THE INVESTIGATION OF FLUIDIZATION OF SOLIDS MIXTURE WITH DIFFERENT PARTICLE DENSITY

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Abstract – High temperature chemical looping combustion is one of the promising technologies for CO_2 capture The CLC reactor system consists of two principal reaction chambers: the air reactor (AR) and the fuel reactor (FR). High solid circulation rate between fuel and air reactors is very important for effective combustion of solid fuels in CLC. Some part of fuel ash particles is implemented in metal oxides flow. Thus, the circulation flux consists of heavy metal particles and light ash particles. Fluidization conditions of binary mixture of particles with different density and sizes are very important for CLC hydrodynamics.

The investigation was made on cold model with interconnected reactors. The narrow fraction of Al2O3 with Sauter diameter 0.22 and 0.347 mm and density 3930 kg/m3 were used as bed materials. Silica sand with Sauter diameter 0.21 mm and density 2600 kg/m3 was a "coal ash".The pressure gradients and minimal fluidization velocities were determined for each material. Experimental data have a good agreement with calculation by Ergun formula. The investigations were made for mixture with volume fraction 50-95% of Al2O3.

INTRODUCTION

Currently, according to A. Lyngfelt et al. (2008), there are more than 600 works on various aspects of use the chemical loops. The experiments revealed that almost complete fuel conversion is possible at 100% capture of CO_2. A lot of works focused on the study of firing the gas fuel. There are only a few test rigs and pilot plants using solid fuel P. Markström at al. (2012). World operating experience of firing fuels in chemical loops involving oxygen carriers with base oxides on Ni, Fe, Mn, Cu and Co, has over 4000 hours. In recent years, considerable interest is growing in use the low cost natural minerals - ilmenite and ferromanganese minerals. This primarily applies to firing of solid fuels Juan Adánez (2012). Reliable high circulation ratio of the material between the reactors and maintaining the desired process temperature is critical for this technology. During the combustion and gasification of solid fuels the ash particles is inevitably added to the circulating metal oxides. This results in a problem of fluidization of binary mixture of solid particles with significantly different densities. In these circumstances, it is important to determine the hydrodynamic parameters and minimal fluidization velocities of binary mixtures, to examine the circulation modes in a system with interconnected reactors.

There are a lot of investigations in field of fluidization of binary mixture of particles. In Rowe and Nienow (1975) general equation was proposed for predicting the minimum fluidization velocity of a mixture of particles of various sizes but all of the same shape and density. It requires knowledge of the change in voidage that occurs on changing the mixture composition. It predicts change in the minimum fluidization velocity with small changes in the fines content of, for example, a commercial catalyst. Investigations of Isemin at al. (2007) were made with anracite particles (0 – 5 mm fraction) and pellets 12.5 mm. Prediction of minimum fluidization velocity of binary systems was done by Chiba et al. (1979) and Obata et al. (1982). More complete information and equations was done by Rincon et al. (1994). In investigations of Rincon et al. (1994) glass particles (mean diameter 2.9 -0.46 mm, density 2700 kg/m^3) and plastic granules particles (mean diameter 2.9 mm, density 2900 kg/m^3) were used. Narrow fraction and near the same density are used in mostly investigations. Coal based CLC technology characterized narrow fraction of high density particles (metal oxides) and wide fraction of relatively small density particles (ash).

