Mutual Verification of the Gas-Phase Volumetric Mass-Transfer Coefficients Determined in the Packed Absorption Column

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Abstract. When using the rate-based approach to the modeling of packed columns, there are problems with the agreement of data obtained at different workplaces with different systems. This work should experimentally verify the mutual consistency of transport data and a common model of a packed absorption column. The two most used methods for k_Ga determination are the chemisorption of sulfur dioxide into a sodium hydroxide solution and of ammonia into a diluted sulfuric acid solution. However, the published k_Ga values obtained at different workplaces using the same method differ by up to several times. It is therefore not possible to verify the assumed dependencies of k_Ga on the system properties. The aim of this work is to determine k_Ga by two methods, in order to compare the values measured at one workplace by two different methods and to determine whether the conversion approaches are correct.

Keywords Verification, Mass-transfer, Gas coefficient, Packed columns

Introduction

Absorption is important process mainly due to its wide application in the chemical industry. Absorption is used in the purification of waste gases, for example in the removal of acid gases from industrial exhalations (washing with chlorine with sodium hydroxide, sulfur dioxide with ammonia solution, carbon dioxide with amine solution, absorption of acetone vapors into water, etc.). However, this process can also be used in the production of important inorganic acids, because the absorption of nitrogen dioxide or sulfur dioxide into dilute solutions, nitric or sulfuric acid is produced. Furthermore, absorption is applicable in the food industry for the preparation of carbonated beverages, in the aeration of fermenters and in many other processes. Absorption columns can be packed with random or structured packing.
However, it is not possible to measure packing properties for all different combinations of industrially used packings, systems and conditions. Thus, there is an effort to create general correlations that can recalculate the transport and hydraulic properties to a completely different conditions than the ones on which they are based or have been verified. However, the current recalculations fail and the literature values of transport properties obtained by different models differ from each other even in comparison with the experimentally obtained data[6].
The ambition of this work is to contribute to the study of mass transport in packed towers by determining the volumetric mass transfer coefficient in the gas phase (k_Ga) by two methods at one workplace and one device and consequently evaluate k_Ga dependence on the diffusion coefficient, which is an important indicator of the mechanism of mass transport. The k_Ga is a transport characteristic of the packing and is most often obtained by two methods, chemisorption of sulfur dioxide into a solution of sodium hydroxide and ammonia into a solution of dilute sulfuric acid.

Experimental part

The measurements were performed on two columns with different diameters (DN150 and DN290) and adjustable heights. The columns were assembled from several sections of different heights, so that the height of the packed bed and the total height of the column could be adjusted. The column was packed with Mellapak 250Y structured packing from Sulzer Ltd. The measurement was performed with 1-4 packing elements, where the height of one piece is 210 mm. When using several elements, the individual elements were always rotated by 90° compared to the previous element.
In the determination of k_Ga by the classical method (Eq. 1), gas phase samples are taken just below and above the packing in order to determine the mass transfer, which takes place only at the packing and not in other parts of the column. For the calculation of k_Ga, we assume a countercurrent plug flow of the liquid and gas phases.

$$k_G a = \frac{u_G}{H} ln \frac{c^{in}_{NH_3,G}}{c^{out}_{NH_3,G}} \quad (1)$$

With the subtraction method, better reproducibility of the k_Ga values has been achieved, but this method is more complicated to perform experimentally. The gas phase samples are taken before the gas stream enters the bottom of the column and behind the measuring orifice, where the outlet gas is well mixed. To determine k_Ga related only to packing, a series of measurements have to be made on different numbers of packing with the same liquid and gas flow rates, the same NH_3 input concentration (dependence of end effects on NH_3 concentration). In order to maintain the same size of ends the column was always set up so that the distance of the packing from the bottom of the column and from the liquid phase distributor was the same. The k_Ga related only to packing is possible to evaluate from measurements on two packed bed heights from the Eq. 2.

$$k_G a = \frac{1}{H_2 - H_1} \cdot (k_G a_2 \cdot H_2 - k_G a_1 \cdot H_1) \quad (2)$$

