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HEAT TRANSFER IN BUBBLE COLUMNS

by

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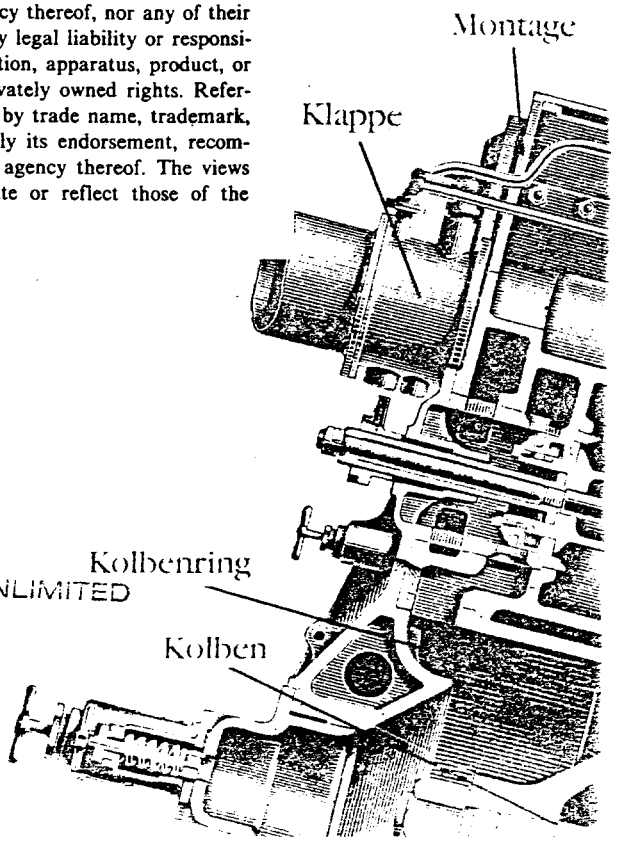

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HEAT TRANSFER IN BUBBLE COLUMNS

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Sufficient dimensioning of cooling and heating surfaces in bubble column reactors requires knowledge of the heat transfer coefficient under different operating conditions. The heat transfer on pipes that were suspended in bubble columns were investigated as a function of various parameters. The heat transfer is particularly strongly dependent on the gas velocity and the viscosity of the liquid. The results can be described by exponential functions.

Object of the Study and Measurement Principle

Liquid columns carrying a steady bubble stream ("bubble columns") are suitable not only for reactions between gaseous phase and liquid phase, but also for reactions between several liquids or for processing involving heterogeneous catalysis. In the liquid reactions, the rising gas bubbles are used for stirring (pneumatic stirring), and they bring about an intensive, uniform mixing of the liquid in the entire column [1, 2]. In the gaseous-phase reactions with solid catalyst, which are of particular importance in industry, the catalyst is suspended in the solid form in a liquid, and the gas rising through this suspension engages in the reaction during its rise. Such a liquid-phase reactor for Fischer-Tropsch synthesis was described by H. Kölbel and P. Ackermann [3].

Sufficient dimensioning of cooling and heating surfaces in bubble column reactors requires knowledge of the heat transfer coefficient for fixed walls in bubble columns. Studies carried out on cooling and heating pipes with a diameter of 30 mm, which were suspended in bubble columns with a height of ca. 1500 mm and with diameters of 92 and 292 mm, will be reported in this paper. The heat transfer coefficient was always determined as a function of the gas velocities (empty tube velocities up to 35 cm/second). The mean bubble size,

the viscosity and interfacial tension of the liquid, the depth of immersion of the pipes, as well as the height at which the test point is located on the pipe, were varied.

The measurements were carried out according to the principle employed by E. Wicke et al. [4, 5] in fluidized beds, according to which a certain thermal output is generated by a heating cartridge in a metal cylinder, which formed part of the suspended pipe in this case. The heat transfer coefficient α can be calculated from this output Q, the surface area F of the metal wall, as well as the temperature difference ΔT between the cylinder surface and the inside of the column according to the formula

$$\alpha = \frac{Q}{F \Delta T} \qquad (1)$$

ΔT was measured by means of surface thermocouples in the metal cylinder as well as a mobile temperature sensor in the bubble column.

