

Article

Effect of Fines Content on Fluidity of FCC Catalysts for Stable Operation of Fluid Catalytic Cracking Unit

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Abstract: Effect of fines content (weight % of particles with diameter less than 45 μm) on bed fluidity was determined to get a base for good fluidization quality in the fluid catalytic cracking (FCC) unit. The fines content in equilibrium FCC catalysts (Ecat) from commercial units were controlled by adding or removing the fines to simulate commercial situation. To get the fluidity values ($U_{\text{mb}}/U_{\text{mf}}$) of seven different FCC catalysts (2 Ecats and 5 fresh catalysts) and their mixture, minimum fluidization velocity (U_{mf}) and minimum bubbling velocity (U_{mb}) were measured in a fluidized bed reactor (0.05 m ID). The fluidity decreased with loss of fines content and increased with increments of makeup of fresh catalysts or additive with the controlled fines content. The fluidities of catalysts increase with increases of normalized particle diameter variation by the fines addition. The obtained fluidities have been correlated with the fines contents and the catalyst and gas properties. The proposed correlation could guide to keep good catalyst fluidity in the FCC unit.

Keywords: fluid catalytic cracking; catalysts; fluidization; fluidity; fines content

1. Introduction

Fluid catalytic cracking (FCC) processes are used to upgrade heavy hydrocarbon fractions into valuable light fractions, such as propylene, gasoline, and gas oil, etc. [1]. The FCC unit consists mainly of a riser, stripper, and catalyst regenerator. The FCC process utilizes a catalyst circulation system, whereby a hot circulating catalyst contacts the oil feedstock in the riser where the catalytic cracking reaction occurs. The cracked product vapor from the reactor is sent to a main fractionator for product separation. The catalyst after the reaction is stripped of volatiles with steam in the stripper, then is sent to the regenerator. The catalyst is reactivated and heated by burning off the coke with air in the regenerator. The heated catalyst is transported to the riser to meet again with the fresh feedstock. The catalyst circulation loop is completed by re-injecting the regenerated catalyst into the riser [2,3].

It is essential to define fluidization phenomena of catalysts in the FCC unit, since all catalyst beds in the circulation loop are in fluidized beds at all times.

The fluidity or fluidization index is defined in many ways and there have been as yet no unique definition and correlation to be fully accepted [4]. Nevertheless, it has long been known in the industry that fluidity ($U_{\text{mb}}/U_{\text{mf}}$) is a useful criterion for judging the hydrodynamic behavior and fluidization properties of the FCC catalysts in fluidized beds [5–7]. To get the fluidity value, many experimental studies have provided correlations between operating parameters and minimum fluidization velocity (U_{mf}), while less work has been directed at minimum bubbling velocity (U_{mb}) [8]. In the experimental measurement of U_{mf} and U_{mb} , the U_{mb} has ambiguity in judgment of the first bubble, unlike U_{mf} . Recently, computational fluid dynamics (CFD) simulation determined U_{mb} point of the bubble can

be well defined [9,10]. However, experimental measurements are often recommended in the refinery, because difficulties may arise with fluidization characteristics of the catalysts changing over time [2,8].