Results and discussion

The confirmation of the methodology (subtraction method) was ensured by measuring on a different number of packing elements and with different input NH_3 concentration for liquid velocities B = 5 m/h and 10 m/h. For B = 5 m/h, a series of measurements was performed on 2, 3 and 4 packing elements for u_G in the range of 0.5m/s to 3 m/s. For B = 10 m/h, a series of measurements was performed on 1, 2 and 3 packing elements for u_G in the range of 0.5 m/s to 2.5 m/s. Each point was measured several times with different input concentrations ranging from about 1600 to 200 ppm (the error bars are shown in Fig. 5, 6, 7 and 8). The choice of the input concentration depended on the gas phase velocity and the number of packing elements, so that the value at the outlet did not fall below 5 ppm and it was possible to determine it with analyzer reliably. When calculating the k_Ga value according to the equation (1) from the NH_3 concentration measured just below and above the packed bed, a significant dependence of k_Ga on the input concentration is evident. However, when calculating k_Ga by the subtraction method, this dependence is greatly suppressed and at lower gas velocities even disappeared completely. This confirmed one of the basic assumptions of the methodology. Because the measurement took place at 3 different packed bed heights, it was possible to determine k_Ga from three combinations of packed bed elements. From the calculated values the average value for individual velocities was determined and all these values were plotted in the graph. Fig. 1 and Fig. 2 show that all values are the same for both liquid velocities, thus confirming another measurement assumption that the k_Ga values do not depend on the used packed bed height.

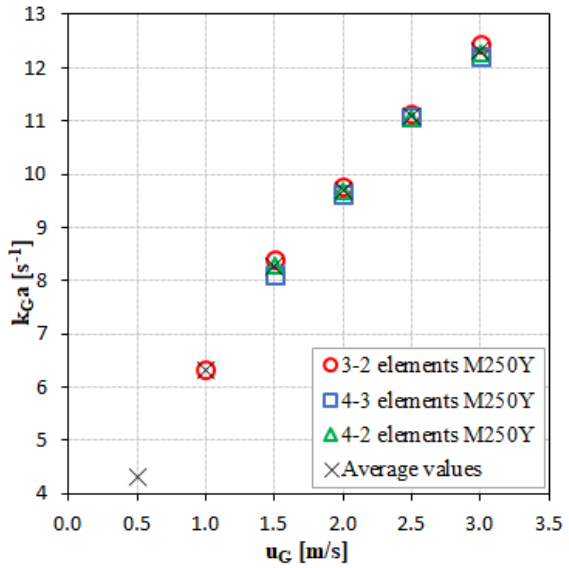

Fig. 1 Dependence of k_Ga values on packed bed height, DN150, B = 5 m/h, M250Y

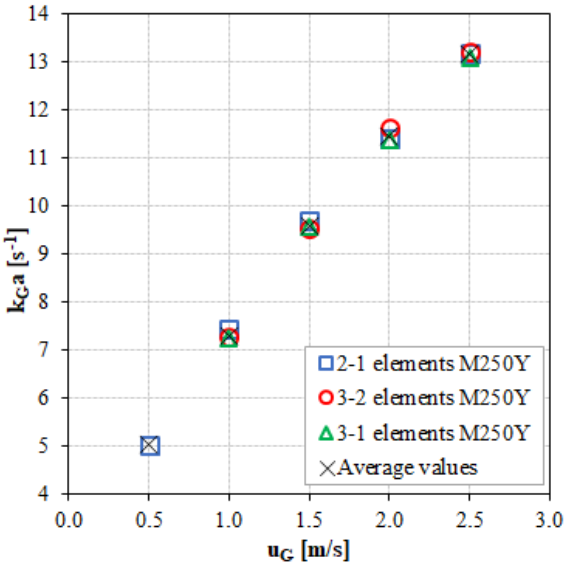

Fig. 2 Dependence of k_Ga values on packed bed height, DN150, B = 10 m/h, M250Y

The last verification of the methodology was a comparison of the values obtained on a column with a larger diameter, on which it was previously verified in our laboratory on the SO_2 / NaOH system that the effect of the input concentration on k_Ga values is significantly smaller. Measurements were performed on a DN290 column on 2 and 3 packing elements, again for liquid velocities B = 5 m/h and 10 m/h. The k_Ga values for DN290 were determined by

the subtraction method at two different concentrations and were plotted together with the average values obtained on the DN150 column. From the Fig. 3 and Fig. 4 it is obvious at first sight that these values are the same (the values differ by a maximum of 2%) and thus the correctness of the measurement methodology was definitively confirmed.

