

Further Development of the Guideline for Standardized Characterization Measurements of Packed Columns

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Abstract. Modeling is commonly applied for dimensioning of absorption columns. Therefore, high significance of the underlying mass-transfer parameters is required. However, in literature an inconsistency of reported measurement data is present. Thus, several papers [1–5], as well as the VDI guideline 2761 Part 2 [6], deal with the accurate experimental determination of hydraulic and mass-transfers characteristics of packing. The aim of the current work is to experimentally test the measurement method and develop it further. Therefore, hydraulic and mass-transfer characteristics of the packing Raschig Super-Pak 250Y were determined. The measurements were performed with a pilot plant consisting of a DN 450 measurement column and a DN 600 saturation column. In a next step, those parameters and procedures will be investigated in detail, which are not adequately described yet.

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Keywords Standardization, Mass-Transfer Parameters, Absorption Column, Packing, Characterization Measurements

Introduction

Absorption is a process widely used in chemical engineering. During this process one or more gaseous components are dissolved in a scrubbing liquid. This can be either by physisorption, whereby no chemical reaction takes place, or chemisorption. The reverse process is called desorption. To increase the mass-transfer area in absorbers, internals are applied. Most commonly, these internals are random or structured packings. In packed columns, high mass-transfer areas and low pressure drops can be realized as well as wide ranges of gas and liquid loads. Further, a high number of separation stages can be achieved in counter-current operation [7].

For dimensioning of packed columns, modeling is applied to reduce experimental effort and costs [3, 4]. Therefore, considerable research has been put into the development of suitable models [1, 8]. However, the models cannot deliver reasonable results if the underlying mass-transfer parameters are inaccurate. These parameters, meaning the effective interfacial area and the gas- and liquid-side mass-transfer coefficients, have to be determined experimentally. Thus, the accuracy of the models depends on the quality of the experimental packing characterization. Since data obtained by various authors in literature is inconsistent, high safety factors have to be applied and columns are often overdimensioned. Therefore, it has been aimed for standardization of the characterization measurements of packed columns [1–5]. A method was developed and summarized in the VDI guideline 2761 Part 2. This guideline gives, amongst others, recommendations about the test column, measurement procedure and model-assisted evaluation [6].

By a standardized procedure for mass-transfer measurements, the comparability and reliability of obtained data can be increased [1, 3–5]. More reliable data can also be beneficial for further developing models [3]. In consequence, more accurate models reduce the need for oversizing in industrial column design [1–5] and therefore investment and operating costs [4, 5]. Furthermore, more accurate data provides also advantages for packing manufacturers. As the performance classification of newly developed packings is improved, advantages against well-established packings can be proven [3].

However, the influences of some parameters on packing characterization are not completely clear yet. For example, the influence of the sodium hydroxide concentration on the determination of the effective interfacial area has still to be clarified. Additionally, the sampling procedure in the two-phase region requires further investigation. Therefore, the aim of this work is to check the guideline experimentally and develop it further. The overall goal is the establishment of a generally valid method, which ensures comparable results even if obtained at different test facilities.

Experimental procedure and calculations

The experiments were performed using the pilot-scale test facility at Montanuniversität Leoben. This test facility made of polypropylene consists of a DN 450 measurement column and a DN 600 saturation column. A scheme of the test facility can be taken from Fig. 1, a detailed description from Wolf et al. [9]. However, in this work, the gas volume flow was determined by ultrasonic measurement (Flexim, Fluxus G72) and the gas-phase analysis by non-dispersive infrared (NDIR) spectrometers (ABB, Uras26, measuring ranges: CO₂ 0-2000 ppm, SO₂ 0-2000 ppm and 0-5000 ppm, respectively) with upstream gas conditioning (filtration, cooling and condensate removal). Additionally, the gas temperature in the connecting duct of the columns was measured. The trough-type liquid distributor had a density of 128 dripping points per square meter and an operation range of 10-60 m/h.

