# OPTICAL MEASUREMENT METHOD FOR GAS-LIQUID MASS TRANSFER INVESTIGATION IN HELICALLY COILED PIPE

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## ABSTRACT

In many industrial processes the transfer of reactants from the gas to the liquid phase is of essential importance. For these processes several types of mass transfer reactors can be found, such as tubular reactors, within those helically coiled pipes. The present study concentrates on mass transfer behavior in a helical coiled pipe in case of gas-liquid two phase flow system. Oxygen transfer from air bubbles to the liquid phase is experimentally investigated via an optical measurement method. The colored reaction used for this, is known as the 'blue bottle reaction'.

## INTRODUCTION

*Dietrich et al*. [1] visualized gas-liquid mass transfer via a new colorimetric technique, using an oxygen sensitive dye. They used resazurin to indicate the mass transfer around Taylor bubbles in a straight square channel. Present investigation actualizes a similar optical technique in a helical coiled pipe with a different indicator: methylene blue. Helical coiled pipes are widely used in the industry, due to their large number of advantages. Comparing with straight tubular reactors the helical tubular reactors achieve better mixing, mass- and heat transfer properties in addition within compact sizes. The background behind these good features can be attributed to the spiral shape causing centrifugal force which causes secondary flow, or in another appellation Dean-vortices [2].

First of all, to be able to describe the details of the investigation, a short terminology about the helical coiled pipes and two phase flows must be presented. Figure 1 gives a schematic illustration about the helical coiled pipe.
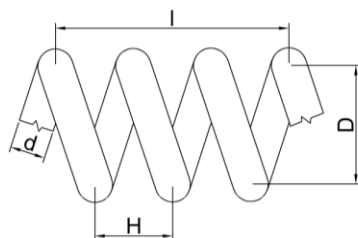


Figure 1
Schematic illustration of helical coiled pipe

In Fig. 1 above $d$ is the inner diameter, $D$ is the coil diameter which is also called pitch circle diameter (*PCD*). $L$ is the total length of the tube and $l$ is the length of the spirally shaped body. $H$ indicate the pitch of the tube, it means the distance

between two adjacent turns. The ratio of inner diameter to coil diameter is called curvature ratio ($\delta$) which can be written as follows [3]:

$$\delta = \frac{d}{D} \ [-] \ .$$
(1)

The flow conditions of this two phase flow system are determined by the following variables. Gas hold-up ($\varepsilon$) is the ratio of the flow rates of gas phase and the total mixture, which can be written as [4]:

$$\varepsilon = \frac{Q_g}{Q_g + Q_l} \ [-] \ .$$
(2)

In equation (2) $Q_g$ is the flow rate of the gas phase and the $Q_l$ is that of the liquid phase. The superficial velocity is the velocity that the gas phase ($v_{sg}$) or the liquid phase ($v_{sl}$) can reach if both of them flow alone in the tube. So the superficial velocity can be calculated from the actual phase flow rate and the cross-sectional area of the tube, as follows [5]:

$$v_{sg} = \frac{4}{d^2\pi} Q_g \ [m/s] \ , \qquad\qquad v_{sl} = \frac{4}{d^2\pi} Q_l \ [m/s] \ .$$
(3)

The mixture passes from the inlet to the outlet during the residence time ($t$). If we assumes that the bubble velocity equal with the mixture velocity, the residence time can be written as:

$$t = \frac{L}{Q_g + Q_l} \frac{d^2\pi}{4} \ [s] \ .$$
(4)