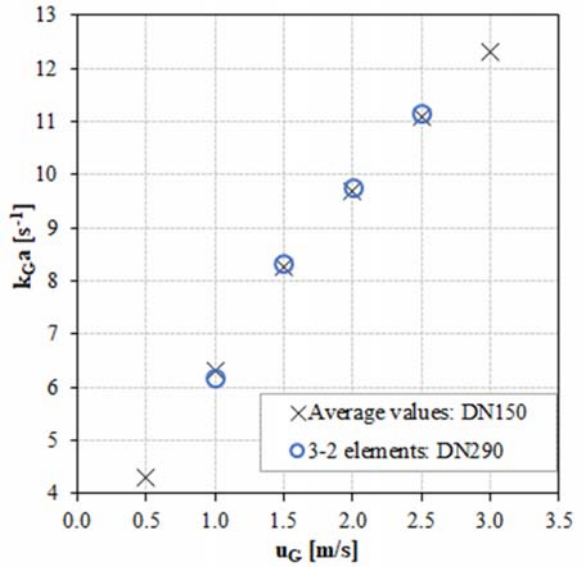

Fig. 3 Comparison of k_Ga values determined on columns with DN150 a DN290, *B* = 5 m/h, M250Y

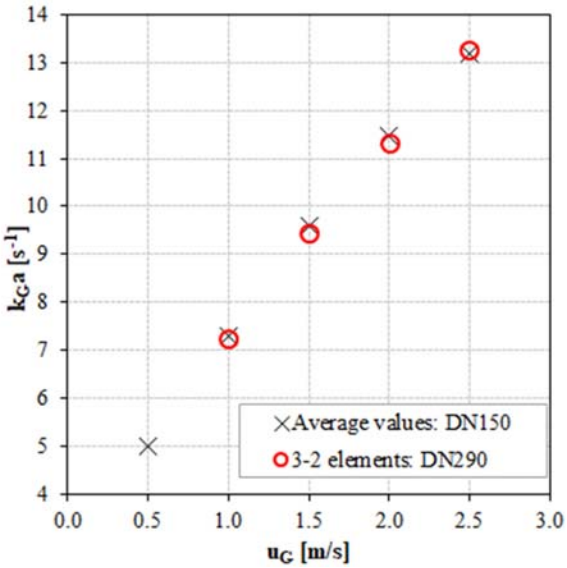

Fig. 4 Comparison of k_Ga values determined on columns with DN150 a DN290, *B* = 10 m/h, M250Y

Due to measurements at different input NH_3 concentrations and different combinations of packing elements subtraction (e.g. 4 - 3 elements, 3 - 2 elements, etc.), several k_Ga values were determined for one combination of velocities B and u_G. From all variants, the average k_Ga values were calculated and plotted as a function of u_G in the Figs. 5, 6, 7 and 8 for liquid velocities B = 5, 7.5, 10 and 20 m/h. For each point, error bars calculated by the student distribution with a 90% confidence interval are displayed. The points were fitted by the power function and the equations are shown in the corresponding figures. For B = 5 m/h and 10 m/h k_Ga depends on u_G with a power of about 0.6, for B = 7.5 m/h the power is slightly higher and has a value of 0.66 and for B = 20 m/h the value of the exponent even higher and equal to 0.75. All dependences are in the range of values reported in various computational models[1, 2].

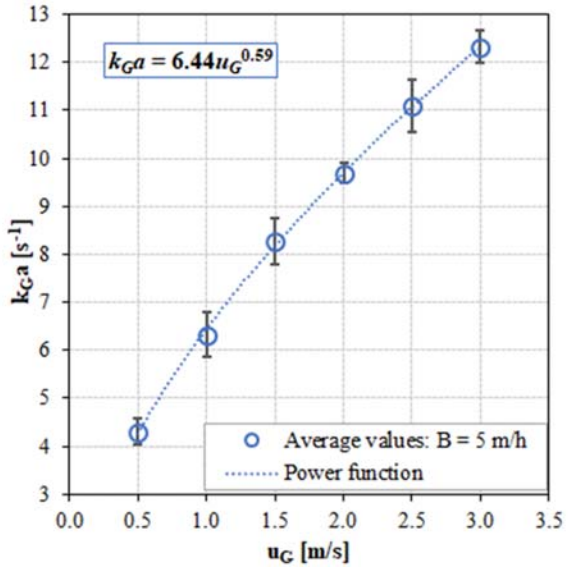

Fig. 5 Dependence of k_Ga values on u_G, DN150, M250Y, B = 5 m/h, error bars represent 90% confidence interval