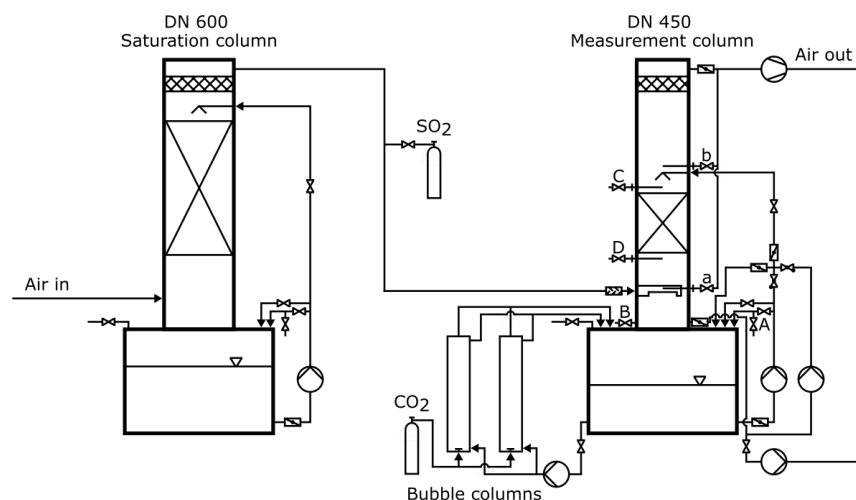


Fig. 1 Scheme of the test facility consisting of a DN 450 measurement column and a DN 600 saturation column;
A, B, C, D: liquid sampling; a, b: gas sampling

The packing Raschig Super-Pak 250Y was chosen for the investigations. Hydraulic characterization and determination of the effective interfacial area were performed with seven packing elements and two initial distribution elements, which were also obtained from Raschig. This resulted in a total packed height of 1.845 m. Further, the gas- and liquid-side volumetric mass-transfer coefficients were determined with two packing elements and without initial distribution elements. The gas phase was sampled continuously in the gas distributor and a few centimeters above the liquid distributor, whereby heated sampling tubes were used. Samples of the liquid phase were taken from the feeding pipe. The liquid load u_L and the gas velocity u_G were calculated by dividing the liquid and gas volume flows through the cross-sectional area of the column, respectively. However, for the hydraulic evaluation the F-factor was used, which is defined as the gas velocity multiplied by the square root of the gas density. Since the gas volume flow was measured downstream the blower, it was corrected for the pressure in the column via the ideal gas law. The ideal gas law was also used for calculation of the gas density, whereby the air leaving the saturation column was assumed to be completely saturated. The saturation pressure was estimated using the Antoine equation with the constants 18.30, 3816.44 and -46.13 (mmHg and K) [10]. The pressure drop was determined without the storage tank below the column using an air-water-system. It was normalized to the distance between the pressure sensors.

The mass-transfer parameters were evaluated using the two-film model and the HTU/NTU modeling. Hereby, three absorption systems were used for the determination of the effective interfacial area and the gas- and liquid-side volumetric mass-transfer coefficients. The effective interfacial area a_{eff} was determined by absorption of atmospheric CO₂ from air into an aqueous sodium hydroxide solution (0.11 mol/L to 0.20 mol/L). Within this absorption system, a rapid irreversible reaction of pseudo-first order takes place. Therefore, the CO₂ concentration in the liquid phase can be neglected [6]. Liquid-phase analysis was performed by titration (Metrohm, 736 GP Titrino) with 0.06 mol/L hydrochloric acid, gas-phase analysis by the NDIR spectrometer. The effective interfacial area was calculated according to the VDI guideline [6]

$$a_{\text{eff}} = \frac{He_{\text{CO}_2}}{\sqrt{D_{\text{CO}_2,\text{L}}k_{\text{OH}^-}c_{\text{OH}^-}}} \frac{u_{\text{G}}}{HRT} \ln \frac{c_{\text{CO}_2,\text{G}}^{\text{in}}}{c_{\text{CO}_2,\text{G}}^{\text{out}}}, \quad (1)$$