Many studies have tried to determine the fines' on the fluidization quality of the catalyst beds [7]. Direct measurement of the FCC catalyst fluidity could provide refiners a tool to routinely check for the catalyst circulation problems in the FCC unit [11]. The control of catalyst fluidity is often difficult due to changing operation conditions and the limited range of available treatment options. The operational condition, hardware design, and catalyst characteristics causes the emission of catalysts, especially catalyst fines. The loss of fines in catalyst inventory can lead to a reduced catalyst fluidity, catalyst defluidization in FCC circulation loop, or worse, shut-down of the unit. In the case of poor fluidization trouble caused by the fines content loss, it is usually recommended to quickly shift the catalysts size distribution (PSD) to the normal distribution of the equilibrium catalyst in the unit. Refiners purposely add fresh catalyst with high fines content, because the catalyst fines cannot be added independently for fast effect in the fluidity. The catalyst vendor can change the particle size distribution of the fresh catalyst, such as adding fines and re-design of the catalyst to assist in restoring normal PSD of the bed. Finally, the refiners take the action of adding the fresh catalysts or fluidity-enhancing-additives with high fines content articles into the regenerator [11,12]. However, the refiners often face the challenging task of judiciously selecting catalysts and additives and estimating the amount of catalyst required for fast effect in the fluidity. It is very difficult to select a catalyst, either by the catalyst vendor's claims or through a performance test directly at the plant [13]. Therefore, it is important to apply reliable prediction for the selection and to exploit the selected catalyst capabilities to the full extent based on the prediction. A few correlations [5,6,12] to predict the fluidity have been reported relating to the fluidity prediction, because less correlation work has been directed at U_{mb} compared to U_{mf} [8]. Abrahamsen and Geldart's [3] correlation for prediction of fluidity has been widely used in the refinery [2]. However, Whitcombe et al. [8] showed that previous correlations on fluidization characteristics did not predict well experimental data of E_{cat} due to metal contamination on the catalyst surface. It indicates that the fluidity correlation should be improved with experimental data of E_{cat} only and E_{cat} -fresh catalyst mixtures for reliable prediction and its application. For the same reason, it is very important to investigate the effect of fresh catalyst and additive addition on the catalyst fluidity in an E_{cat} fluidized bed, to ensure stable operation even under changing operating conditions of the FCC unit.

In this study, the fluidities of commercial fresh and equilibrium FCC catalysts with various contents of fine particles were measured to investigate the effect of fresh catalyst and additive addition for maintaining good fluidization quality of the FCC unit. An improved correlation based on the experimental data has been proposed for practical application in the FCC unit, such as fines makeup of the E_{cat} bed considering the situation of fines loss. A guidance for control of bed fluidity has been proposed based on the correlation.

2. Materials and Methods

Experiments were carried out in a cold model reactor, which is made of a transparent Plexiglas column, as shown in Figure 1. It consisted of a main column (0.05 m ID) and an expanded column (0.20 m ID), which is covered with wire mesh screen to retain fluidized particles inside the bed. Primary components are the compressor (NH-5, Hanshin), cylindrical column, wind box, sintered plate distributor (Stainless Steel Filter: pore size 40 μ m, Sunwoo sintered filter Co.), flow meter (RK1150, Kojima instrument), and manometer (series 1221, Dwyer). Air was injected into the column through the wind box and the distributor. Pressure taps were mounted flush with the wall of the column to measure pressure drops with gas velocity. Bed materials of 0.45 kg were loaded. To get the fluidity values (U_{mb}/U_{mf}) of the FCC catalysts, the U_{mf} and the U_{mb} were measured in a fluidized bed reactor.

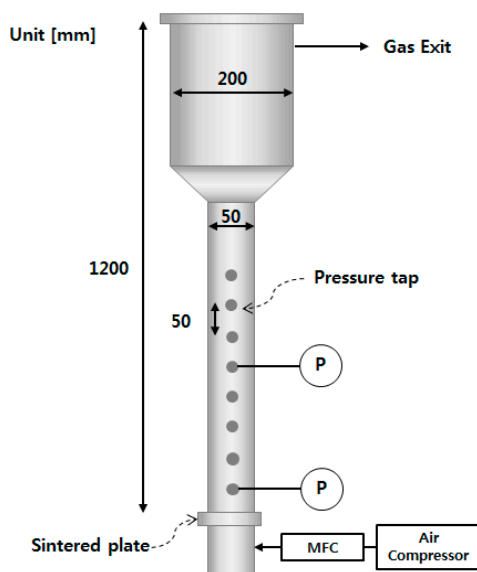


Figure 1. Schematic diagram of experimental apparatus.

The U_{mf} of the catalyst was obtained from a plot of pressure drop across the bed as a function of superficial velocity (U_g). The point of minimum fluidization was taken at the intercept of the fixed bed pressure drop, usually a linear function of U_g , and the pressure drop when the bed is fluidized, essentially determined by the weight of particles per unit area of the bed [14]. The U_{mb} was determined by the inspection of the appearance of the first obvious bubble in the bed after the minimum fluidization state as U_g is increased [12,14].

