The investigation of movement conditions of particles binary mixtures in chemical looping combustion of solid fuels

Ryabov G\textsuperscript{1} Folomeev O\textsuperscript{2} Dolgushin I\textsuperscript{3}

1 Head of Laboratory, JSC VTI, Moscow, RF
2 Senior scientist, JSC VTI, Moscow, RF
3 Senior scientist, JSC VTI, Moscow, RF
E-mail; vti@vti.ru

Abstract. High temperature chemical looping combustion is one of the promising technologies for CO\textsubscript{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 Al\textsubscript{2}O\textsubscript{3} with Sauter diameter 0.22 and 0.34 mm and density 3930 kg/m\textsuperscript{3} were used as bed materials. Silica sand with Sauter diameter 0.21 mm and density 2600 kg/m\textsuperscript{3} was a “coal ash”. The investigations were made for mixture with volume fraction 50-95\% of Al\textsubscript{2}O\textsubscript{3}. Small addition of sand gives a significant reduction of minimal fluidization velocity. There is a linear proportion between share of Al\textsubscript{2}O\textsubscript{3} and pressure gradient. Some other phenomena were obtained in case of 0.34 mm Al\textsubscript{2}O\textsubscript{3}. Further investigations have aim to determine of light fraction addition on hydrodynamics of interconnected reactors.

1. Introduction

Currently, according to A. Lyngfelt et al. [1], 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\textsubscript{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. [2]. 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 [3]. 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. Coal based CLC technology characterized narrow fraction of high density particles (metal oxides) and wide fraction of relatively small density particles (ash). It should be noticed that during the coal combustion in CFB the average circulating ash particles size is in 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.

2. Experimental setup and methods

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. [4]. 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 $0.1 \times 0.1$ 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 $44 \times 94$ mm cross-section and 420 mm length in horizontal part. FB reactor has a lower section with $0.28 \times 0.2$ m cross-section and a height of 0.5 m, a transition cone section and an upper section of $0.4 \times 0.4$ 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.

Methods of determining the parameters of fluidization are described in [5]. 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 $\sim 96 - 55\%$). The field of mass flow and the share of $\text{Al}_2\text{O}_3$ near to the wall of CFB reactor were determined with the S-shaped probe (fig. 1).

![Schematic diagram for measuring of local mass flows](image)

**Fig.1** Schematic diagram for measuring of local mass flows

**3. Results and analysis**

Complete experimental data on minimal fluidization velocity, maximum pressure gradients and pressure gradients in minimal fluidization regime of sand and two types of aluminum oxide were done in [6]. 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. 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 [6] was obtained dependence for the relative pressure gradient on the relative minimal fluidization velocity:
(1)

\[
\left( \frac{\Delta P}{L} \right)_{mf} = \overline{U}_{mf} \left( \frac{d_{Al}}{d_m} \right)^2 \left( \frac{1}{1 - \varepsilon_{mf}} \right)^2 \left( \frac{\varepsilon_{mf}}{\varepsilon_{mf}} \right)_{mf} \left( \frac{1}{1 - \varepsilon_{mf}} \right)_Al
\]

Where: \(d_{Al}\) and \(d_m\) - settlement average diameter of \(\text{Al}_2\text{O}_3\) and mixture, mm, \((\varepsilon_{mf})_{Al}\) and \((\varepsilon_{mf})_m\) - minimal fluidization porosity of \(\text{Al}_2\text{O}_3\) and mixture

Figure 2 shows the experimental data on the relative pressure gradient against the volume fraction of \(\text{Al}_2\text{O}_3\). For a \(\text{Al}_2\text{O}_3\) with small particles mixed with sand, this dependence is close to linear. For \(\text{Al}_2\text{O}_3\) with large particle small addition of sand leads to a significant reduction in the relative pressure gradient at minimal fluidization. Figures 3 shows the relative minimal fluidization velocity between against the volume fraction \(\text{Al}_2\text{O}_3\) (small particle sizes) mixed with sand. Also, there are presented calculated by the eq. (1) relative values of minimal fluidization velocity.

With increased volume fraction of \(\text{Al}_2\text{O}_3\), 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. 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. 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, first of all - CFB reactor. The experiments were made with pure \(\text{Al}_2\text{O}_3\) with Sauter diameter 0.236 mm, real density 3940 kg/m3, bulk density 1970 kg/m3. The recirculation rate in the standpipe, pressure drops and average concentration on the height of CFB reactor were obtained as well as profile of up and down mass fluxes in upper part of the reactor (near cyclone inlet). Then, about 1.5 kg of sand (Sauter diameter 0.22 mm, real density 2600 kg/m3, bulk density 1560kg/m3) was fed in CFB reactor. It was made 3 time, so the past experiment was made by feeding 4.5 kg of sand. There are 2 average air velocity 3.6 and 4.3 m/s and near the same mass in the reactor (40 – 45 kg).