EXPERIMENTAL SETUP AND CHARACTERISTICS OF APPLIED MATERIALS

To study the hydrodynamics of interconnected CFB and FB reactors the experimental setup was constructed. The main elements are CFB and fluidized bed reactor (FB) reactors with associated overflow system. A detailed description of the installation and layout of reactors is given in Ryabov et al. (2014). CFB reactor is a vertical column with cross-section 0.2 × 0.3 m and 5.4 m height, to the top of column the inlet cyclone duct is attached. The air is discharged from the cyclone to the settling chamber, at the top of which installed the removable filter. To the conical part of cyclone attached the riser with cross-section 0.1 × 0.1 m. In the middle part of the riser is installed shutoff rotary valve, which is used to determine the flow rate of material through the circulation loop. The riser is connected to the upper loop seal. The design of the loop seal allows to release one part of the material directly to the CFB reactor, and the other part to the lower part of FB reactor through the riser with L-valve with 44 × 94 mm cross-section and 420 mm length in horizontal part. FB reactor has a lower section with 0.28 × 0.2 m cross-section and a height of 0.5 m, a transition cone section and an upper section of 0.4 × 0.4 m cross-section and 1.5 m height. It is connected to pipe with loop seal placed in the conical part of reactor and providing feed of the material to lower section of CFB reactor.

An important issue is to simulate the hydrodynamic conditions using mixtures of metal oxides and particles of ash. The most significant is the choice of size of these particles. Therefore, it was necessary to analyze the published data on the use of oxides during the combustion of solid fuels in firing installations. In Bolh à ret al. (2008) the South African coal burned with an average particle size of 0.17 mm. Information about the size of petroleum coke burned in N. Berguerand and A. Lyngfelt (2008) is not given, but in these two studies as a metal oxide, ilmenite was used with a particle size of 0.09 - 0.25 mm at a bulk density of 2100 kg/m^3. In L. Shen et al (2009, a, b, c) [8, 9, 10] were used metal oxides on the nickel and aluminum basis with dimensions of 0.2 - 0.4 mm and a bulk density of 2350 kg/m^3. The average size of carbon particles was equal to 0.38 mm, and the biomass particles - 1.5 mm. Pulverized coal with 0.123 mm particle size and coal with big particles (up to 8 mm) were combusted in 1 MW capacity plant (P. Ohlemüller et al. 2014).

It should be noticed that during the coal combustion in CFB the average circulating ash particles size is in a narrow range of 0.15 - 0.25 mm. In many cold models are used the particles of sand with the same dimensions as the particles of the circulating material. Thus, we can conclude that for simulation of ash behavior in cold model is likely to use the sand with an average size of about 0.2 mm. Oxides particles such as aluminum or iron oxides should have dimensions close to sand particle size.

The characteristics of materials used in experiments are shown in Table 1.

Table 1 – The characteristics of studied materials.

№	Characteristics/material	Sand	Al2O3 (Triakor D)	Al2O3 (Beleit)
1	real particles density, ρ, kg/m^3	2600	3930	3960
2	Bulk density, ρ_b kg/m^3	1550	1900	1970
3	Vibration bed (tapped) density, ρ_v кг/м^3	1650	2080	2120
4	Average diameter on weight, d_m, mm	0.250	0.240	0.380
5	Average diameter on surface, $d_{s,}$ мм	0.212	0.204	0.347
6	Settlement average diameter, d_p, mm (with shape factor)	0.215	0.220	0.350

Methods of determining the parameters of fluidization are described in [12]. Minimal fluidization was determined visually and by the pressure gradient change curve. Initially the fluidization parameters were separately determined for two fractions of sand and aluminum oxides. There were the traditional curves of changes in pressure and level swelling. Then, 5 - 10% of each aluminum oxide fraction was removed and sand was fed to the same bed level. Sand and metal oxide then mixing with large amount of air and after that the experiment starts with certain sand fraction. In the next experiment, the mixture was removed again and fills up with sand. As a result, the range of volume fractions of sand changed from 95% to 50% in the volume of aluminum oxide (mass fraction – 96 – 55%).

RESULTS AND ANALYSIS

Table 2 shows the experimental data of minimal fluidization velocity, maximum pressure gradients and pressure gradients in minimal fluidization regime of sand and two types of aluminum oxide. There's also shows the calculated values determined by Todes formula (M. E. Aerov and O. M. Todes, 1968) [13], simplified formula J. H. Goo et al. (2009) [14] and Ergun formula according to measured pressure gradient.
The experimental and calculated data for minimum fluidization velocity for sand is in good agreement with each other. Velocities at the beginning of fluidization of aluminum oxides were higher than calculated by dependencies M. E. Aerov and O. M. Todes, 1968 and J. H. Goo et al. (2009) which may be due to the difference between the actual shape factor and voidage at minimal fluidization from values in calculated dependencies. The data on calculated velocity by measuring pressure gradients satisfactorily agree with measured values.