Seven types of FCC catalysts (Geldart group A [5]) and their mixtures were used as the bed material. The FCC catalysts have four major components: zeolite, matrix, filler, and binder. They have an internal porous structure with acid sites to crack larger molecules to desired size ranges [2]. The catalysts include two types of Equilibrium catalysts (Ecats), four types of fresh catalysts (Fcats), and an FCC additive. The Ecats were obtained from a commercial FCC unit in Korean refineries, which uses the different type of catalysts depending on main products of the unit, such as gasoline or propylene. The Fcats and additive in the study are commercial FCC catalysts provided by different catalysts companies. The Fcat-1 was manufactured by BASF, and the Fcats-2, 3, and 4 were manufactured by Albemarle. The additive is a commercial product (SmoothflowTM, Albemarle) having a high content of fine particles for improving catalyst fluidity in the process. The particle size distributions (PSDs) of all catalyst types are shown in Figure 2. All catalysts show a broad particle size distribution, mostly in the range between 10 to 180 μm , while the additive is in the range between 10 to 100 μm . In the PSD comparisons of FCC catalysts, the Ecats show lower amounts of particles less than 50 microns than the Fcats, due to the gradual loss of fine particles in the catalyst bed through the cyclone system during the numerous catalyst circulations in the unit. The Fcats differ in the amount of fine particles less than 50 microns between the catalysts, because the catalyst vendors produce the catalysts by controlling the amount of the fine particles according to the requirements of the refiners. The physical properties of the FCC catalysts are shown in Table 1. In the table, $d_{p,45+}$ represents the Sauter mean diameter of catalysts, except for the fine particles with a diameter of less than 45 μm . The scanning electron microscopy (SEM: S-4800, Hitachi) images of the catalyst particles are shown in Figure 3. As shown in the figure, the Ecats have a different shape compared to the Fcats and additives, because the Ecat particles are exposed to severe conditions, such as high temperature and attrition during the circulation in the unit. X-ray fluorescence spectroscopy (XRF: Axios-Petro, PANalytical) was used for the metal contents analysis of the catalysts. The Ecats show high metal contents, such as 0.53 wt. % of Ni, 1.21 wt. % of V, and 0.70 wt. % of Fe, while no Ni and V were detected in Fcats, and the amount of Fe was at 0.3 wt. %.

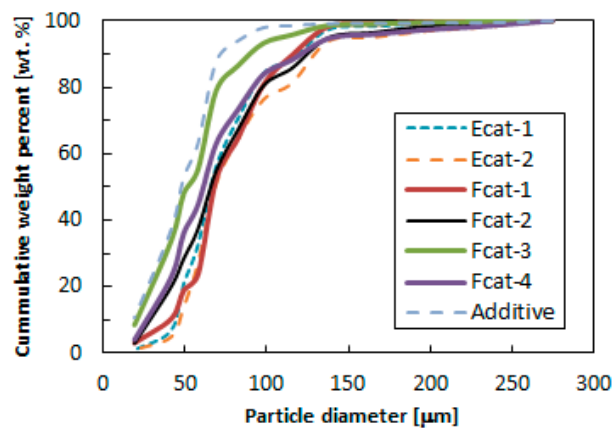


Figure 2. Cumulative particle size distributions of FCC catalysts used in this study.