whereby the parameters suggested by Tsai [11] were used. Hereby, H corresponds to the height of the packed bed, R to the ideal gas constant, T to the temperature (average of gas in the connecting duct and head of the column and liquid with the latter weighted more heavily similarly to Tsai [11]) and $c_{\text{CO}_2,\text{G}}$ to the CO_2 concentration in the gas phase. The diffusion coefficient $D_{\text{CO}_2,\text{L}}$ [12] and the Henry's constant He_{CO_2} [12, 13] were evaluated using the correlations of Barrett. For evaluation of the latter, the electrolyte compounds NaOH and Na_2CO_3 were taken into consideration, whereby the contributions for Na^+ , OH^- and CO_3^{2-} were taken as 0.091 L/mol, 0.066 L/mol and 0.021 L/mol, respectively. The contribution of CO_2 was determined using a correlation based on the values of Barrett [12, 13]. The correlations of Barrett were also used by Pohorecki and Moniuk [14]. Further, the correlations of Pohorecki and Moniuk were used for evaluation of the reaction rate constant k_{OH^-} [14], as well as the viscosity of the caustic solution [15]. For the viscosity of water the correlation proposed by Tsai [11] was used, which was based on data from the National Institute of Standards and Technology [16]. Since the influence of the gas-side mass-transfer coefficient k_{G} on the effective interfacial area is small [6], it was neglected.

The gas-side volumetric mass-transfer coefficient $k_{\text{G}}a_{\text{eff}}$ was determined by absorption of SO_2 from air into an aqueous sodium hydroxide solution (0.34 mol/L). Therefore, SO_2 was fed into the gas duct between the saturation column and the measurement column. The SO_2 concentrations in the gas phase were measured by the NDIR spectrometers. $k_{\text{G}}a_{\text{eff}}$ could then be evaluated according

$$k_{\text{G}}a_{\text{eff}} = \frac{u_{\text{G}}}{H} \ln \frac{c_{\text{SO}_2,\text{G}}^{\text{in}}}{c_{\text{SO}_2,\text{G}}^{\text{out}}}, \quad (2)$$

whereby the mass-transfer resistance was assumed to be solely in the gas phase [6].

For evaluation of the liquid-side volumetric mass-transfer coefficient $k_{\text{L}}a_{\text{eff}}$, CO_2 was desorbed from water into air. The mass-transfer resistance in the gas phase of this system was assumed to be negligible and $k_{\text{L}}a_{\text{eff}}$ was calculated via

$$k_{\text{L}}a_{\text{eff}} = \frac{u_{\text{L}}}{H} \ln \frac{c_{\text{CO}_2,\text{L}}^{\text{in}}}{c_{\text{CO}_2,\text{L}}^{\text{out}}}, \quad (3)$$

using the CO_2 concentrations in the liquid phase [6]. In preparation for the measurements, deionized water was enriched with CO_2 via circulation between the column's sump and two bubble columns connected in parallel. The measurements itself were the only ones which were conducted in an open-loop instead of a closed-loop configuration. Thus, liquid samples had to be taken also from the sump and not only from the feeding pipe. At higher liquid loads, a pump was used to support the liquid flowing out of the sump and ensure unaltered samples. Additionally, liquid samples were taken a few centimeters above and below the packing in the two-phase region. For this, two identical sampling devices were used, see Fig. 2. These sampling devices were similar to those Wolf et al. [9] used, but had different dimensions. After the sampling, sodium hydroxide was added in excess to all samples to hinder CO_2 from degassing. However, since visually degassing occurred during the sampling, the measurements were performed a second time to check for reproducibility, whereby it was sampled only from the two-phase region.

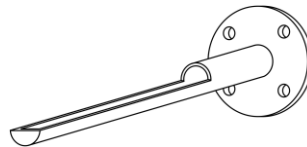


Fig. 2 Scheme of the liquid-phase sampling device.