The aim of the current measurements is the following: visualize and quantify mass transfer from oxygen to water in a helically coiled tube in case of various flow conditions. During the measurements sixteen measurement points are investigated. As seen in Figure 2 the investigated points are embedded in the plug flow regime on the flow map, because in this case due to the emerging Taylor-bubbles the mass transfer rate is the highest [6].
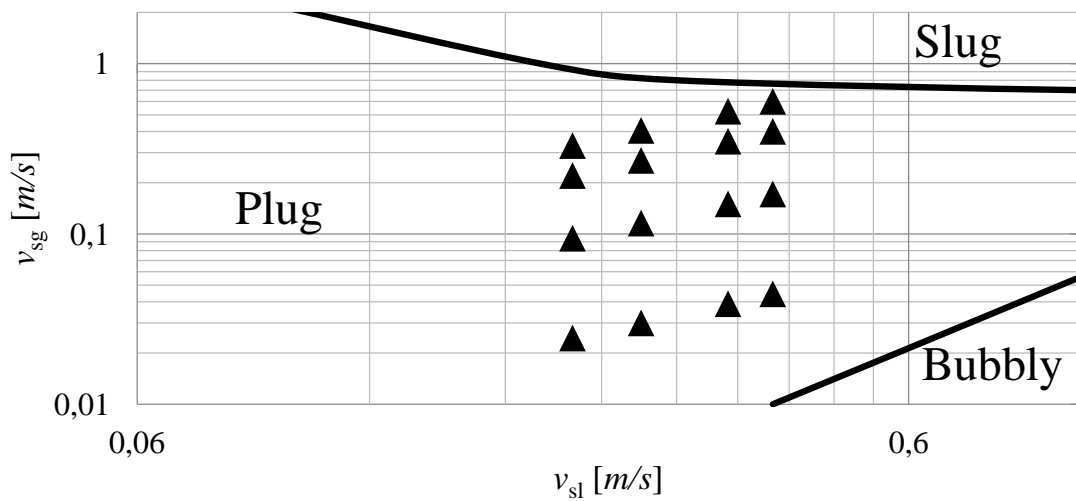


Figure 2
Flow map for the investigated measurement points
(Flow map based on the work of *Murai et al.* [4])

## MEASUREMENT METHOD

The used non-intrusive measurement technique is based on a redox indicator, namely methylene blue. For this experiment methylene blue (10 $mg/l$) is added to a water-based solution of glucose (40 $g/l$) and sodium-hydroxide (5 $g/l$). Glucose in alkaline medium can be oxidized easily and the oxidized methylene blue can be reduced by the glucose and becomes leucomethylene blue, consequently the blue solution becomes colorless. On the other hand, due to the oxidization of leucomethylene blue the colorless solution becomes blue. In our case this oxidation caused color change is utilized for the mass transfer visualization. Figure 3 shows the oxidized and reduced forms of methylene blue.
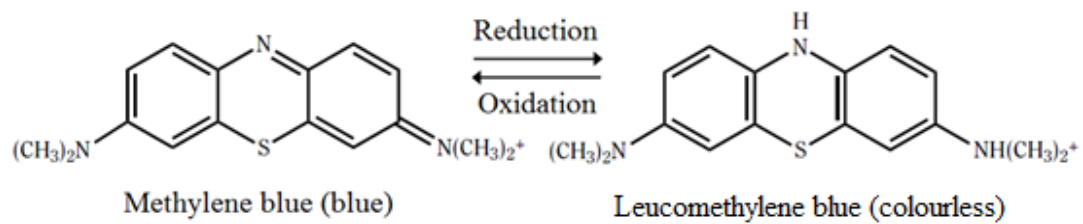