Only one report, that by Z. Novosad [6], was found in the literature on heat transfer on a bubble column with a diameter of 38 mm, where the heat transfer was measured on the entire column wall. This study will be discussed later. Studies on heat transfer in a vertical evaporator pipe [7] and flowing gas-liquid mixtures [8] can be used for comparison to the results obtained in this study. Heat transfer in a bottom-phase reactor was mentioned by H. Kölbel, P. Ackermann and F. Engelhardt [9].

Experimental Setup

The experimental setup is shown in Figure 1. Air was always used as the gas admitted into the column. The air was fed into the bubble column via a Woulf bottle a, a dry gas meter b and a pressure gauge c. The bubble column consisted of an acrylic glass pipe with a length of 2000 mm. A gas distribution chamber, which was defined in the upward direction by interchangeable,

porous distribution plates d made of ceramic material, as is available from the company of Schumacher*, Bietigheim, under the tradename Aerolith, was located under the column. For the design of the columns, see previous studies [10].

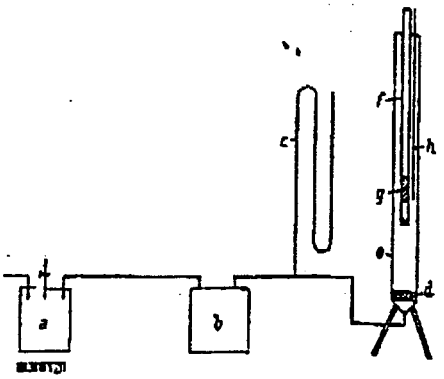

Figure 1. Schematic representation of the experimental setup. a - compensating vessel, b - gas meter, c - pressure gauge, d - distribution plate, e - column, f - acrylic glass pipe with a diameter of 30 mm, g - heating and measuring cylinder, h - temperature sensor.

A pipe f with a diameter of 30 mm was suspended in the column; the pipe screwed together from its parts, i.e., the heating and measuring cylinder g and pipe sections of varying lengths, and its bottom was closed with a hemispherical acrylic glass cap. It was possible to vary the depth of immersion of the pipe in the column freely, and it was also possible to vary the space between the measuring cylinder and the lower end of the pipe by using lower pipe sections of various lengths.

According to Figure 2, the measuring section consisted of a copper cylinder a with a height of 100 mm (excluding the threaded parts), an external diameter of 30 mm and an internal diameter of 18 mm. It had three holes e staggered at 120°C relative to one another and located at 5, 50 and 95 mm from the lower edge. Insulated constantan wires 1, which were soft-soldered with

* We wish to express our thanks to the company of Schumacher for providing the plates.

copper to the surface, were pulled through the said holes; the wires were pulled up through grooves in the inner wall of the cylinder. In addition to these constantan wires, a copper wire soldered to the cylinder was led to the electrical measuring device. The heating cartridge located inside the cylinder consisted of one copper cylinder with a diameter of 16 mm and a length of 100 mm, around which was wound a heating resistor wire d with a resistance of 80 Ω (mica insulation). The winding was corrected until the deviations between the temperatures indicated by the individual surface test points remained below 5% of the temperature difference ΔT against the inside of the column. 220-V direct current was used for heating; a series resistor made it possible to set thermal outputs between 50 and 460 W.

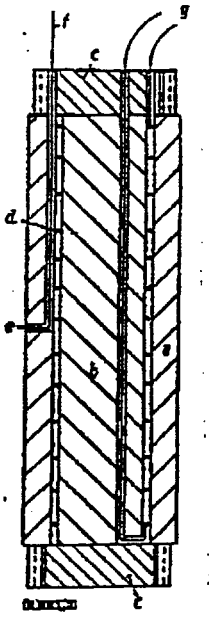

Figure 2. Section through the heating and measuring cylinder.
a - copper cylinder, b - copper heating cartridge,
c - plastic caps, d - heating coil, e - hole,
f - thermocouple wires, g - heating current lead-in.

A copper-constantan couple, which was located at the lower end of a brass pipe (h in Figure 1) and had an external diameter of 4 mm, was used to measure

the temperatures; the pipe was bent such that it was possible to measure the entire radial temperature field in the column by rotating the pipe.

The electrical circuit used to measure the thermal voltages corresponded to that of a Feussner compensator with auxiliary current setting. It was possible to turn on the three surface test points as well as the measuring probe alternatingly with a mercury switch.

