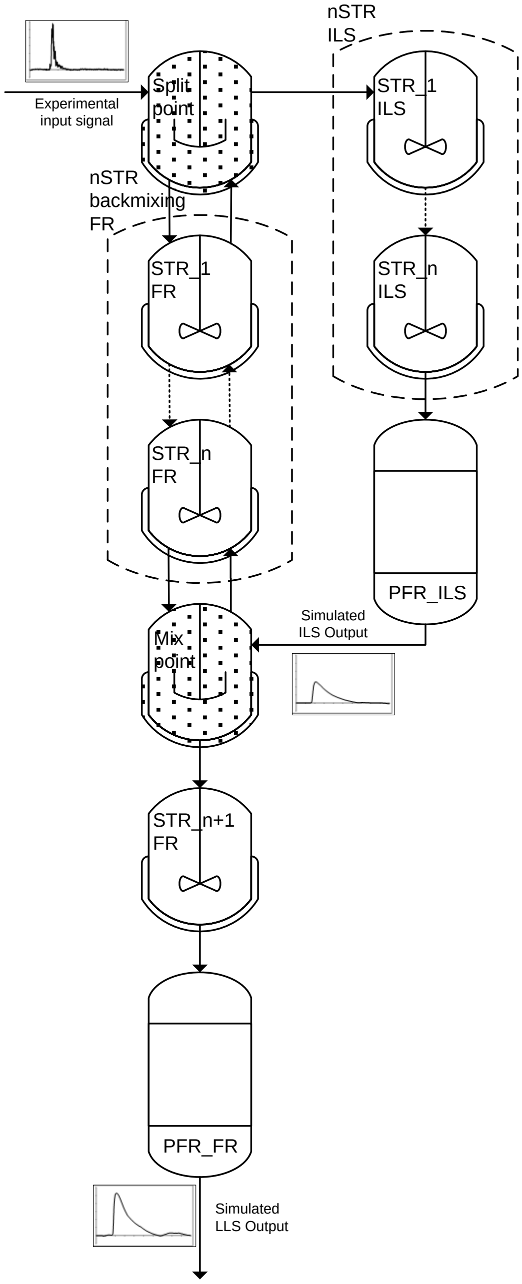


Figure 4: Proposed RTD model.

(b) The fuel reactor itself, namely its countercurrent section, it is, the section below the upper loop seal. In this section particles flow both up- and down-wards but the net flow is downwards to the lower loop seal.

(c) The lowest section in the FR is modeled separately because of two reasons: it sums up material from both global and internal loops and contains a bed denser than in the rest of the reactor.

The assumptions made with this model include:

- There is no backflow of material in the internal loop. This backflow can certainly occur at two points, the upper part of the FR since many of the particles reaching this section fall again in the bed instead flowing directly to the cyclone; here the low density of the bed allows assuming that no backmixing would take place. The second point is the return leg of the ILS, if the pressure difference in the seal is not appropriate, particles can flow from the reactor into the seal; this effect is however unlikely when the internal circulation is stable and high enough.
- In the FR, since bed density deceases with the height of the reactor, the relation between the streams going up- and down-wards from each stirred tank might vary as well. This split condition is considered constant for simplification.
- The n stirred tanks in the body of the FR are considered of same size. The stirred tank at the bottom has, however, its own and different size. The relation between these sizes is calculated from the relative pressure differences of the two sections.
- The n tanks in the internal loops are of same size as well (but different from the ones in the FR).

Figure 5 shows an example of the results obtained with the proposed model, measured signals are shown as well and parameters for the simulation appear in Table 2. Both output signals are successfully measured, processed and simulated; for each of them the corresponding **E** curve is calculated with a satisfactory agreement. The impact of the internal circulation on the global one can be successfully simulated with the proposed model. The signal from the internal loop seal shows an unexpected width