Evaluation of the total CO_2 concentration in the respective liquid samples was performed by degassing CO_2 and analysis by the NDIR spectrometer. Therefore, approximately 5 mL samples were weighed and transferred by a syringe with needle into a gas-washing bottle filled with diluted phosphoric acid. The CO_2 was transported to the NDIR spectrometer by N_2 , which was added via a gas cylinder and a pressure regulator. To ensure small bubbles, a

frit was used. The ratio of the detected peaks corresponded to the ratio of the CO_2 concentrations, whereby the CO_2 loss while opening the gas-washing bottle was neglected. The method is described in detail in the dissertation of Hilliard [17]. However, contrary to this method, no drying beds were used and not the whole gas stream was analyzed, as a gas-washing bottle and a t-piece were used. Further, different sample amounts were analyzed.

Results and Discussion

Fig. 3 and Fig. 4 show the results of the pressure drop measurements in comparison with data from Vysoká škola chemicko-technologická v Praze (English abbreviation UCT) [18, 19] and Separations Research Program (abbreviation SRP) at the University of Texas at Austin [20], respectively. It can be seen that the dry and irrigated pressure drops are in accordance with the data of SRP and also with the data of UCT at low F-factors.

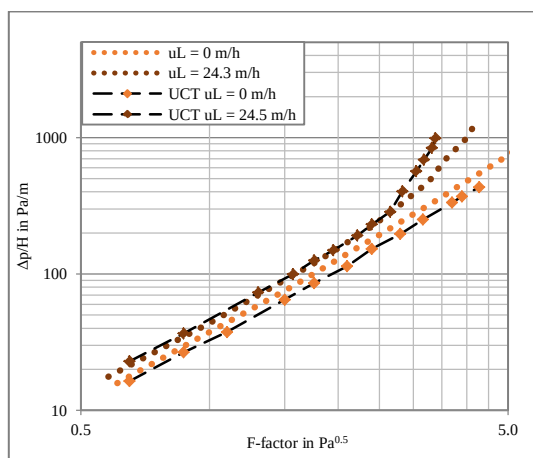


Fig. 3 Specific pressure drop as function of the F-factor; comparison with data of UCT [18, 19].

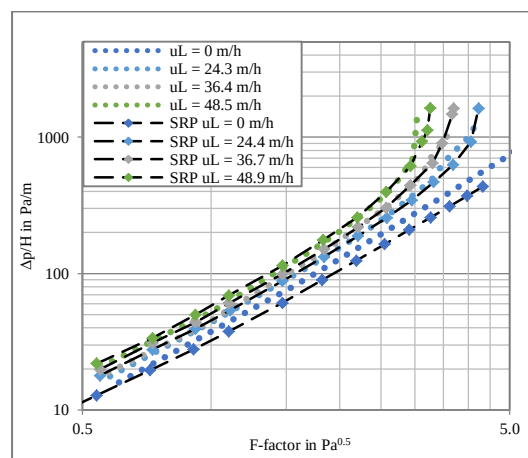


Fig. 4 Specific pressure drop as function of the F-factor; comparison with data of SRP [20].

The preliminary results of the effective interfacial area at a gas velocity of 0.94 m/s indicate a non-negligible influence of the sodium hydroxide concentration, as shown in Fig. 5. An increase of the sodium hydroxide concentration from 0.11 mol/L to 0.20 mol/L resulted in an average reduction of the determined effective interfacial area of approximately 11 % at different liquid loads. However, as can be seen in Fig. 6, the effective interfacial areas are comparable to those obtained by UCT [18, 19] (average relative deviation of 8 %) and SRP [20] (average relative deviation of 7 %).