Determination of the Heat Transfer Coefficient - Scope of the Experiment

After coarse adjustment of the gas velocity with the stopcock of the compressed air line and fine adjustment with the bleed cock of the Woulf bottle for the first gas velocity of one measurement series, the throughput was determined on the dry gas meter; it was converted into the pressure prevalent at half height of the column. The heating current was subsequently applied and adjusted. A stationary temperature field became established on the measuring cylinder after ca. 15 minutes, and it was possible to determine the surface temperatures, as well as the temperatures in the bubble column at different distances from the cylinder wall. It was necessary to perform the measurements within the shortest time possible, since the temperature in the bubble column was steadily rising slightly due to the heating and mixing of the liquid.

Further measurements of one measurement series, carried out at gas velocities varied in increments of one cm/second, were performed after a waiting period of ca. five minutes after setting the gas velocity. The measuring cylinder was wiped with a rubber wiper before each measurement to prevent the measuring results from being distorted by stagnant air bubbles on the measuring cylinder.

The measurements were evaluated according to Equation (1). Q was obtained from the heating voltage and the heating current, F was given and ΔT followed from the surface temperature averaged from the three test point temperatures

and the spatially constant column inside temperature. The heating current was set such that ΔT was between 10°C and 15°C, and ΔT rose to 20°C-40°C only during the measurements carried out on highly viscous solutions (low heat transfer coefficients). Preliminary experiments failed to reveal the influence of the heat flux on the heat transfer coefficient under otherwise identical conditions.

The gas velocity (empty tube velocities) was varied between 1 and 10 cm/second, and measurements were carried out in one case at 34 cm/second as well. The influence of viscosity was investigated on cane sugar solutions with viscosities up to ca. 1000 centistokes, and the effect of the interfacial tension was investigated with aqueous solutions of potassium ethyl xanthate with a surface tension of 30 dynes/cm. The mean ideal* bubble diameter at the test point was between 1.6 and 2.4 mm in the different columns under otherwise identical conditions. These experiments were carried out at an equal stagnant liquid level of 1400 mm in the column. The measuring height, i.e., the distance between the lower edge of the measuring cylinder and the distribution bottom was 200, 700 or 1200 mm. An acrylic glass cap with a length of 40 mm was located below the measuring cylinder. Moreover, by inserting inserts of varying lengths, the distance between the lower edge of the measuring cylinder and the acrylic glass cap was extended by 48 or 460 mm. The bubble columns investigated had internal diameters of 92, 192 and 292 mm.

Interpretation of the Measuring Results

Figure 3 shows an example for the radial temperatures in the column for different gas velocities beginning from the wall of the measuring cylinder. A great temperature reduction at the wall and constancy of the temperature inside

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* The ideal diameter is defined as the diameter of a sphere of equal volumn.

the column up to a few mm from the wall are seen in all cases. Since similar profiles were obtained for all the investigated conditions, it was not difficult to determine the inside temperatures in the column which are required for calculating the heat transfer coefficient according to Equation (1).

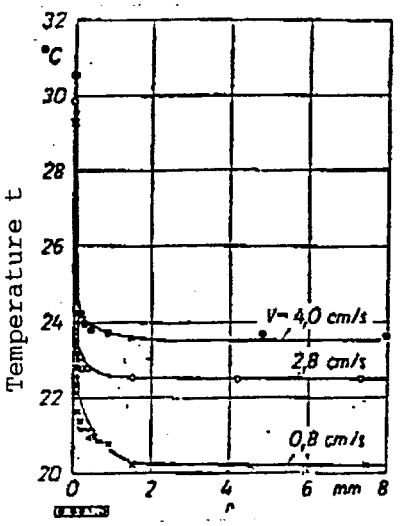

Figure 3. Temperature t in the column as a function of the distance r from the wall of the measuring cylinder at a heating surface load of 30,000 kcal/m^{2}h for different gas velocities V through the column. 192-mm column with air bubbles in water. Measuring height 700 mm.

The constancy of temperature inside the column can be attributed to the intensive mixing of the liquid by the rising gas bubbles. According to Siemes and Weiss [2], the mixing coefficient in an air-water bubble column is higher by ca. five orders of magnitude than the self-diffusion coefficient of water even at a low gas velocity of ca. one cm/second, and it is higher by ca. seven orders of magnitude at a velocity of 10 cm/second. It is easy to estimate that this intensive mixing must practically lead to perfect equality of the temperature inside the column, which has been known from industrial applications [3].