It is very important to determine an initial mass inventory in the system and a part of light fraction (ash or sand). The specific solid inventory in the fuel reactor (FB) varied from 360 -750 kg/MWth [7]. As air reactor is CFB reactor, which operated in range of 2 – 3 time large velocity than
FB reactor, solid inventory in CFB reactor is about 150 - 250 kg/MWth. The largest experimental unit 1MWth [8] has CFB reactor cross section 0.273 m² and operated with coal rate 150 kg/h. So, mass inventory in CFB reactor is about 200 kg and bed height – 0.35 m. For our experimental test rig with cross section 0.06 m² the equivalent mass is about 40 – 50 kg. There are near 20 kg/h of ash are added in 1 MWth system. Most of ashes are removed from the system by drain and flue ash. The gross estimation gives about 3 – 12% of ash in total mass inventory of coal based CLC systems.

Typical up and down mass fluxes around the width of the installation are shown on fig. 4. Maximum up flow mass fluxes are in the core and maximum down flow mass near the wall. This profile characterized flow behavior in CFB reactor [9]. The share of light fraction (sand) gives a little increasing of upward local mass fluxes near the wall (0 – 20 mm).

![Fig. 4. Profile of local up and down mass fluxes. Velocity – 3.6 m/s](image)

1. 2 up and down, no sand, 3.4 – 3.5% of sand, 5.6 – 7% of sand

The profiles of summary (as sum of up and down mass flux) local mass flux are done on fig. 5 and 6. There is no influence of sand share on summary local mass flux. The width of down flow zone near the wall is about 10 mm.

![Fig. 5 – Profile of summary mass fluxes. Velocity – 3.6 m/s](image)

1- no sand, 2:3.5% of sand, 3 – 7% of sand, 4 – 11% of sand
Fig. 6 – Profile of summary mass fluxes. Velocity – 4.3 m/s
1- no sand, 2 - 3.5\% of sand, 3 – 7\% of sand, 4 – 11 \% of sand

Results of the investigations of CFB hydrodynamics demonstrated a slow influence of light fraction addition on circulation rate and local mass flux in range of light fraction share 3 – 12 \%. This fact means that calculation of circulation rate and mass fluxes could be carried out by previous dependences for pure metal oxides.

Acknowledgments
This paper is supported by Russian Fund of Fundamental Research (Project NO: 16-08-00294/17).

References
[1] Lyngfelt A et al 2008. Chemical looping combustion – Status of development, Proc. of CFB 9, May 13-16 2008, Hamburg, Germany, pp 39-53
[2] Markström P et al 2012. Chemical Looping Combustion in a 100 KW Unit for Solid Fuels, Proc. of Int. Conf. FBC 21 June 4-6, 2012, Naples, Italy, pp. 285-292.
[3] Adánez J Chemical-Looping Combustion of Coal: Recent Developments and Technology Challenges, plenary lecture, Proc. of Int. Conf. FBC 21 June 4-6, 2012, Naples, Italy, pp. 12-27.
[4] Ryabov G et al 2014. Pressure Balance model for Dual CFB-FB Reactors Systems, Proc. of the 22nd Int. Conf on Fluidized Bed Conversion, 12-14 June, Turky, Finland.
[5] Ryabov G and Folomeev O 2014. The Methods for Calculation of Fluidization Parameters for Solid Return Systems of Circulation Fluidized Bed Reactors, Knowledge of Russian Academy of science, Energetics, # 5, 2014 (in Russian).
[6] Ryabov G et al 2017 The Investigation of fluidization of solids mixture with different particles density, Proc. of CFB12, May 24-26, Krakow, Poland, pp 179 – 186.
[7] Mattisson T. et al 2017 Chemial-looping technologies using circulating fluidized bed systems: status of development, Proc. of CFB12, May 24-26, Krakow, Poland, pp 23 – 34.
[8] Ohlemüller P et al 2015. Operation of a 1 MWth Chemical Looping Plant Proc. of the 22nd Int. Conf on Fluidized Bed Conversion, 12-14 June, 2015. – Finland, Turku, 2015.
[9] Ryabov G et al 2002. Solid mass fluxes in the exit region of CFB furnace// Proc. of 7th Int. Conf. on CFB, Niagara Falls, Ontario, Canada, 2002.