Table 2 - The experimental data for start of fluidization of sand and aluminum oxides.

№	Parameter	Dimension	Sand	Al2O3 (Triakor D)	Al2O3 (Beleit)
1	Experimental pressure gradient at the minimum fluidization.	kPa/m	9.2	13.8	17.2
2	Experimental maximum pressure gradient	kPa/m	11	15	19
3	Experimental minimum fluidization velocity	m/s	0.042	0.095	0.23
4	Calculated minimum fluidization velocity by M. E. Aerov and O. M. Todes, 1968 [13]	m/s	0.04	0.07	0.15
5	Calculated minimum fluidization velocity by J. H. Goo et al. 2009 [14]	m/s	0.04- 0.045	0.06 – 0.07	0.17-0.19
6	Minimum fluidization velocity calculated according to Ergun formula and experimental pressure gradient	m/s	0.04–0.045	0.07-0.09	0.19-0.23

Figure 1 shows the pressure gradient at the minimal fluidization against the volume fraction of Al2O3 (0.35 mm). The pressure gradient at the minimal fluidization decreases linearly with increasing volume fraction of sand. When increasing the share of sand the minimal fluidization velocity decreases not linearly (Fig. 2).

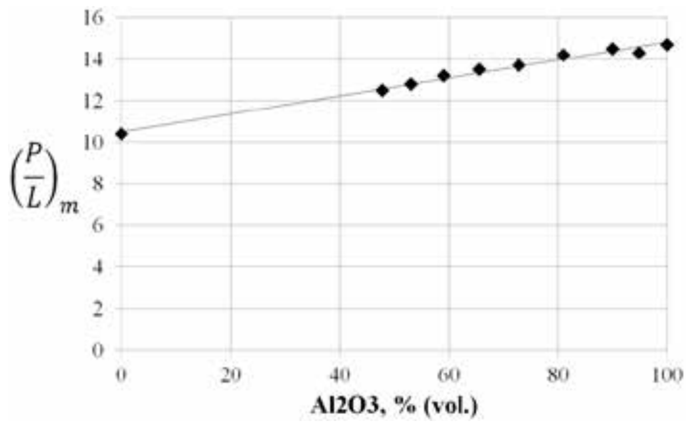

Fig. 1. The pressure gradient at the minimal fluidization against the volume fraction of Al_2O_3.

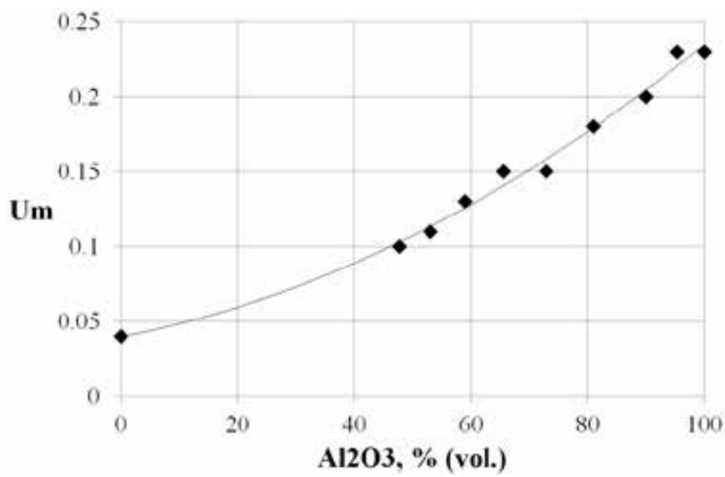

Fig. 2. Minimal fluidization velocity against the volume fraction of AL_2O_3.