The rapid temperature drop near the wall can certainly be attributed to a boundary layer adhering to the wall, in which the heat is mainly transported by conduction. This layer will apparently not be thicker even in the case of

high gas velocities. If, according to E. Wicke [11], we estimate the thickness Δ of this film to be

$$\delta \sim \lambda/\alpha \qquad (2)$$

in which λ is the thermal conductivity of the liquid at the wall, a boundary layer thickness of ca. 0.2 mm will be obtained for the cases shown in Figure 3, which is apparently in good agreement with the measurements shown in the figure. This confirms the hypothesis on the existence of a boundary layer, since Equation (2) is based on such a hypothesis.

Heat Transfer Coefficient as a Function of Various Parameters

Dependence on the Gas Velocity

Figure 4 shows the heat transfer coefficient α as a function of the velocity V (empty tube velocity) for the 192-mm column (air bubbles in water). A strong increase in a at low V values is followed by a range in which the heat transfer remains almost independent of the velocity; a measurement performed on the 92-mm column revealed constancy of a up to a velocity of 34 cm/second. It follows from this for the wall boundary layer, which is the cause of the heat transfer resistance, that this boundary layer decreases at low velocities to become practically independent of the velocity at velocities of ca. 2 to 3 cm and lower; consequently, this boundary layer is not broken up at these velocities by either the rising bubbles or the movements of the liquid, which are very intensive at higher gas velocities. The great reduction at low velocities can be attributed to the fact that bubbles do not constantly rise at all points of the wall at a short distance from the wall, so that there are always areas in which the liquid is temporarily moving only slightly up to fairly great distances (approximately a few bubble diameters) from the wall. This is also reflected by the α values, which represent mean values determined for the entire measuring cylinder surface. Since the number of rising bubbles is

approximately proportional to the velocity in the range of low velocities, the only slightly moving liquid zones disappear as the velocity rises until gas bubbles eventually rise constantly everywhere at the measuring cylinder up to the shortest possible distance from the wall; this velocity is ca. 2-3 cm per second.

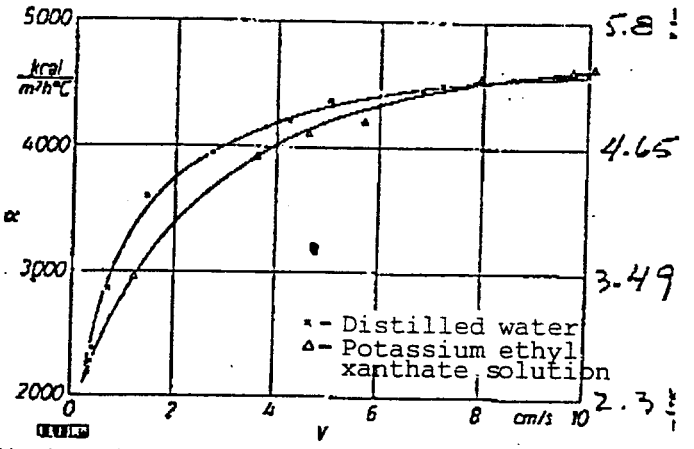

Figure 4. Heat transfer coefficient α as a function of the velocity V for water and an aqueous solution of potassium ethyl xanthate (0.5 g/L). 192-mm column. Measuring height 700 mm.

These circumstances make it understandable that the different hydrodynamic states of the bubble columns for low gas velocities ("laminar range"), on the one hand, and for high velocities ("turbulent range")*, on the other hand, (with a transition between 4 and 5 cm/second) are not reflected by the α values. These states pertain to the inside of the column, the processes that determine the heat transfer should be sought near the wall, and they are apparently influenced by the stream inside the bubble column only slightly.

It is surprising that the α values of bubble columns reach values that were measured during evaporation in a vertical pipe [7, 8]. During evaporation, the boundary layer is constantly broken up and agitated by the vapor

* For a detailed discussion of these states, see previous studies [2, 10, 12].

bubbles located directly at the wall. This agitating effect presumably fails to cause a substantial increase in the α values compared to bubble columns at lower thermal outputs since, according to M. Jakob and W. Fritz [13], bubbles are always formed in a few fixed points, i.e., the agitation is limited to small areas.

Verschoor and Stemerding [8] determined heat transfer coefficients between 2000 and 8000 kcal/m^2h/degree in flowing air/water mixtures in a pipe with a diameter of 14 mm. These figures cannot be directly compared to the values reported here since the experiment was carried out under totally different conditions, but they are on the same order of magnitude.

