Determination of pressure drop in a fixed bed catalytic reactor during ammonia oxidation on nanostructured catalyst

Oksana Dobrovetska[1], Diana Kindzera[2], Roram Hosovskyi[3], Volodymyr Atamanyuk[4]

1. Department of Chemical Engineering, Lviv Polytechnic National University, UKRAINE, Lviv, 12 S. Bandery Str, E-mail: oksanadobrovetska@gmail.com
2. Department of Chemical Engineering, Lviv Polytechnic National University, UKRAINE, Lviv, 12 S. Bandery Str., E-mail: kindzera74@ukr.net
3. Department of Chemical Engineering, Lviv Polytechnic National University, UKRAINE, Lviv, 12 S. Bandery Str., E-mail: roma5477@meta.ua
4. Department of Chemical Engineering, Lviv Polytechnic National University, UKRAINE, Lviv, 12 S. Bandery Str., E-mail: atamanyuk@ukr.net

Abstract* – The effect of the increasing gas velocity on the pressure drop in the fixed beds with a length of $35\cdot10^{-3}$, $45\cdot10^{-3}$, $55\cdot10^{-3}$, $65\cdot10^{-3}$ and $75\cdot10^{-3}$m which were generated in the downflow reactor has been established by experimental method. Ergan and Darcy-Weisbach equations have been used as the numerical methods to calculate the pressure drops within the Reynolds number range of $312 \leq Re \leq 1177$. The difference between experimental and calculated values of pressure drops was found to be increased with the increase of the Reynolds number. Maximal relative error between experimental and theoretical values does not exceed ± 3%.

Keywords – nanostructured catalyst, fixed bed reactor, low-temperature ammonia oxidation process

Introduction

Nowadays, ammonia and methanol synthesis, methane reforming with carbon (IV) oxide, the ethane, hydrochloric acid and sulfur (IV) oxide oxidation, as well as production of hydrogen by the rich combustion of heavy fuel oil and waste treatment are carried out in fixed bed catalytic reactors, also called packed bed catalytic reactors [1-3]. These reactors are the most commonly used in large-scale production of chemicals. In the reactors the continuous phase is a fluid or gas and the porous phase consists of catalytic granules which can be loaded into the reactor as single bed units, multi-bed units, and multi-tube units [4]. Oxidation of ammonia (NH_3) in the presence of catalysts takes place with the formation N_2, N_2O and NO and the resulting product depends on the process conditions and the catalyst nature [5]. The catalytic oxidation of ammonia to nitrous oxide (N_2O) at low temperatures is an efficient technique and it can be intensified by the application of new selective catalysts [6].

High activity at low-temperature ammonia oxidation and high selectivity for N_2O are shown by Pt-based catalysts [7]. The technology of obtaining nanostructured catalysts attracts the special attention of researchers due to the high activity of the catalyts [8]. Therefore, nanostructured platinum catalysts obtained by electrochemical method are promising and their application will provide the intensification of the ammonia oxidation process in the fixed bed reactor.

Results and discussion

The pressure drops were measured for fixed beds with length of $35\cdot10^{-3}$, $45\cdot10^{-3}$, $55\cdot10^{-3}$, $65\cdot10^{-3}$, $75\cdot10^{-3}$ m and experimental curves are presented in Fig. 1.

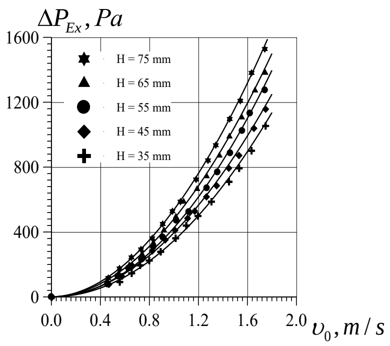

Fig. 1. The pressure drop in fixed beds of different length vs. gas superficial velocity.

In general, Fig. 1 demonstrates that the increase in the gas superficial velocity increases the pressure drop in fixed beds. The effect of fixed beds' length on the pressure drop is significant and, as the example, it is evident, that at the actual velocity of 1.78 m/s the pressure drop is changing from 985 Pa for the length of the fixed bed H = $35 \cdot 10^{-3}$ m to 1530 Pa for the length of the fixed bed H = $75 \cdot 10^{-3}$ m.

To determine the pressure drop in the fixed beds of different length when passing the gas flow through them, the Ergan equation (1) and Darcy-Weisbach equation (2) were used.

$$\frac{\Delta P}{H} = 150 \frac{\mu \cdot (1-\varepsilon)^2 \cdot}{d_p^2 \cdot \varepsilon^3} \upsilon_0 + 1{,}75 \frac{\rho \cdot (1-\varepsilon)}{d_p \cdot \varepsilon^3} \upsilon_0^{\,2} \quad (1)$$

$$\Delta P = \lambda \cdot \frac{H_e}{d_e} \cdot \frac{\rho \cdot \upsilon_0^2}{2} \quad (2)$$

So, Ergan equation allows to predict accurately the pressure drop in the fixed bed up to Reynolds number of 850, but with the increasing of Reynolds number above 850, the error between experimental and calculated values increases due to increased gas turbulence in the channels between particles of the catalyst, when the effect of inertial component increases and the effect of the viscous component decreases.