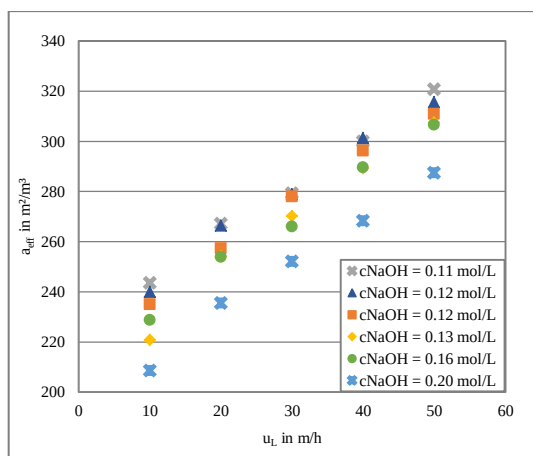


Fig. 5 Effective interfacial area as function of the liquid load; gas velocity of 0.94 m/s; influence of the sodium hydroxide concentration.

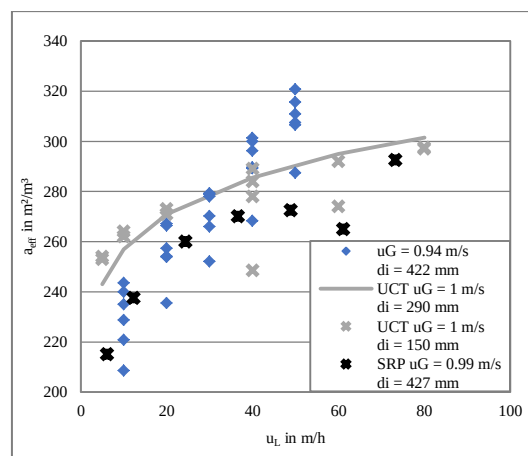


Fig. 6 Effective interfacial area as function of the liquid load; comparison with data of UCT [18, 19] and SRP [20].

For determination of the $k_G a_{\text{eff}}$ values, an inlet SO_2 concentration of approximately 1000 ppm was used, except for the gas velocity of 0.6 m/s, as the SO_2 could not be detected in the output gas stream in the ppm range. Thus, an inlet SO_2 concentration of approximately 2000 ppm was used. As can be seen in Fig. 7, the $k_G a_{\text{eff}}$ values are in accordance with the data from UCT [18, 19] (average relative deviation of 9 %) and the data from SRP [20] (average relative deviation of 15 %). However, further increase of the gas velocity resulted in flooding.

The $k_L a_{\text{eff}}$ values based on the samples of the feeding pipe and sump differ significantly from those based on the samples of the two-phase region with an average relative deviation of about 100 %, as can be seen in Fig. 8. This is an indication that not negligible degassing occurred outside the packing, which resulted in an overestimation of $k_L a_{\text{eff}}$. The average relative deviation between the $k_L a_{\text{eff}}$ values obtained from the samples in the two-phase region was 13 %. UCT and SRP used different absorption systems for the $k_L a_{\text{eff}}$ determination. UCT desorbed oxygen from water into nitrogen [18, 19], whereas SRP desorbed toluene from water into air [20]. Thus, similarly to Rejl et al. [18], the published $k_L a_{\text{eff}}$ of the component i (O_2 or toluene) were correlated to the CO_2 - H_2O system via

$$(k_L a_{\text{eff}})_{\text{CO}_2} = (k_L a_{\text{eff}})_i \left(\frac{D_{\text{CO}_2, \text{H}_2\text{O}}}{D_{i, \text{H}_2\text{O}}} \right)^{0.5}, \quad (4)$$

whereby the relation of the penetration theory was used [21]. The diffusion coefficients for CO_2 , O_2 and toluene in water at 20°C were taken as $1.67 \cdot 10^{-5} \text{ cm}^2/\text{s}$, $2.01 \cdot 10^{-5} \text{ cm}^2/\text{s}$ and $0.85 \cdot 10^{-5} \text{ cm}^2/\text{s}$, respectively [22]. The resulting $k_L a_{\text{eff}}$ are shown in Fig. 8. The average relative deviation from the data of UCT was 14 % and from SRP 30 %.