In Rincon (1994) presented the dependence for calculating the minimal fluidization velocity of particle mixtures based on geometrical constructions according to the relative changes in pressure to the particles and the mixture of particles. For a mixture of aluminum oxide particles and sand it looks like:

$$\frac{1}{U_m} = \frac{X_s}{U_s} + \frac{X_{Al}}{U_{Al}} \tag{1}$$

It seems that this dependence is quite suitable for spherical particles with similar voidage. The slope of the pressure gradient line is defined by Ergun formula and depends on diameter of particles and their voidage. Our experimental data gave similar values of angle for binary mixtures, the largest slope characteristic of the sand, and the least - for the large fraction of alumina.

The following model is proposed to calculate the fluidization parameters for binary mixtures of particles with different densities and sizes:

- Determination of equivalent diameter of the particle mixture based on Kruger formula:

$$\frac{100}{d_m} = \sum \frac{q_i}{d_i} \tag{2}$$

- The pressure gradient at the minimal fluidization is determined by the first term of Ergun formula:

$$\left(\frac{\Delta P}{L}\right)_{mf} = \frac{150\mu}{d_p^2} U_{mf} \frac{(1-\varepsilon_{mf})^2}{\varepsilon_{mf}^3} \tag{3}$$

- At a constant bed volume its density at minimal fluidization can be written as:

$$\rho_m\left[1-(\varepsilon_{mf})_m\right] = q_s\left[1-(\varepsilon_{mf})_s\right]\rho_s + q_{Al}\left[1-(\varepsilon_{mf})_{Al}\right]\rho_{Al} \tag{4}$$

- The equivalent density of mixture varies linearly with changing the volume share of metal oxide

$$\rho_m = \rho_s + q_{Al}\left[\rho_{Al} - \rho_s\right] \tag{5}$$

Based on these premises the voidage of particle mixture at minimal fluidization is

$$(1-\varepsilon_{mf})_m = \frac{q_{Al}\left[\left(1-\varepsilon_{mf}\right)_{Al} - (1-\varepsilon_{mf})_s \frac{\rho_s}{\rho_{Al}}\right] + (1-\varepsilon_{mf})_s \frac{\rho_s}{\rho_{Al}}}{\rho_s + q_{Al}\left[\rho_{Al} - \rho_s\right]} \tag{6}$$

To analyze the experimental data they should be presented as a function of the relative pressure gradient and relative minimal fluidization velocity with respect to these values for the metal oxide. In accordance with eq. (2) – (6) was obtained dependence for the relative pressure gradient on the relative minimal fluidization velocity:

$$\left(\frac{\Delta \overline{P}}{L}\right)_{mf} = \overline{U}_{mf}\left(\frac{d_{Al}}{d_m}\right)^2 \frac{(1-\varepsilon_{mf})^2}{(\varepsilon_{mf})_m^3} \frac{(\varepsilon_{mf})_{Al}^3}{(1-\varepsilon_{mf})_{Al}^2} \tag{7}$$

Figure 3 shows the experimental data on the relative pressure gradient against the volume fraction of Al_2O_3. For the Al_2O_3 with small particles mixed with sand, this dependence is close to linear. For Al_2O_3 with large particle small addition of sand leads to a significant reduction in the relative pressure gradient at minimal fluidization.