Influence of Bubble Size, Test Point and Starting Distance

To determine the influence of the bubble size on the heat transfer coefficient, different distribution plates were first used, after which measurements were carried out at different heights of the column with a given distribution plate, since the bubbles coalesce during their rise in pure liquids [10, 12].

Figure 5 shows the measuring results for different distribution plates as well as different immersion depths of the pipe in the 192-mm column. The mean ideal bubble diameters at the height of the test point according to E. Borchers [14] are presented in Figure 5; they are between 1.6 and 2.4 mm. It should be noted in this connection that these values are only valid for the laminar range of gas velocities between 1.5 and 4 cm/second; they are lower below 1.5 cm/second, and there are no well-defined bubbles above 4 cm/second (in the turbulent range), and the phase boundaries change constantly in the liquid-gas mixture. The measured values apparently coincide within the error of measurement, so that the bubble size does not practically influence the heat transfer coefficient α. This is understandable since the mechanism according to which

the individual bubbles within the size range investigated rise and consequently the influence on the boundary layer are the same [9].

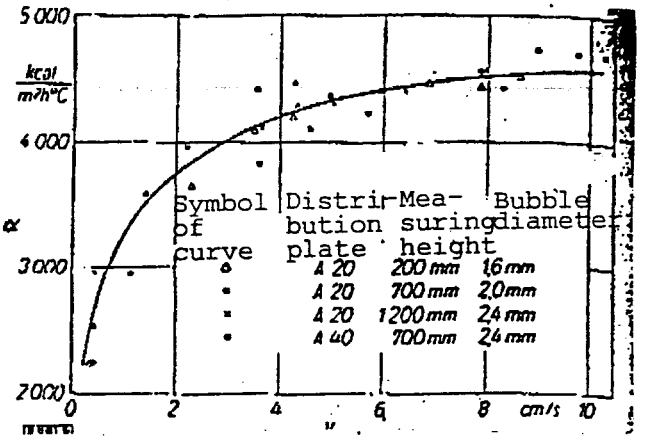

Figure 5. Heat transfer coefficient α as a function of the velocity V for different distributors and measuring heights, corresponding to different bubble sizes. 192-mm column with air bubbles in water.

Measurements performed at constant immersion depth of the pipe but at different distances between the measuring cylinder and the lower end of the pipe also yielded identical α values.

Influence of Surface-Active Additives

Figure 4 shows the heat transfer coefficients obtained for a bubble column with a diameter of 192 mm after adding 0.5 g potassium ethyl xanthate per liter of water, which corresponds to a static interfacial tension of 29.7 dynes/cm in the solution along with the value obtained for distilled water (interfacial tension 71.5 dynes/cm). A slight difference is observed between the α values, but the difference does not exceed 10%. This difference can probably be attributed to the reduction of the rise velocity of the bubbles in the solution due to the formation of a bubble boundary layer with high xanthate content; this reduction, which was conclusively demonstrated by B. Stuke [15], leads to reduced movement of the liquid in the vicinity of the boundary layer. The

difference also almost completely disappears in the range of high velocities, in which individual bubbles with defined boundary layer are no longer formed.

The measurements show no effect of the interfacial tension on the heat transfer in bubble columns (aside from the effect of the interfacial enrichments in the case of solutions).

Dependence on Viscosity

The viscosity of the liquid was varied between 0.85 and 947 centistokes by using cane sugar solutions of various concentrations. Since the temperature rose appreciably due to heating of the measuring cylinder during one measuring series (variation of the gas velocity from high to low values under otherwise identical conditions), the viscosity decreased with decreasing velocities in each measurement series. Figure 6, which contains the results of these measurements, contains the viscosities that correspond to the column temperatures determined immediately before and after the measurement series*. These viscosities were determined in an Ubbelohde viscometer. The curves should be slightly adjusted downward as the velocity decreases so that the highest viscosity is the exact curve parameter for all velocities.

The diagram indicates the strong effect of viscosity on α at low viscosities; the effect becomes weaker beginning from ca. 5 centistokes, but still remains noticeable. This decrease in α with increasing viscosity shows an increase in the thickness of the boundary layer, which is just as understandable as the limited nature of this increase at higher viscosities. It should be borne in mind that the mean bubble size in the column increases considerably with increasing viscosity and the path of the bubbles becomes greatly linear, which facilitates the formation of thicker boundary layers.