Figure 3
Structure of the oxidized and reduced state of methylene blue

The investigated helical pipe is made by glass and it is placed in a PMMA decagon prism, which is filled up with glycerin. Due to same refractive indices the wall of the glass tube becomes nearly invisible for the camera. The parameters of the helical pipe are the following: $d=6$ $mm$, $D=25$ $mm$, $H=15$ $mm$, $l=210$ $mm$, $L=1318$ $mm$, $\delta=0.24$. During the measurements, the liquid solution is circulated in the helical pipe by a speed controlled gear pump (*Watson Marlow 5003U Micropump*). The gas phase is controlled by a mass flow controller (*Bronkhorst El-flow F-201CV*) and it is added to the liquid phase using a T-junction. Due to the backlight illumination (40$W$ white LED lamp, type: *Strakler EEK*) the dissolved oxygen caused liquid colour change is perceived as a light intensity change by the camera (*Lavision Imager pro HS*) and in this way the mass transfer visualization become possible. This technique gives integrated information about the whole line of sight (depth). For each measurement the camera works with 100 $Hz$ recording frequency and 116 $\mu m$ image resolution. One measurement series can be divided into three parts: (i) 50 pictures about the oxygen-free solution, (ii) 3000 pictures about the measurement point, (iii) 50 pictures about the saturated blue solution. The oxygenation of the liquid (oxidation) and stripping of dissolved oxygen (reduction) are carried out with the mini bubble column reactor and this device ensures the oxygen-free liquid phase during the measurements. In case of (i) and (ii) helium is used to the reduction and oxygen protection, during the oxidation (iii) air is bubbled in the reactor. The schematic drawing of the applied measurement system can be seen in Figure 4.
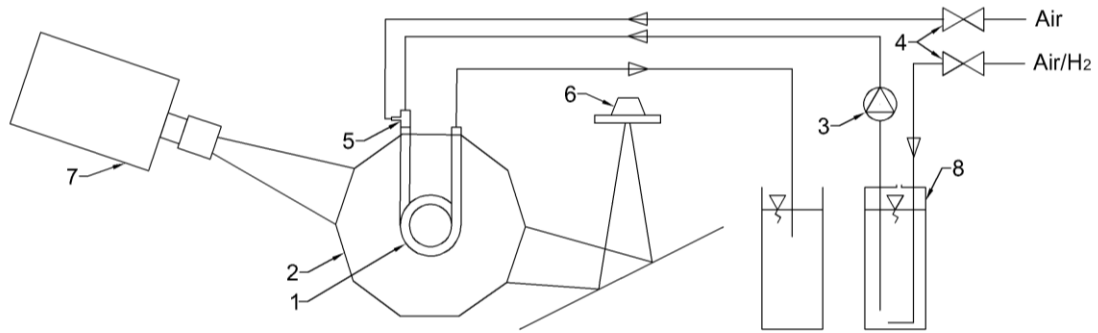
Figure 4
Schematic illustration of the experimental setup with all mentioned subunits:
helical pipe in side view (1), decagon prism (2), pump (3), mass flow controller (4),
T-junction (5), LED lamp (6), HS camera (7), mini bubble column reactor (8)

## PROCESSING METHOD

After the measurements the pictures are processed using Davis software (*LaVision, Göttingen, 1989*). The goals of the picture post-processing are the following: correction of the backlight inhomogeneity, elimination of shadows and reflexions, masking out the helix and bubbles and preparation for further processing. The steps of the image processing in Davis are: *'convert to grayscale', 'sheet correction', 'geometric mask', 'algorithmic mask'* and *'average'*. The results of the measurements are RGB pictures; nevertheless during the additional evaluations greyscale images are more favourable, so in the first step the RGB pictures are converted to grayscale. Using the '*sheet correction*' intensity differences of the backlight can be corrected. For the sheet correction a so-called sheet image is needed, which gives information about the intensity distribution of the backlight. The geometric mask can be successfully used if masking of fix areas is required. For the investigated helical pipe, the applied geometric mask has a dual function: first, eliminate the outer area of the helix, and second, mask out inner parts of the helical pipe with reflexions, shadows and other optical aberrations which influence the results negatively. For algorithmic mask, using an intensity threshold the program can distinguish between valuable areas and unimportant areas. In our case this kind of masking has become necessary because of the moving bubbles. Due to the *'algorithmic mask'* the bubbles are covered and only the intensity change of the liquid phase is visualized. After these operations the *'average'* function makes an average from all images of the series for every pixel.