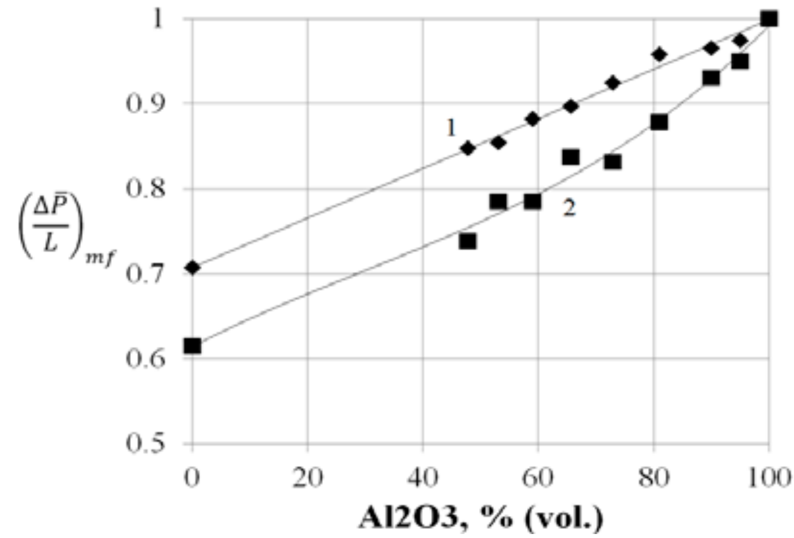

Fig. 3. Relative pressure gradient at minimal fluidization against the volume fraction of Al_2O3.

Figures 4 and 5 shows the relative minimal fluidization velocity between against the volume fraction $Al2O_3$ (small and large particle sizes) mixed with sand. Also, there are presented calculated by the eq. (1) and (7) relative values of minimal fluidization velocity.

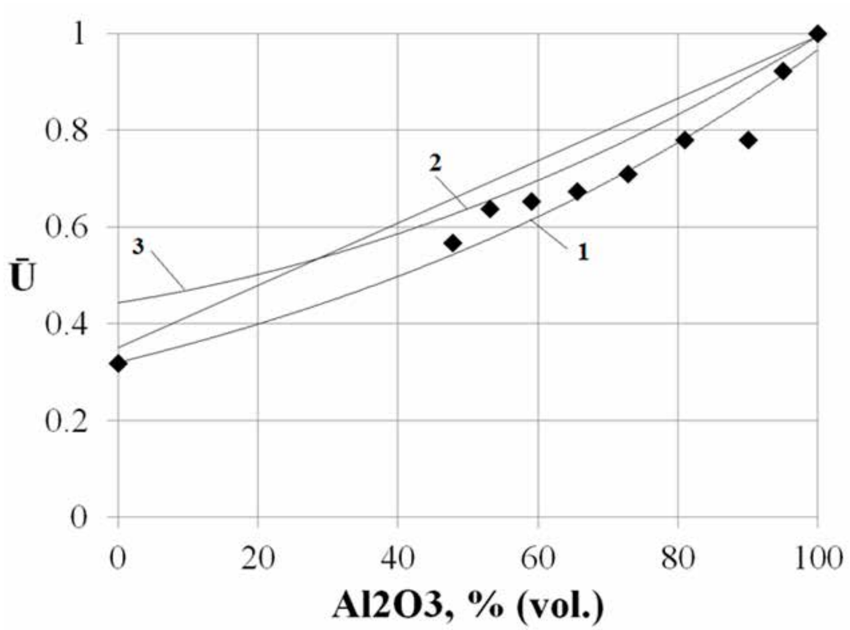

Fig. 4. The relative minimal fluidization velocity against the volume fraction of small fraction Al_2O_3 in a mixture with sand. 1 - experimental data, 2 – according to eq. (7), 3 - according to eq. (1).

The real values of density of sand and Al_2O_3 mixtures were also identified by pycnometric method. It was found that a linear relationship is broken only for the volume of sand fraction of less than 10%. As a result, significant changes in calculated minimal fluidization velocity has not occurred (curve 4 in Fig. 5).

The calculated values for the smaller fraction of Al_2O_3 provide increased minimal fluidization velocity in comparison with experimental data. For a larger fraction the estimated values are below the experienced. The reason for that is a simplified approach for definition of equivalent diameter and equivalent real density of the particles. However, with increased volume fraction of Al_2O_3, which occurs in practice in high-temperature chemical looping cycles with metal oxides (oxygen carriers) - the use of estimated dependency yields a relatively small error in determining the fluidization parameters.