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* Previous measurements [12] had given values that were slightly too low.

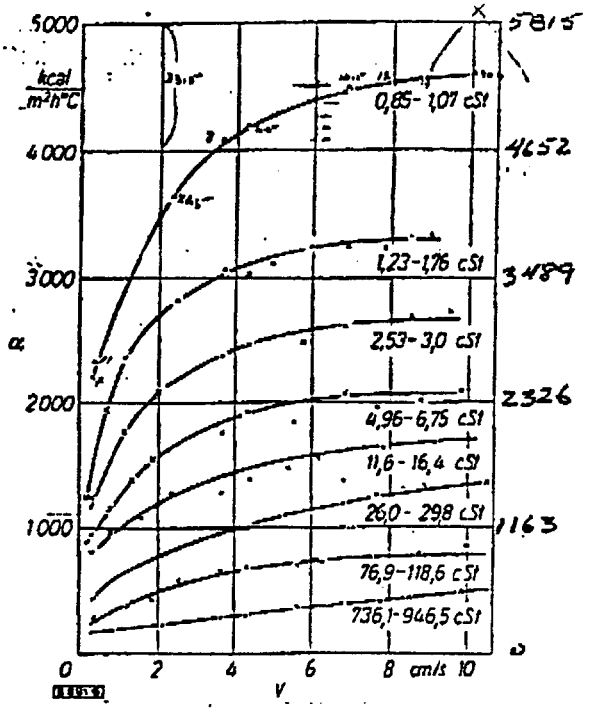

Figure 6. Heat transfer coefficient α as a function of the velocity V for different viscosities (cane sugar solutions). 192-mm column, measuring height 700 mm.

Influence of the Column Diameter

Figure 7 shows measurement series carried out on columns with different diameters, as well as some test points published by Novosad [6] for a column with a diameter of 38 mm, in which the liquid was passed through at low velocities (below 1.2 cm/second) in parallel flow with the gas. All our values are within a rather narrow range, and Novosad's heat transfer coefficients are below these measured values.

The values measured directly in the 92-cm column are somewhat higher than the points shown in Figure 7. It was possible to attribute this to the temperature rise, which is appreciable in this relatively narrow column during the measurements as a consequence of the heating and the resulting decrease in viscosity. Using Equation (5), which will be discussed later, the measured values were converted to the conditions applicable to the wide columns.

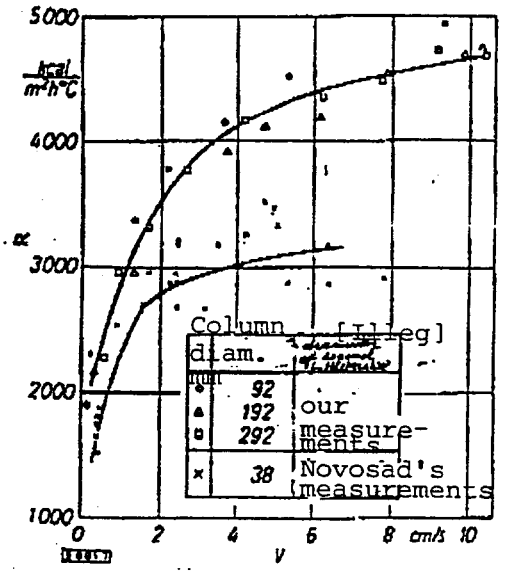

Figure 7. Heat transfer coefficient α as a function of the velocity V for different column diameters D according to our measurements and the measurements of another author (Z. Novosad [6]). Column content: aqueous potassium ethyl xanthate solution (0.5 g/L) in our measurements and water in the measurements performed by the other author. Measuring height 700 mm.

Consequently, the measurements show that the heat transfer is independent of the column diameter if the ratio of the column diameter to the bubble diameter is above ca. 20.

The fact that the values published by Novosad are too low compared to ours can certainly be attributed in part to the somewhat less sensitive method he used, but these lower values may also reflect the influence of the column diameter in this range, in which the real horizontal bubble diameter can account for more than 1/5 of the column diameter. However, these values are on the same magnitude as ours.