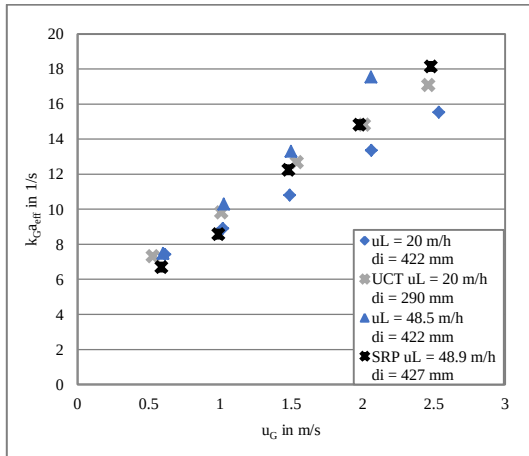


Fig. 7 Gas-side volumetric mass-transfer coefficient as function of the gas velocity; comparison with data of UCT [18, 19] and SRP [20].

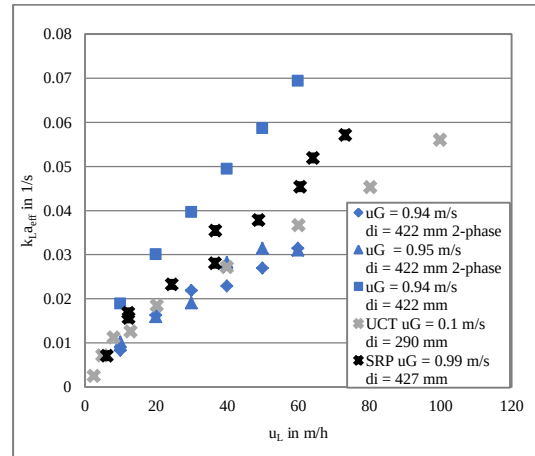


Fig. 8 Liquid-side volumetric mass-transfer coefficient as function of the liquid load; 2-phase indicates sampling in the two-phase region; comparison with data of UCT [18, 19] and SRP [20].

Conclusion

Hydraulic and mass-transfer characteristics of the packing Raschig Super-Pak 250Y were determined using the pilot-scale test facility at Montanuniversität Leoben. Preliminary results of the specific pressure drop, the effective interfacial area and the gas- and liquid-side volumetric mass-transfer coefficients proved to be comparable with data of UCT [18, 19] and SRP [20]. Average relative deviations from available data were calculated for the results obtained at the closest gas and liquid loads. Thus, it is indicated that method standardization for determination of hydraulic and mass-transfer characteristics of packed columns has well advanced.

However, with the sodium hydroxide concentration and the sampling method, two influences have been determined, which require further investigation. Therefore, not all specifications given in the VDI guideline [6] appear to have a sufficient level of detail, which can lead to discrepancies in experimental data. It is suggested that the key for transparent determination of hydraulic and mass-transfer characteristics of packing is an extremely high level of detail, which will be required subsequently for the establishment of a generally valid method. Thus, the need for particularly detailed documentation including the test facility, preparations, measurement procedure, analytical methods and data evaluation is emphasized.

Outlook

In a next step, more measurements will be performed and the procedures refined. Then, those parameters and procedures, which are not adequately described yet, will be systematically investigated. Therefore, the influence of the sodium hydroxide concentration on the determination of the effective interfacial area will be examined in detail first. In this context, also the influence of the sodium carbonate concentration will be investigated. The second parameter, which will be examined in detail, is the packing height, especially when determining the gas- and liquid-side volumetric mass-transfer coefficients. In the preliminary experiments, two packing elements were used for $k_G a_{\text{eff}}$ and $k_L a_{\text{eff}}$ determination. However, to minimize the influence of end effects, it is desired to use higher packing heights [6]. As shown in Fig. 8, also the sampling method can have a significant impact on the experimental result. Therefore, detailed investigations will be performed to optimize the sampling locations and procedures. In this regard, also the evaluation of mass-transfer measurements with data obtained from the two-phase region will be investigated. The above discussed examinations are expected to give a valuable contribution to the establishment of a generally valid method for the determination of hydraulic and mass-transfer characteristics of packed columns.

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