After the picture processing the visualized mass transfer had to be quantified from the obtained intensity change over the helix. The final average pictures from Davis are exported in '.DAT' format to a MATLAB (*The MathWorks, U.S.A., 1984*) script and the following calculations are carried out using the software package. First of all the intensity of each coil of the helical pipe is averaged. In the interest of the progress of mass transfer from gas to liquid phase can be described. The progress variable (*pv*) is defined as:

$$pv = \frac{I - I_c}{I_b - I_c} \ [-] \ , \tag{5}$$

where $I$ is the actual averaged intensity value, $I_c$ is the intensity of the totally oxygen free solution and $I_b$ is the intensity of the saturated solution. This variable is computed in each turn of the helical pipe. The difference between the progress variable values of the first and the last spiral characterizes the rate of the mass transfer:

$$\Delta pv = pv_{15} - pv_1 \ [-] \ . \tag{6}$$

The progress variable change gives information about the mass transfer rate, but this variable is not independent of the residence time. Because of this, a new variable is created in favor of the better comparison. The integral time ($\tau$) can be calculated by the ratio of the progress variable change ($\Delta pv$) and the residence time ($t$), as follows:

$$\tau = \frac{\Delta pv}{t} \ [1/s] \ . \tag{7}$$

The progress variable and the integral time typify well the characteristic and the speed of mass transfer in the various flow conditions. Nevertheless, without calibration these parameters do not give numerical values about the amount of dissolved oxygen content. For the calibration we relied on the study of *Dietrich et al.* [1]. The base of the calibration is the following: methylene blue solutions with different concentrations ($n_{methylene \ blue}$) are saturated and circulated through the helical coiled pipe. Due to the described measurement and processing method in case of every concentration a progress variable ($pv$) value is created. Using stoichiometry of the reaction between oxygen and leucomethylene blue the number of moles of dissolved oxygen ($n_{O2 \ reacted}$) can be deduced easily from the number of moles of leucomethylene blue ($n_{leucomethylene \ blue}$). This can be written in the following formula [7]:

$$n_{O_2 \ reacted} = \frac{n_{methylene \ blue}}{2} = \frac{n_{leucomethylene \ blue}}{2} \ [mol] \ . \tag{8}$$

Due to this method, the connection between the dissolved oxygen concentration ($C$) and the progress variable ($pv$) is specified as seen in Figure 5.
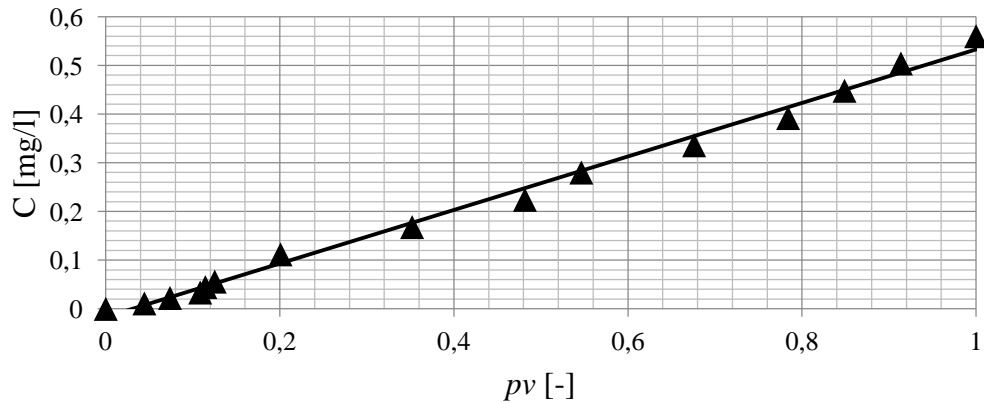