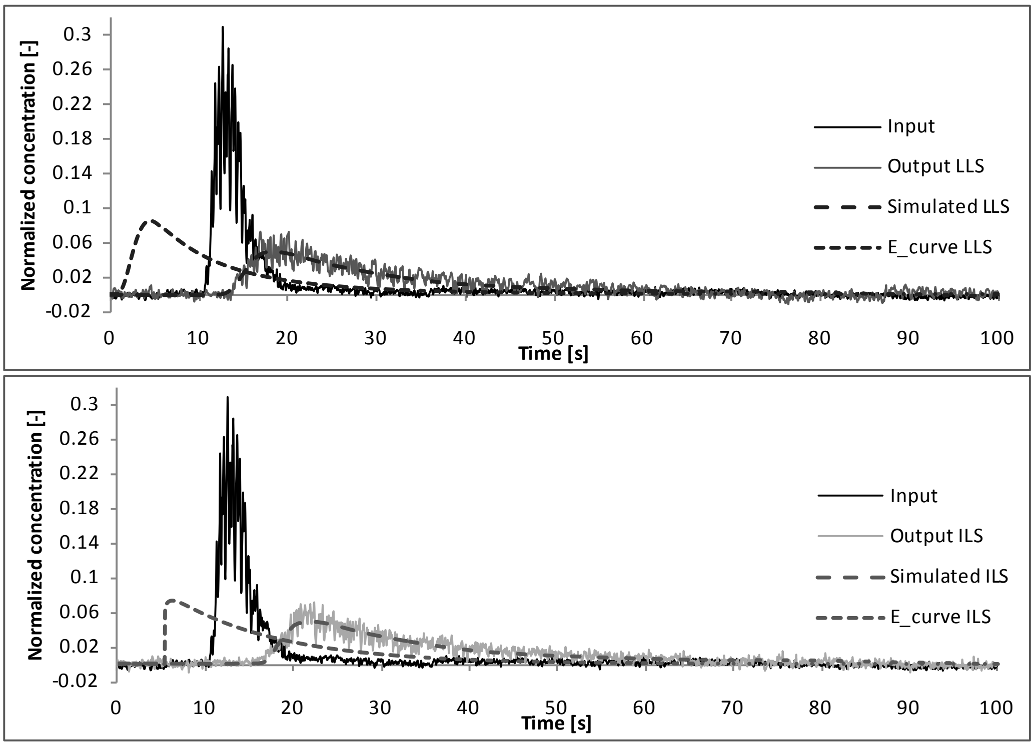


Figure 4: Example of measured and simulated signals of a typical RTD test. LLS signal up and ILS signal down. Conditions: 2.83m·s^{-1} AR, 1.82 m·s^{-1} FR, 1 Nm^3·h^{-1} ULS, 1 Nm^3·h^{-1} LLS, 0.7 Nm^3·h^{-1} ILS, total inventory 5kg, circulation rates in terms of FR: 68.73 kg·m^{-2}·s^{-1} global and 27.10 kg·m^{-2}·s^{-1} internal.

Table 2: Simulation parameters for the example presented in Figure 4.

Parameter		Internal loop	Global loop	
τ	Experimental	18.98	15.20	[s]
n	Number of tanks STRn model	1	6	[-]
τ_{PFR}	Plug flow model	5.35	0.11	[s]
τ_{STR}	Stirred tanks in series model	12.35	7.97	[s]
τ	Simulation	20.15	16.56	[s]
τ	E curve	17.81	12.66	[s]
σ^2	var^2 E curve	168.65	130.97	[-]
	Square error (sim / exp)	0.0558	0.0796	[-]
Ratio global/internal+global circ.		0.80		[-]

(high τ is calculated); rather than indicating the mean time that the particles need to flow one-way from ULS to the ILS, it represents the residence time of the particles circulating in the internal loop including the residence time of these particles inside the FR riser. This effect is reflected as well in the τ for the global loop. The result for the ratio of global circulation to global plus internal circulation (actually corresponding to the split condition in the FR model), keeps a coherence with the expectations, a tentative ratio calculated based on measured circulation rates differs only slightly (0.72 for measured circulation rates versus 0.80 derived from the RTD measurements).

CONCLUSIONS

A method for determination of solids residence time distribution has been successfully developed. Precise measurement of ferromagnetic material concentration has been performed based on inductance changes of simple coils. The system allows for accurate capture of RTD in the cold flow model presented. The results obtained experimentally are realistic, reproducible, accurate, and suitable for modeling calculations. An RTD model has been properly fit to the experimental data. The inductance measurements in conjunction with cold flow models are seen as a promising technique for the study of fluid-dynamics of fluidized beds and especially valuable for fluid dynamic model validation and scale-up of fluidized bed systems. The characteristics of the method reflect the possibility for its implementation not only in RTD determination but also in measurement of circulation rate and bed density in similar units.

NOTATION

A	Cross-section area of the conductor, [m^2]
AR	Air reactor
Ar	Archimedes Number, [-]
B	Magnetic flux density, [T]
C_{in}	Concentration function of input signal
C_{out}	Concentration function of output signal
CFB	Circulating fluidized bed
d_P	Mean particle diameter, [m]
D*/*uL	(Eq. 9)Vessel dispersion number [-] (10)
DCFB	Dual circulating fluidized bed
E	Residence time distribution function
FR	Fuel reactor
H	Strength of the generated magnetic field, [$A \cdot m^{-1}$]
I	Current, [A]
l	Length of the coil, [m]
ILS	Internal loop seal
L	(Self) inductance, [H]
LLS	Lower loop seal
N	Number of turns in the coil
n	Number of tanks
Re_{mf}	Particle Reynolds Number at minimum fluidization velocity, [-]
RTD	Residence time distribution
t	*T*ime, [s]
$\bar{t}_i$	Mean residence time for the tank *i,* [s]
U	Superficial gas velocity, [$m \cdot s^{-1}$]
U_{mf}	Minimum fluidization velocity, [$m \cdot s^{-1}$]
ULS	Upper loop seal
σ^2	Variance of the residence distribution curve
δ	*Dirac delta function*
Φ	Mean particle sphericity, [-]
ρ_P	Particles density, [$kg \cdot m^{-3}$]
τ	Mean residence time, [s]
τ_{PFR}	Mean residence time - plug flow reactor model, [s]
τ_{STR}	Mean residence time - stirred tank reactor model, [s]
μ	Permeability, [$H \cdot m^{-1}$]
μ_0	Permeability of vacuum, [$H \cdot m^{-1}$]
μ_r	Relative permeability, [-]

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