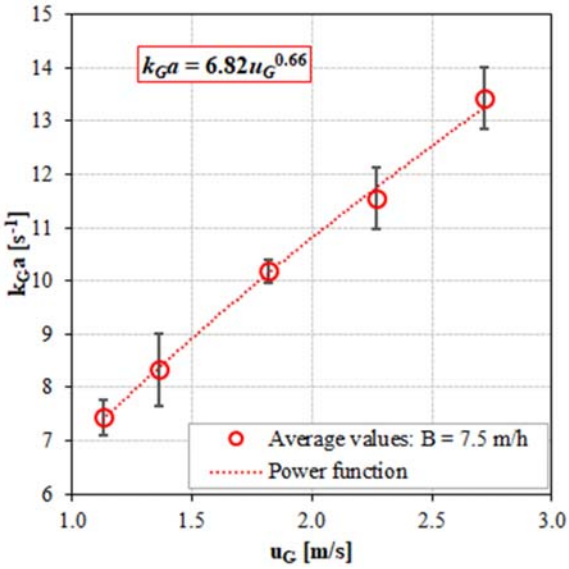

Fig. 6 Dependence of k_Ga values on u_G, DN150, M250Y, B = 7.5 m/h, error bars represent 90% confidence interval

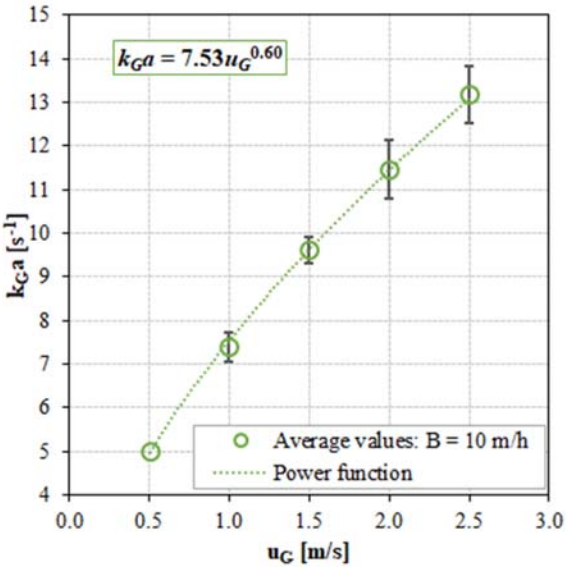

Fig. 7 Dependence of k_Ga values on u_G, DN150, M250Y, B = 10 m/h, error bars represent 90% confidence interval

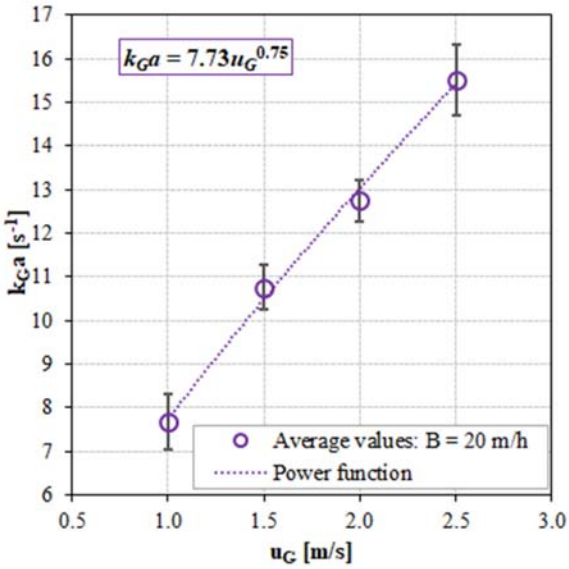

Fig. 8 Dependence of k_Ga values on u_G, DN150, M250Y, B = 20 m/h, error bars represent 90% confidence interval

It is now possible to compare k_Ga values determined by two different methods and thus determine the dependence of k_Ga on the diffusion coefficient from experimental data on the packinng, which has not yet been published. The method of SO_2 / NaOH chemisorption has been measured previously in our workplace and published in Rejl[4]. The measurement took place at the same workplace, the same experimental equipment was used, the same structured packing and measurement methodology was very similar. The change of the measuring system does not change the physical properties of the carrier phases and thus, from these experimental values, the power of the power dependence of k_Ga on the diffusion coefficient can be calculated according to Eq. (3). The k_Ga values used for the calculation are given in Table 1 and the individual powers were calculated using both the average k_Ga values determined by the NH_3 / H_2SO_4 method and the values after subtracting or adding the value of the standard deviation. The results of the powers are given for all variants in Table 2. The power calculated from the average k_Ga values are higher for a higher liquid velocity B = 10 m/h than for B = 5 m/h and in both cases they decrease with increasing gas velocity. For B = 5 m/h