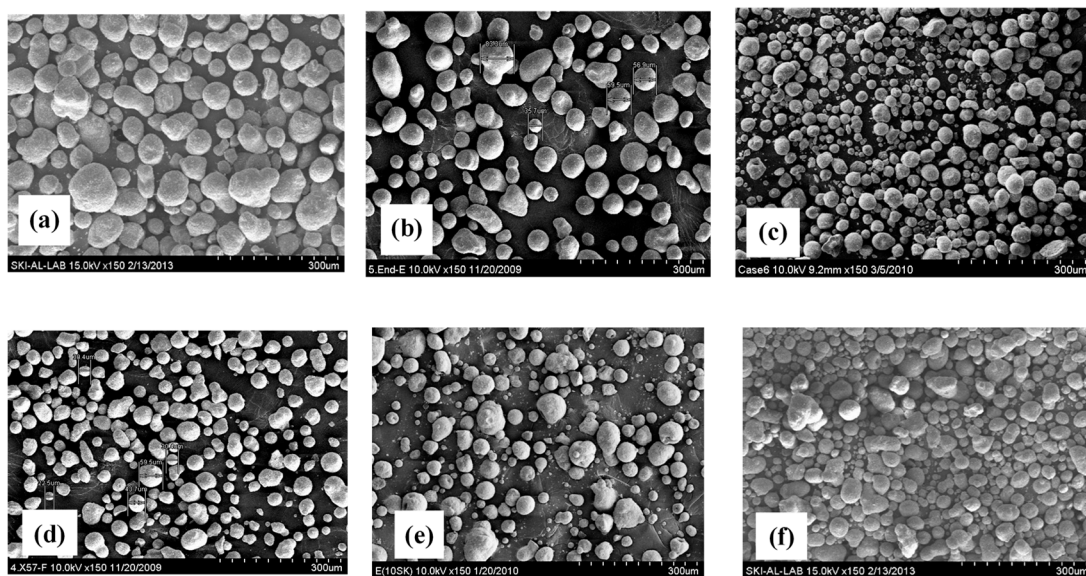


Figure 3. SEM (scanning electron microscopy) images of FCC catalysts used in this study: (a) Ecat-1, (b) Ecat-2, (c) Fcat-1, (d) Fcat-2, (e) Fcat-3, (f) additive.

Table 1. Physical properties of FCC particles in the study.

FCC Catalysts	Average Particle Diameter, d_p [μm] ^(a)	$d_{p,45+}$ [μm] ^(a)	Fine Contents [wt. %]	Apparent Density, ρ [kg/m ³] ^(b)
Ecat-1	65.7	67.5	6.8	1647
Ecat-2	68.6	70.1	4.9	1646
Fcat-1	67.8	71.1	10.4	1402
Fcat-2	65.3	74.0	20.0	1608
Fcat-3	50.3	63.1	33.9	1580
Fcat-4	60.5	67.8	23.0	1720
Additive	47.2	60.5	37.4	1625

Note: (a) based on the average of 3 repeated measurements; (b) based on the average of 5 repeated measurements.

In the experiments, 4 sets of catalyst mixtures were prepared in order to simulate the refinery situation, as summarized in Table 2. In set 1, the fluidity of each catalyst particle (7 catalysts) was measured. In set 2, the content of fine particles was adjusted by addition or removal of the fines in the Ecat-1 and Ecat-2 beds to simulate the plant's differential fines loss situation, such as the cyclone trouble [15]. In set 3, the Fcat-1 and Fcat-2 were added into the Ecat-1 bed without fine particles to simulate the Fcats make-up for restoring the fluidity of the bed from the fine loss situation. In set 4,

the additive was added into the Ecat-1 bed without the fine particles to show the effect of make-up of catalyst with much higher fines content on the fluidity of the Ecat bed.

Table 2. Summary of experimental sets on the fluidity measurement.

SET	Method	Remark
1	Measurement with 7 FCC catalysts in Table 1.	Effect of physical properties.
2	Measurement with the Ecat bed with controlled fines content by (1) adding fines to Ecat-1 without fines. (2) adding fines to Ecat-2 without fines.	Effect of fines contents in Ecats.
3	Measurement with the Ecat bed with controlled Fcat content by (1) adding Fcat-1 to Ecat-1 without fines. (2) adding Fcat-2 to Ecat-1 without fines.	Effect of Fcats make-up in Ecat bed without fines.
4	Measurement with the Ecat bed with controlled additive content by adding additive to Ecat-1 without fines.	Effect of additive make-up in Ecat bed.

3. Results

3.1. Effect of Fcat or Additive Make-up on Bed Fluidity

Effect of fines contents on fluidity of FCC catalysts (Ecats) is shown in Figure 4. The fluidity of the Ecats increases with increase of the fines content. It is often speculated that the fines act as a kind of lubricant. This lowers the apparent viscosity of the fluidized bed and leads to suppression of the forming of bubbles, with increased overall bed expansion by the more uniform gas-solid distribution. The amount of gas flowing interstitially is a function of the fines content [7,12]. Figure 4 shows that increment of 1 wt. % of fines increases about 0.03–0.04 of fluidity, depending on the Ecat type. The Geldart Group A particles with high fines contents retain gas longer than those with low fines, indicating that a certain fines loss from the bed could make poor fluidization in some regions, such as standpipe or wall region of the reactor [16].