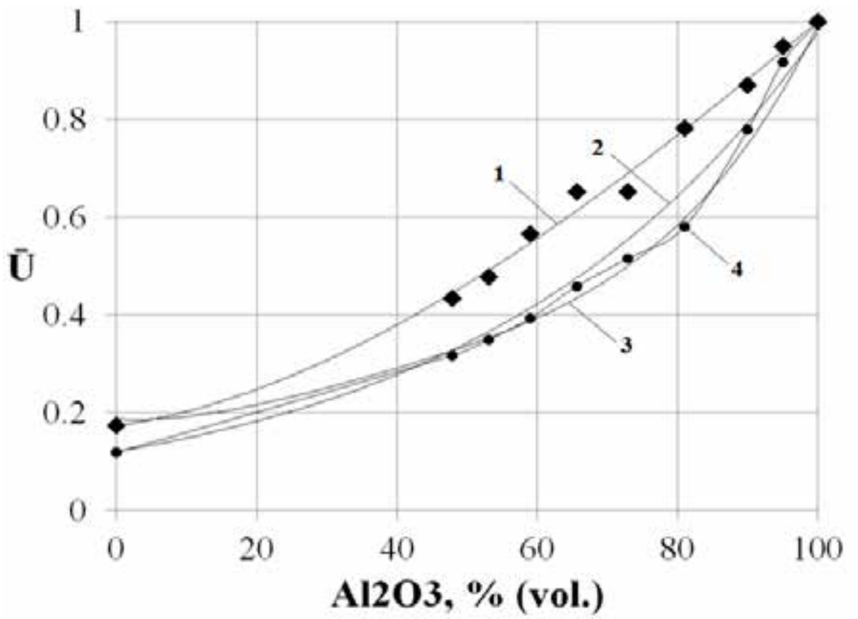

Fig. 5. The relative minimal fluidization velocity against the volume fraction of large fraction Al_2O_3 in a mixture of sand for a large fraction of Al_2O_3. 1 - experimental data, 2 – according to eq. (7), 3 – according to eq. (1), 4 - according to eq. (7) with experimental value of real density of mixture.

It should be noted,when using alumina and sand with approximately the same particle dimensions the mixing along the height of the riser is quite good, however, when using alumina with larger particle sizes than sand has been observed some sand segregation. Sand mostly was closer to the top of the bed (Fig. 6). These, apparently explains the fact nonlinear reducing the minimal fluidization velocity with small additions of sand.

Further investigations have aim to determine of light fraction addition on hydrodynamics of interconnected reactors, standpipe and loop seal. The researches of hydrodynamics in the interconnected reactors system with binary mixtures of particles is currently conducting. The field of mass flow and the share of Al2O3 near to the wall of CFB reactor were determined with the S-shaped probe This paper is supported by Russian Fund of Fundamental Research (Project NO: 16-08-00294/16).

Fig. 6 – Picture of bed (90% AL_2O_3 of 0.35 mm fraction, 10% sand).

CONCLUSIONS

By using the advanced combustion technology and gasification of fuels in chemical loops with CO_2 capturing arises the problem with motion of binary mixture of solid particles with a significantly different density. Considering that the questions hydrodynamics of particles in the conditions of use the circulation circuits with interconnected CFB and FB reactors has not been studied so far in the world practice, the relevance of proposed researches is very high.

The results of fluidization condition study for binary mixtures of metal oxides and sand showed quite strongly decreasing of minimal fluidization velocity at low initial addition of sand.

Dependency of relative pressure gradient against the volume fraction of heavy particles is linear. It is noted that when using alumina with larger particle sizes than sand particles has been observed some segregation, sand for the most part is closer to the top of the bed. Dependences for calculating the fluidization parameters of binary mixtures were proposed. With increased volume fraction of Al_2O_3, which occurs in practice in high-temperature chemical looping cycles with metal oxides (oxygen carriers) - the use of estimated dependency yields a relatively small error in determining the fluidization parameters.

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