Since the Euler number includes the velue of the pressure drop, the equation may be represented as:

$$\Delta P = 49 \cdot Re_{exp}^{\;-0{,}11} \cdot \left(\frac{H_e}{d_e}\right) \cdot \left(\frac{d_p}{D}\right) \cdot \rho \cdot \upsilon^2 \quad (3)$$

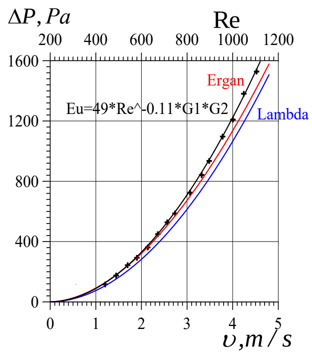

Fig. 2. Pressure drops in the fixed bed of the length H=$75 \cdot 10^{-3}$ m versus actual gas velocities within the Reynolds number range of 312 ≤ Re ≤ 1177

Graphic dependencies in Fig. 2 show that values of the pressure drop obtained by using Eq. (3) accurately describe the experimental data.The maximal relative error between experimental and theoretical values does not exceed ± 3%. Therefore, the proposed equation (3) allows us to calculate theoretically with high accuracy the pressure drop in the downflow fixed bed reactor within Reynolds number range of $312 \leq Re_{exp} \leq 1177$.

Conclusion

The effect of the increasing gas velocity on the pressure drop in the fixed beds of nanostructured platinum catalyst with the length of $35 \cdot 10^{-3}$, $45 \cdot 10^{-3}$, $55 \cdot 10^{-3}$, $65 \cdot 10^{-3}$ and $75 \cdot 10^{-3}$m which were generated in the downflow fixed bed reactor has been established by experimental method. Ergan and Darcy-Weisbach equations have been used as the numerical methods to calculate the pressure drops and the graphic dependencies have been plotted to compare the results with experimental data. The error between values of pressure drops was found to be increased at Reynolds number above 850.

Generalizing the experimental results, the equation $\Delta P=49 \cdot Re_{exp}^{-0,11} \cdot (H/d_e) \cdot (d_p/D) \cdot \rho \cdot \upsilon^2$ has been proposed which allows to calculate theoretically with high accuracy the pressure drop in the downflow fixed bed reactor with new catalyst within Reynolds number range of $312 \leq Re_{exp} \leq 1177$. The maximal relative error between experimental and theoretical values does not exceed ± 3%.

References

[1] Sandeep, K. C., Mohan, S., Mandal, D., Mahajani, S. (2019) Determination of gas film mass transfer coefficient in a packed bed reactor for the catalytic combustion of hydrogen, v. 202, 508-518. https://doi.org/10.1016/j.ces.2019.02.042 (Chemical Engineering Science)

[2] Sun, Z., West, D. H., Balakotaiah, V. (2019) Bifurcation analysis of catalytic partial oxidations in laboratory-scale packed-bed reactors with heat exchange, v. 377, 1197651. https://doi.org/10.1016/j.cej.2018.08.151 (Chemical Engineering Journal)

[3] Gómez, J., Mmbaga, J.P., Hayes R.E., Toledo, M., Gracia, F. (2018) Modeling hydrogen production in a catalytic-inert packed bed reactor by rich combustion of heavy fuel oil, v. 43, 2677-2688. https://doi.org/10.1016/j.ijhydene.2017.12.056 (International Journal of Hydrogen Energy)

[4] Bazmi, M., Hashemabadi, S.H., Bayat, M., (2011) Modification of Ergun equation for application in trickle bed reactors randomly packed with trilobe particles using computational fluid dynamics technique, v. 28, 1340-1346. https://doi.org/10.1007/s11814-010-0525-8 (Korean Journal of Chemical Engineering)

[5] Manna, M.V., Sabia, P., Ragucci R., Joannon M. (2020) Oxidation and pyrolysis of ammonia mixtures in model reactors, v. 264, 116768, https://doi.org/10.1016/j.fuel.2019.116768 (Fuel)

[6] N.Yu. Masalitina, Ph.D. Thesis, National Technical “Kharkiv Polytechnic Institute”, Ukraine, Kharkiv, 2017

[7] Ma, H., Schneider., W. F. (2020) DFT and microkinetic comparison of Pt, Pd and Rh-catalyzed ammonia oxidation, v. 383, 322-330. https://doi.org/10.1016/j.jcat.2020.01.029 (Journal of Catalysis)

[8] Saldan, I., Girella, A., Milanese, C., Fratini, F., Dobrovetska, O., Levchuk, I., Kuntyi, I. (2018) Size and distribution of palladium nanoparticles electrodeposited on graphite, v. 25. 82-87, doi:https://doi.org/10.15407/fm25.01.082 (Functional Materials)