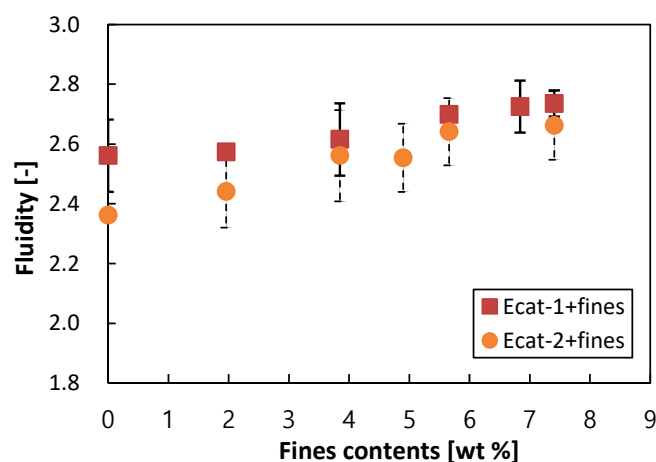


Figure 4. Effect of fines content on catalyst bed fluidity.

Effects of the fines content and average particle diameter (d_p) on minimum fluidization velocity (U_{mf}) are shown in Figure 5. The U_{mf} decreased with increasing content of fine particles in the catalysts with make-up of Fcats or additive into the bed. The U_{mf} is sensitive to parameters such as particle diameter, particle density, and fluid properties [17]. The fine particles introduced between the coarse particles could act as a lubricant in the bed to reduce the bed viscosity with the reduction of the

friction between the coarse particles, thereby leading to a decrease in the U_{mf} [5]. Additionally, the fine contents can be related to the voidage in the bed [6]. The increase of fine contents decreases the average particles diameter of the mixed FCC catalysts. The increased fines and decreased d_p affect the decrease of the U_{mf} as in Figure 5b.

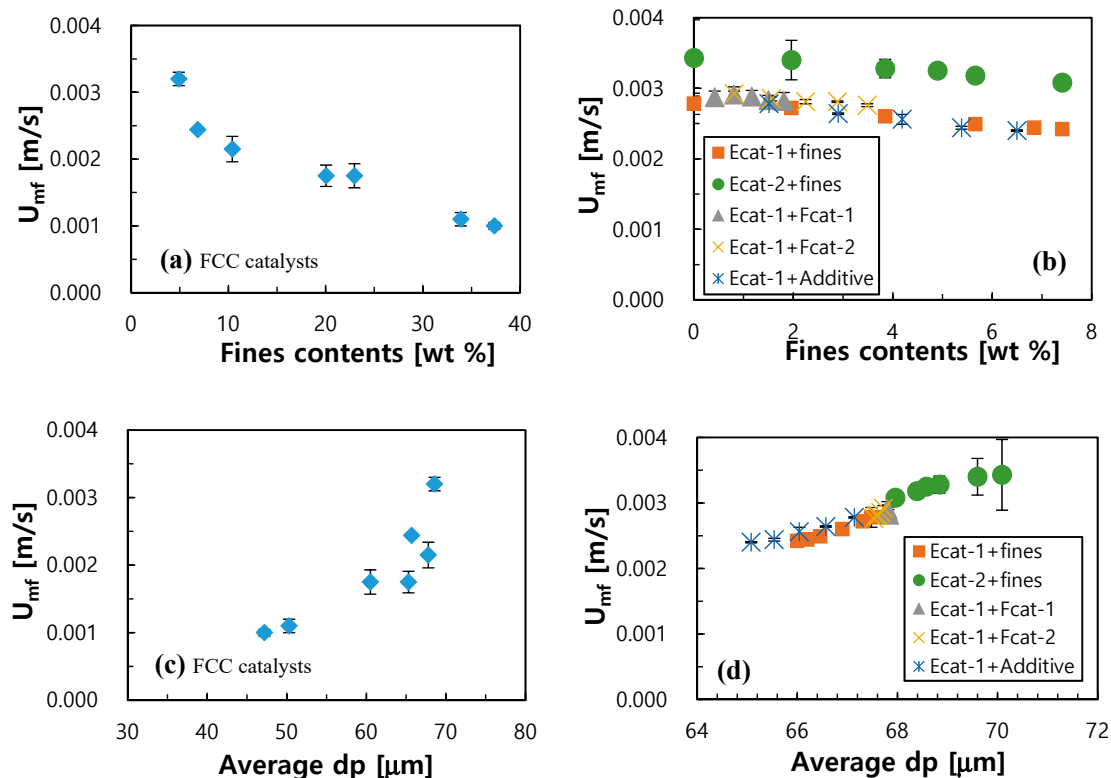


Figure 5. Effects of fine contents and average particle diameter on minimum fluidization velocity: (a) set 1, (b) sets 2–4, (c) set 1, (d) sets 2–4 in Table 2.

