

Diffusion - Determination of Diffusion Constants using the Schlieren Method

Protocol for the PC 2 lab course by
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Abstract: In this experiment, the refractive indices of different salt solutions were measured as $n_{\text{H}_2\text{O}} = 1.3375$, $n_{\text{NaCl}} = 1.3541$, $n_{\text{KCl}} = 1.3514$ and $n_{\text{ZnSO}_4} = 1.3725$. By using the Schlieren method, the diffusion constants of the three salts NaCl, KCl and ZnSO₄ in water were determined as $D_{\text{NaCl}} = 1.414 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $D_{\text{KCl}} = 6.636 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ and $D_{\text{ZnSO}_4} = 2.925 \cdot 10^{-10} \text{ m}^2 \text{ s}^{-1}$.

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1 Theory

Molecular diffusion describes the movement of particles from a region of high concentration to a region of low concentration, which leads to an even distribution of particles.^[1] It was first discovered by Robert Brown, who observed the random movement of tiny particles suspended in a liquid, which is called Brownian motion. Then German physicist Adolf Fick laid the theoretical foundation for diffusion by formulating two laws.

The first Fick's law describes how the particle flux density $j(z)$ is connected to the Diffusion coefficient D and the concentration gradient $\frac{\partial c(z)}{\partial z}$, which is shown in Equation 1.

$$j(z) = -D \cdot \frac{\partial c}{\partial z} \quad (1)$$

The physical meaning of the first Fick's law is that a concentration gradient leads to mutual diffusion of particles, leading to an even distribution. The diffusion coefficient can also be defined by the „Einstein relation” in Equation 2.

$$D = \frac{1}{2} \nu \Delta z^2 \quad (2)$$

ν is the jump rate and Δz^2 the mean square jump distance of particles, assuming that they can only move along the z axis. Second Fick's law describes the temporal course of concentration as shown in Equation 3.

$$\frac{\partial c}{\partial t} = D \cdot \frac{\partial^2 c}{\partial z^2} \quad (3)$$

Equation 3 states that the concentration equalization proceeds faster the stronger the concentration gradient. By integrating Fick's second law from Equation 3 under certain boundary conditions and using the substitution $\mu = \frac{x}{\sqrt{4Dt}}$, Equation 4 for the profile of concentration can be obtained.

$$c = \frac{c_2}{2} \cdot \left(1 - \frac{2}{\sqrt{\pi}} \int_0^\mu e^{-\mu^2} d\nu \right) \quad (4)$$

By deriving Equation 4, the function is simplified into Equation 5, which leads to the concentration gradient profile having the shape of a Gaussian bell curve, as is illustrated in Figure 1.

$$\frac{dc}{dz} = -\frac{c_2}{2\sqrt{\pi Dt}} \cdot e^{-\mu^2} \quad (5)$$

Assuming that the refractive index n is proportional to c and knowing that the gradient has an extreme value at $z = 0$, as can be seen in Figure 1, Equation 6 describes the refractive index gradient at $z = 0$ with the indices of water n_1 and of the salt solution n_2 .^[1]

$$\left(\frac{dn}{dz} \right)_{z=0} = -\frac{n_1 - n_2}{2\sqrt{\pi Dt}} \quad (6)$$

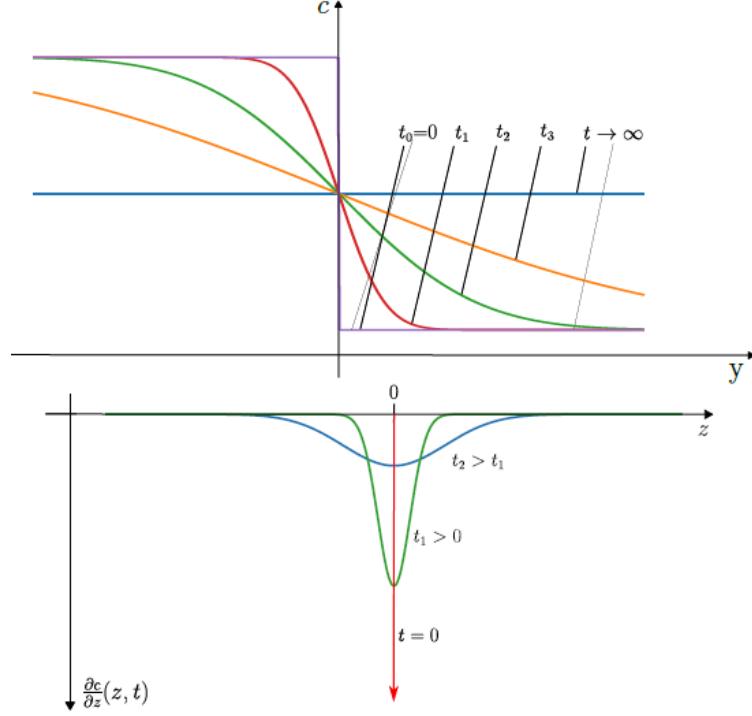


Figure 1: Profiles of the concentration and concentration gradient.^[1]