Figure 5
Calibration curve

**RESULTS**

The progress variable change (*pv*) and the integral time ($\tau$) in case of every investigated flow conditions typify the effectiveness and speed of the mass transfer. Since the calibration curve is linear, the characteristic of the dissolved oxygen concentration (*C*) and the dissolved oxygen concentration per unit time ($\dot{C}$) is proportional like the progress variable change (*pv*) and the integral time ($\tau$). Due to this consideration in this paper just the *C* and $\dot{C}$ results are plotted. These plots determine the mass transfer characteristic as well as the *pv* and $\tau$ plots and additionally give data about the real dissolved oxygen concentrations. Figure 6 shows the dissolved oxygen concentration (*C*) as a function of the gas hold-up ($\varepsilon$) and the superficial liquid velocity ($v_{sl}$).
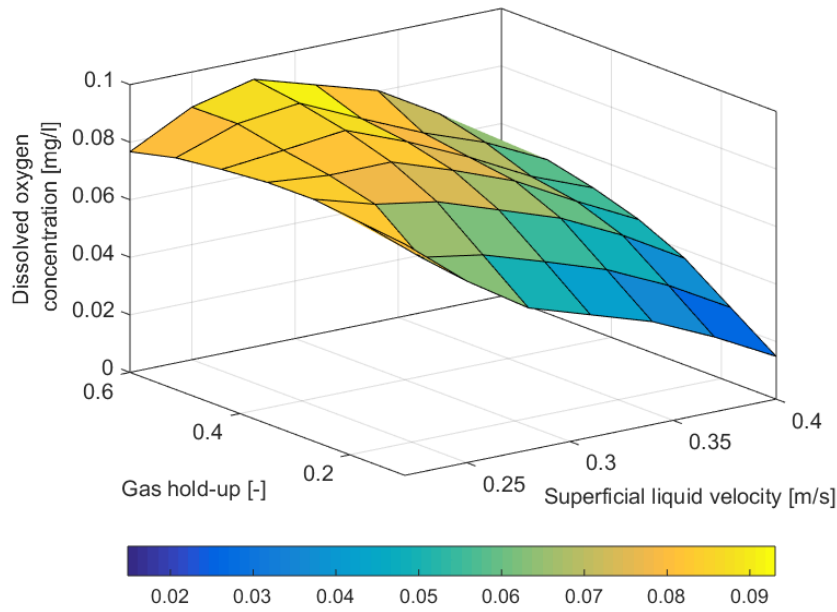


Figure 6
Dissolved oxygen concentration vs. gas hold-up and superficial liquid velocity

It can be seen in Figure 6 that two types of tendencies can be observed. Firstly, the gas hold-up increases leads to the growth of the dissolved oxygen concentration. (The $v_{sl}$=0.22 *m/s* condition is an exception where besides the gas hold-up change the value of the progress variable change stagnates.) Secondly, in case of $\varepsilon$=0.1 and $\varepsilon$=0.3 the superficial liquid velocity increase leads to higher oxygen mass transfer rate. On the other hand, in case of $\varepsilon$=0.5 and $\varepsilon$=0.6 the effect of superficial liquid velocity change are different: although the superficial liquid velocity reduction leads to the dissolved oxygen concentration growth but this tendency reaches the maximum at $v_{sl}$=0.27 *m/s* and then decreases again. The highest dissolved oxygen concentration can be observed in case $v_{sl}$=0.27 *m/s*, $\varepsilon$=0.6 it is 0.0952 *mg/l*, and the lowest dissolved oxygen concentration can be seen at $v_{sl}$=0.4 *m/s*, $\varepsilon$=0.1, it is 0.0148 *mg/l*. Smaller superficial velocity means higher residence time (*t*) so the connection time between the phases is higher as well. Thus, it follows that the higher oxygen dissolution is maybe just caused by the longest connection time between the phases and not because of the increased mass transfer efficiency.