the values of the powers range from 0.15 to 0.33 and for B = 10 m/h in the range from 0.29 to 0.42, the average value from these data is 0.27. If we also take into account the calculations of extreme points with subtraction or addition of the standard deviation, then the values of the power are on average 0.18 and 0.32. These values are much smaller than basic theories and models assume. The RBF[2] and Billet and Schultes[1] models account for a power of 2/3, and the Delft model[5] even uses a dependency with a power of 0.7 to 0.85 in its equations. On the other hand, our power value corresponds to the dependence used in the UCT model[3], where the exponent value is in the range of 0.19 to 0.35. The data obtained in this work and in Haidl et al.[3] show that it is very likely that models, which uses the kGa dependence on diffusion coefficient derived from data measured on a wet-walled column, cannot be applied to structured packings as well, because the values calculated using such models overestimate the dependence of $k_G a$ on the diffusion coefficient.

$$m = \frac{\ln\left(\frac{k_G a_{SO_2/NaOH}}{k_G a_{NH_3/H_SSO_4}}\right)}{\ln\left(\frac{D_{SO_2/air}}{D_{NH_3/air}}\right)} \tag{3}$$

Table 1: Values of $k_G a$ determined at the UCT Prague using the SO_2/NaOH and NH_3/H_2SO_4 methods

Method	SO_2/NaOH (Rejl[4])		NH_3/H_2SO_4 (experimental data)					
	$k_G a$		average $k_G a$		$k_G a$ minus standard deviation (SD)		$k_G a$ plus standard deviation (SD)	
u_G [m/s]	***B*** [m/h]							
	5	10	5	10	5	10	5	10
0.5	3.47	3.82	4.31	5.02	4.03	5.02	4.59	5.02
1.0	5.45	5.99	6.32	7.39	5.85	7.06	6.79	7.72
1.5	7.09	7.80	8.28	9.61	7.79	9.31	8.77	9.91
2.0	8.55	9.41	9.7	11.46	9.48	10.80	9.92	12.12
2.5	9.89	10.88	11.09	13.18	10.55	12.52	11.63	13.84
3.0	11.14	12.25	12.32	-	11.99	-	12.65	-

Table 2: Power values in power dependence of $k_G a$ on diffusivity

Power from Eq. 3	SO_2/NaOH (Rejl[4])					
	NH_3/H_2SO_4 average $k_G a$		NH_3/H_2SO_4 $k_G a$ minus SD		NH_3/H_2SO_4 $k_G a$ plus SD	
u_G [m/s]	***B*** [m/h]					
	5	10	5	10	5	10
0.5	0.33	0.42	0.23	-	0.42	-
1.0	0.23	0.32	0.11	0.25	0.33	0.38
1.5	0.23	0.32	0.14	0.27	0.32	0.36
2.0	0.19	0.30	0.16	0.21	0.22	0.38
2.5	0.17	0.29	0.10	0.21	0.25	0.37
3.0	0.15	-	0.11	-	0.19	-
Average	0.27		0.18		0.32	

Conclusion

The k_Ga values for the structured packing Mellapak 250Y for liquid velocities B = 5, 7.5, 10 and 20 m/h and gas velocities in the range of 0.5 to 3 m/s were determined by a verified methodology. For individual liquid velocities, the dependence of k_Ga on the gas velocity was found to be power with a power value of 0.6 (B = 5 m/h), 0.66 (B = 7.5 m/h), 0.6 (B = 10 m/h) and 0.75 (B = 20 m/h).

Finally, the k_Ga values obtained at one workplace, two different methods of k_Ga determination (ammonia chemisorption into sulfuric acid solution and sulfur dioxide chemisorption into sodium hydroxide solution) were compared and the dependence of the mass transfer coefficient on diffusivity was evaluated. The mass transfer coefficient in the gas phase depends on the diffusivity by power law with an average value of the power of 0.27. This finding confirms a similar dependence used in the UCT model[3], but does not agree with most models and theories used so far, which state the dependence of k_Ga on diffusivity with a power of 2/3 from the boundary layer theory or even 0.7 - 0.85 in the Delft model[5]. This fact could fundamentally affect gas-phase mass transfer models and calls into question the reliability of correlations in transfer to systems and conditions other than those under which they were derived or verified. To confirm this fact, it will be necessary to perform more experiments on different packings, both structured and random. A great advantage in the problem of k_Ga dependence on diffusivity would be obtaining additional data with a measuring system for determining the volume coefficient of mass transfer in the gas phase with a different value of diffusivity of the transition component (experiments with liquid evaporation into the gas stream could be suitable).

Acknowledgements

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