Representation of the Results Using Equations

The measurement results presented above show the dependence of the heat transfer coefficient on the viscosity η (or the kinematic viscosity ν) and the

velocity V, and the heat transfer resistance is located in a velocity-dependent boundary layer. Equation

$$Nu = f\ (Re,\ Pr) \qquad (3)$$

which is often used to describe this dependence and which contains the Nusselt number Nu, the Reynolds number Re and the Prandtl number Pr, was also applied to this case. The velocity V was selected as the characteristic gas velocity, and the material characteristics used were the corresponding values of the pure liquid at the temperature whose mean value is between the surface temperature and the internal temperature of the column (boundary layer mean temperature), and the diameter d of the suspended pipe (f in Figure 1) was selected as the characteristic dimension. Figure 8 shows the Nusselt number

$$Nu = \frac{ad}{\lambda} \qquad (4a)$$

(λ is the thermal conductivity of the liquid) as a function of the Reynolds number

$$Re = \frac{Vd}{\nu} \qquad (4b)$$

for the measurement series shown in Figure 6, and the Prandtl number

$$Pr = \nu/a \qquad (4c)$$

(a is the temperature conductivity of the liquid) being the parameter. Even though Pr was in the range of 5.0-1200 in the measurements recorded, all test points fit one curve, i.e., the Prandtl number does not affect Nu within the error of measurement in this representation, and Equation (3) consequently changes in this case to

$$Nu = f\ (Re) \qquad (3a)$$

As indicated in Figure 8, the curve can be subdivided in the double-logarithmic representation into two almost straight sections, to which correspond to the equations

$$Nu = 43.7\ Re^{0.22} \quad \text{for } Re > 150 \qquad (5a)$$

$$V \leq 10 \text{ cm/second}$$

$$\text{and } Nu = 22.4\ Re^{0.355} \quad \text{for } Re < 150 \qquad (5b)$$

Since α remains constant at velocities above 10 m/second, the α value obtained for this velocity according to Equation (5a) also applies to V > 10 cm/second.

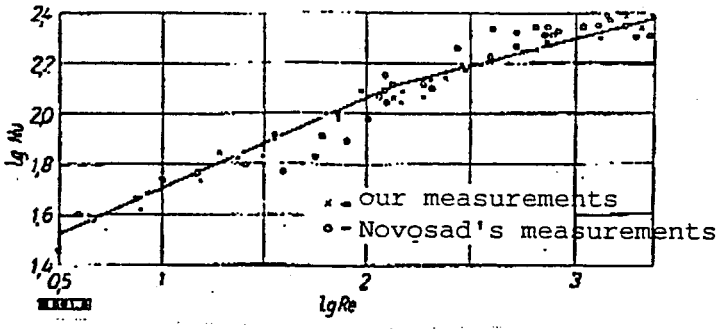

Figure 8. Representation of our measured results and those of another author according to Equation (5).

Equations (5a) and (5b) do not represent strict laws of similarity. For example, the Nusselt number can be expected to depend on the ratio of the bubble diameter to the pipe diameter in ranges of this ratio that are different from those investigated here, and the ratio of the rise velocity of the bubbles to the empty pipe velocity may also play a role. Therefore, it is not permissible to calculate the dependence of the heat transfer on the heating or cooling pipe diameter according to Equations (5a) and (5b); this dependence has yet to be investigated experimentally. Practicable reference values for α can best be obtained even for vertical surfaces other than the wall of 30-mm pipes by retaining the value of 30 mm for the characteristic dimension in these other cases as well.

Figure 8 also shows Novosad's test points, most of which are in agreement with our measurements. The pipe diameter of 38 mm, which is practically almost equal to the heating pipe diameter used in our measurements, was selected as the characteristic dimension.

Novosad tried to describe his measured results by the equation

$$Nu = \frac{Nu_o}{1 + 30\sqrt{z}} + 2{,}28\, Re^{0.7} \cdot Pr^{0.4} \qquad (6)$$

in which Nu_o is the Nusselt number for the heat transfer at zero gas velocity, and z is the relative gas content in the column. Reynolds number is defined here by

$$Re = \frac{D \cdot V}{\nu \sqrt{z}} = \frac{D\, u \sqrt{z}}{\nu} \qquad (7)$$

in which D is the pipe diameter and u is the mean rise velocity of the bubbles. Novosad can describe his results using Equation (6) to an accuracy of ±8%. In contrast, our own measurements show values which deviate from those calculated according to Equation (6) by up to 30%. This is probably due to the different conditions in very narrow and wide columns, which are better taken into account by the simpler Equation (5).

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