Effects of the fine contents and average particle diameter (d_p) on minimum bubbling velocity (U_{mb}) are shown in Figure 6. The U_{mb} did not show noticeable change with increasing the content of fine particles and average particle diameter in the catalysts with make-up of Fcats or additive into the bed. The U_{mb} is dependent upon fines content and particle diameter [5]. The increase of fines content in the bed enhances the inter-particle force, influencing the stability of the bubble-free regime [6]. However, the increase of fines content simultaneously decreases d_p , resulting in decrease of U_{mb} . However, it can be seen that the gap between U_{mf} and U_{mb} was obviously increased with the fines content from Figures 6 and 7, indicating expansion of the bubble-free regime after the fluidization of FCC particles.

Effects of fines content and normalized variation of particle mean diameter on the catalyst fluidity (U_{mb}/U_{mf}) are shown in Figure 7. It is well known that high contents of fines help to maintain good fluidity of the FCC catalyst bed. The increase of fine contents increases the fluidity of the Ecat bed, as in Figure 7a, indicating the addition of the catalyst with a high fines content will help to restore fluidity. The effect of fines in the catalysts on the fluidity has been well defined from a lot of studies with varying fines content in given FCC catalysts and the correlation derived from the results [7]. However, the particle size distribution (PSD) and the average particle size change, as well as the change of the fines content when the catalysts have different PSDs, are mixed with the existing catalyst bed. Therefore, it is necessary to consider the relative influence of the fine particles on coarse ones in the overall bed as the fines content varies [7,18], because it is difficult to quantify the PSD change of the bed. Regarding the PSD change, several studies [6,19] introduced a concept of relative spread in the PSD, but the results for its effect were not shown. In this study, the particles in the bed were

divided into coarse particles and fine particles based on 45 μm . A concept of variation of mean particle size with fines addition (normalized particle diameter variation) is applied to quantify the effect of mixed catalyst bed fluidity when the fine particle content in the coarse particle is changed.

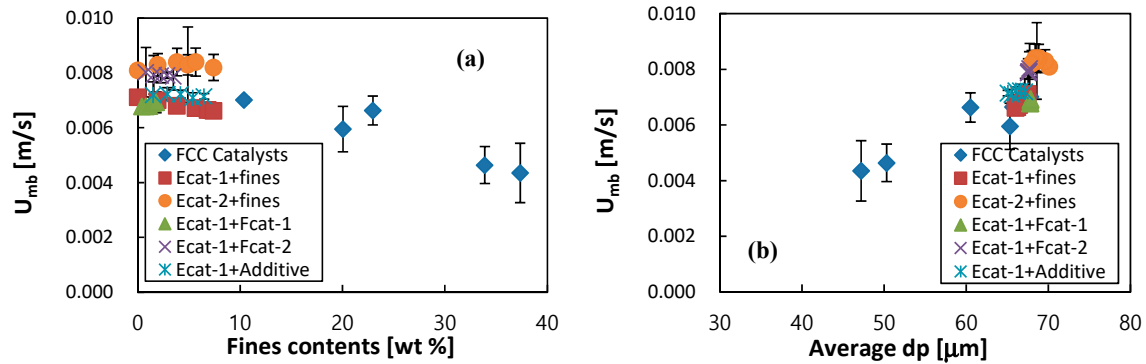


Figure 6. Effects of fine contents (a) and average particle diameter (b) on minimum bubbling velocity.

The normalized particle diameter variation is defined as Equation (1).

$$d_{p,n} = (d_{p,45+} - d_p) / d_{p,45+} \quad (1)$$

The fluidities of catalysts increase with increases of $d_{p,n}$, due to the increase of PSD and the decrease of mean diameter compared initial catalyst bed, as in Figure 7b. The increase of fluidity is affected not only by the PSD of the added catalyst, but also by the physical properties of the catalyst, considering the difference of the increasing slope.