The residence time independent examination is realized with the help of dissolved oxygen concentration per unit time ($\dot{C}$), as seen in Figure 7.
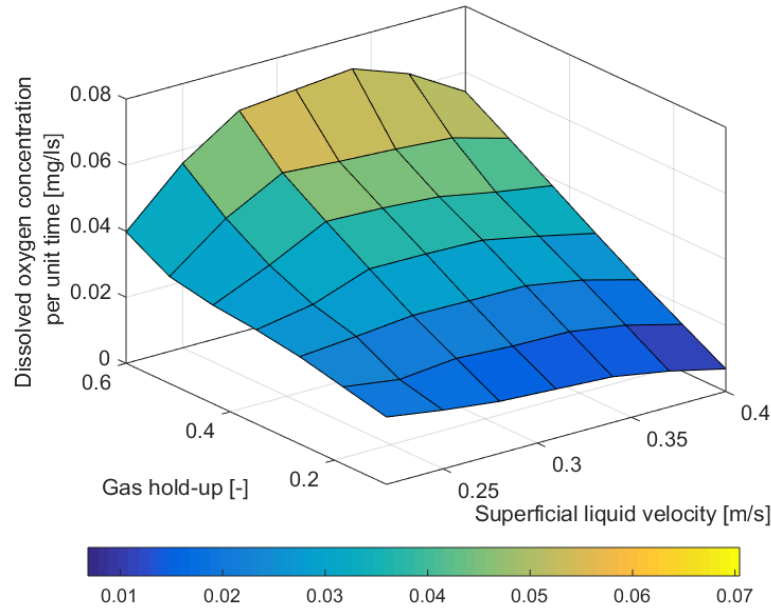


Figure 7

Dissolved oxygen concentration per unit time as a function of the gas hold-up and superficial liquid velocity

In case of Figure 7 the gas hold-up growth leads to the mass transfer efficiency increases as before, but the effect of the superficial liquid velocity is changed. Clear trend cannot be detected, only in case of $v_{sl}$=0.22 $m/s$ condition can be seen significant decrease in the dissolved oxygen concentration per unit time. The highest dissolved oxygen concentration per unit time can be observed in case of $v_{sl}$=0.35 $m/s$, $\varepsilon$=0.6 it is 0.071 $mg/ls$, and the lowest dissolved oxygen concentration per unit time can be seen in case of $v_{sl}$=0.4 $m/s$, $\varepsilon$=0.1 it is 0.0068 $mg/ls$.

**CONCLUSIONS**

During this research the mass transfer behaviour is experimentally studied in a glass helical coiled pipe in case of gas-liquid two phase flow. An experimental method which is applicable in the helically coiled pipe for the non-intrusive oxygen-water mass transfer investigation is mounted. Using the measurement system the mass transfer behavior in case of different flow conditions is determined. Sixteen flow conditions are investigated in the plug flow range. The main idea is changing of the superficial liquid velocity and the gas hold-up. Based on the results, the following conclusions are drawn:

- The applied optical method is suitable to the investigation of the mass transfer behaviour in the helical coiled pipe.
- Within the investigated range the higher gas hold-up ($\varepsilon$) value causes higher mass transfer rate. This tendency can be observed in C as well as $\dot{C}$.

- The dissolved oxygen content ($C$) is increased when the superficial liquid velocity ($v_{sl}$) is decreased, but this tendency caused by the higher connection time in lower superficial liquid velocity. Therefore, the dissolved oxygen content is higher but the efficiency of the mass transfer is not. Investigating dissolved oxygen concentration per unit time (residence time independent examination) clear tendencies cannot be determined between the superficial liquid velocity and the mass transfer efficiency. For the accurate determination of this relationship additional studies will be needed.

Overall it can be said that with similar researches the mass transfer behaviors in helical flows can be determined. It can support the helical coiled tubular reactors designing and this leads optimized mass transfer processes in the industrial area.

## ACKNOWLEDGEMENT

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