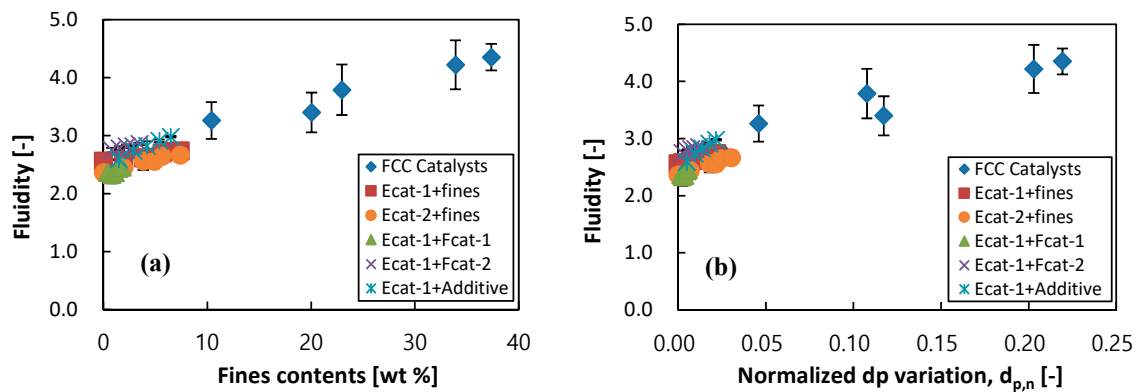


Figure 7. Effects of fine contents (a) and normalized particle diameter variation (b) on catalyst fluidity.

3.2. Correlation on Fluidity of FCC Catalysts

Correlation for the fluidity is useful for monitoring the unit of a given catalyst and interpreting the bed behaviors which may cause problems related to the fluidization [12]. Abrahamsen and Geldart's [5] correlation has been widely used for the fluidity. However, their correlation is not enough for exact prediction of catalysts make-up rate in urgent decisions after fines loss issues, such as catalyst circulation problems, because the correlation is for a wide range of Geldart A particles [11,20]. In this study, a guiding correlation has been proposed for fines makeup to the E-cat bed. The obtained fluidities for FCC catalysts with different fine fractions (F_{45}) have been correlated with dimensionless numbers based on the results of this study:

$$U_{mb}/U_{mf} = 2.09 \text{Ar}^{-0.25} (\exp F_{45})^{0.76} (d_{p,n} + 1)^{1.37} \quad (2)$$

with a correlation coefficient number of 0.93 and a standard error of 0.183. The range of variables in Equation (2) covers $5.8 \leq Ar \leq 19.3$, $0 \leq F_{45} \leq 0.37$, and $0 \leq d_{p,n} \leq 0.22$.

The fluidity values measured in the present study are compared with the values calculated by Equation (2) as shown in Figure 8. As can be seen, the calculated fluidity values predict well the experimental data within $\pm 18\%$. The experimental results are compared with published correlations [5,6,12] by the absolute average error, E as [21,22]:

$$E(\%) = (100/N) \sum |(\text{prediction} - \text{experimental})/\text{experimental}| \quad (3)$$

where N is data number. As can be seen in Table 3, the Equation (2) shows relatively good prediction, with an E value of 5.3%, compared to previous studies [5,6,12]. The correlation of the previous studies underestimated the experimental values. The E value results are well matched with the experimental report by Whitcombe et al. [8], where they concluded the fluidity and fluidization characteristics are affected by the different surface characteristics and shape of the Ecats compared to the Fcats [8,22].

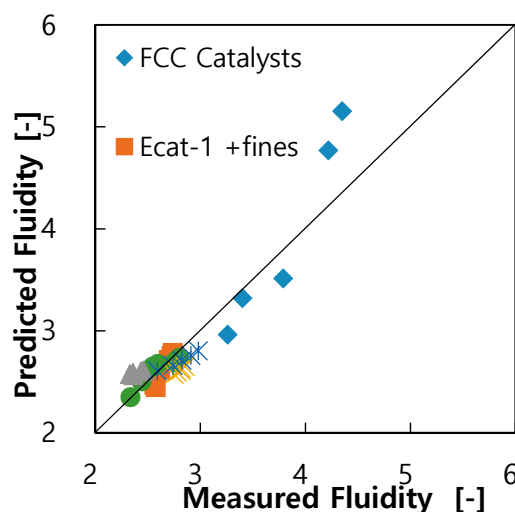


Figure 8. Comparison of the fluidity obtained by the proposed correlation and the experimental data.

Table 3. Mean percentage deviation between predicted and experimental fluidity values.

Authors	Correlations	E [%]
Abrahamsen and Geldart [5]	$\frac{U_{mb}}{U_{mf}} = \frac{2300\rho_g^{0.126} \exp(0.716F_{45})}{(\rho_p - \rho_g)^{0.934} g^{0.934} d_p^{0.8}}$	22.3
Xie and Geldart [6]	$\frac{U_{mb}}{U_{mf}} = \frac{330\rho_g^{0.19} \mu_g^{0.37} \exp(0.716F_{45})}{(\rho_p - \rho_g)^{0.934} g^{0.934} d_p^{0.8}}$	37
Singh and Roy [12]	$\frac{U_{mb}}{U_{mf}} = \frac{0.5231(d_p/D_c)^{1.13}(D_c/h_s)^{-0.0384}(\rho_p/\rho_g)^{0.74}}{[(\rho_p - \rho_g)^{0.934} g^{0.934} d_p^{1.8}] / (1100\rho_g^{0.066} \mu_g^{0.87})}$	674.2 ¹
This study	$\frac{U_{mb}}{U_{mf}} = 2.09 Ar^{-0.25} (\exp F_{45})^{0.76} (d_{p,n} + 1)^{1.37}$	5.3

¹ The correlation is based on U_{mb} correlation from experiments on Geldart B particles.

3.3. Guidance for Control of Bed Fluidity

Many industrial fluidized bed reactors operate on a catalyst bed, with a high fines content of 10 to 50 wt % for improved fluidized bed performance, and thus higher yields [7]. It is noticed that the fines loss usually occurs during the change of operating condition in the FCC unit, including start-up, charging-up of feed, and shut-down. Comparative test data on the fluidity may show quickly if a fines loss problem develops [11]. The refiners take the action of adding fresh catalysts or fluidity enhancing additives with a large number of fine particles into the reactor to restore the fluidity of the catalyst in the unit. However, there is an associated cost in maintaining or restoring a high level of fines. A better guidance for effective catalyst make-up for keeping or restoring the bed fluidity is required, because

the catalyst make-up costs may be higher. The proposed Equation (2) is believed to provide a PSD management guide for making the fluidization quality good in the unit by providing the desired PSD information of fresh catalysts for the make-up [18]. On the basis of the available findings and results, the following suggestion would likely be useful in quickly restoring the bed fluidity in various fines or catalyst loss occasions. First, the monitoring of the fines content and fluidity obtained from the direct measurement [2] or the CFD simulation [9,10] in normal operating conditions is required to define the unit's own fluidity characteristics, due to each FCC unit's operational limits, such as cyclone configuration and efficiency [23]. Second, check the fines and the fluidity in case of temporary abnormal operating conditions, such as catalyst losses. Finally, determine the make-up rate of the fresh catalysts to reach an optimum fluidity level from the proposed correlation before catalyst fluidity gets worse.

4. Conclusions

The effect of catalyst fine contents on the FCC catalyst bed fluidity was determined. The fluidity decreases with loss of fine content and increases with increment of makeup of fresh catalysts or additive with the controlled fines content. The fluidities of catalysts increase with increases of normalized particle diameter variation by the fines addition. A guiding correlation on the FCC catalyst fluidity for the makeup of fines to E-cat bed has been proposed based on the fine contents, Archimedes number, and normalized particle diameter variation. The proposed correlation could guide to keep good catalyst fluidity in the FCC unit.

Author Contributions: S.W.K. and D.Y.L. conceived and designed the research. C.E.Y. analyzed the testing data. S.W.K. wrote the paper. S.W.K. and D.Y.L. supervised.

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Conflicts of Interest: The authors declare no conflict of interest.

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