Following this argumentation, diffusion leads to a continuously changing refractive index profile, which causes a light beam directed to a cuvette of a salt solution with a concentration gradient to be refracted toward the optically denser medium. That is called Snell's law of refraction and shown in Figure 2. The exit angle β und the degree of curvature can be described with Equation 7 and 8, where α is the angle of incidence and r the radius of the curvature of the light beam.^[1]

$$\beta = \frac{n}{n_0} \alpha \quad (7)$$

$$\frac{1}{r} = \frac{1}{n} \frac{dn}{dz} = \frac{d \ln(n)}{dz} \quad (8)$$

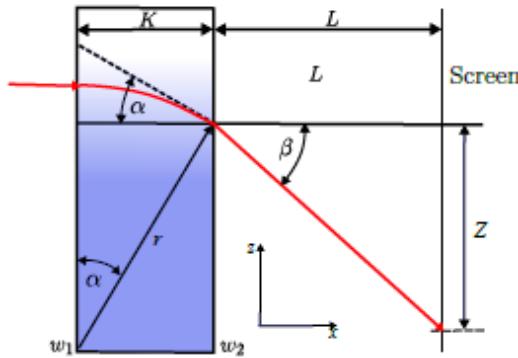


Figure 2: Scheme of the experimental setup geometry.^[1]

The Huygen's principle states that every point of a wavefront can be seen as the origin of a spherical elementary wave. That's why the path of the light beam is curved in a medium with a spatially dependent refractive index $n(z)$ due to a lateral phase shift within the wavefront. The strongest deflection is experienced by the part of the beam that passes through the steepest concentration gradient region. With the small-angle approximation and Equation 7 and 8, the refractive index gradient can be described by Equation 9.

$$\frac{dn}{dz} \approx \frac{Z}{L} \cdot \frac{n_0}{K} \quad (9)$$

L is the distance between the cuvette and the screen, K the thickness of the cuvette, n_0 the refractive index of air and Z the distance between the straight extension of the beam at the exit point and the impact point on the screen. By equating Equation 9 and Equation 6 and rearranging to the diffusion constant D , Equation 10 is derived to calculate D using the Schlieren Method.^[1]

$$D = \frac{(n_1 - n_2)^2 L^2 K^2}{4\pi n_0^2 Z^2 t} \quad (10)$$

2 Procedure

The experimental setup was already built prior to the start of the measurements as shown in Figure 2. The laser beam light path was tilted at about 45°.

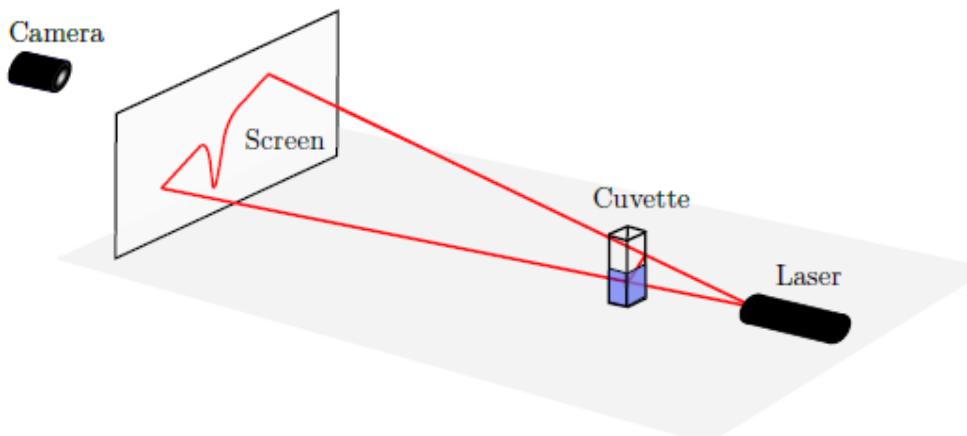


Figure 3: Scheme of the laser beam geometry.^[1]

The experiment began by doing a calibration measurement where the settings of the camera were adjusted, so that the only visible part was the laser beam. Afterwards the cuvette was half filled with deionized water and underlayered with a 2 molar solution of sodium chloride. As soon as the sodium chloride solution was underlayered the video recording was started. After 15 minutes the recording was stopped the resulting video was analyzed by taking snapshots at relevant timestamps.

Those snapshots were uploaded into the python skript "convert-image.py", which converted the snapshots into xy-value pairs. The resulting ".dat" file was then uploaded into the python skript "auswertung diffusion.py" where the amplitude was determined by adjusting slider. These steps were repeated for the 2 molar potassium chloride and zinc sulfate solutions. The potassium chloride solution was also recorded for 15 minutes and the zinc sulfate solution was recorded for 30 minutes. During the recording of the sodium chloride solution the refracting indices of deionized water, and the three solutions were measured using a refractometer.

3 Analysis

3.1 Refractive indices

The refractive indices of deionized water and the different salt solutions were measured with a refractometer and are listed in Table 1.

Table 1: Measured refractive indices of deionized water and different salt solutions.

Solution	n
H ₂ O (deionized)	1.3375
NaCl (2M)	1.3541
KCl (2M)	1.3514
ZnSO ₄ (2M)	1.3725

The NaCl and KCl solutions have similar refractive indices of $n_{\text{NaCl}} = 1.3541$ and $n_{\text{KCl}} = 1.3514$, while the ZnSO₄ solution has a significantly higher refractive index of $n_{\text{ZnSO}_4} = 1.3725$ than the others. This is due to the much higher polarizability of the larger SO₄²⁻ ion with a higher ionic charge in comparison to the Cl⁻ ion. Additionally, the higher molar mass of ZnSO₄ results in a significantly higher mass density at the same molar concentration (2M). Na⁺ has a higher charge density than K⁺ due to its smaller size, which polarizes the surrounding water molecules more, leading to a slightly higher refractive index.

3.2 Diffusion coefficients

In order to determine the diffusion coefficients, the pixel value from the graphs has to be converted into a real length, using an appropriate conversion factor F , that is defined as:

$$F = 0.139 \frac{\text{mm}}{\text{pixel}} \quad (11)$$

3.2.1 NaCl

Figure 4 shows the picture out of the laserbeam on the screen plotted and quantified in pixels.

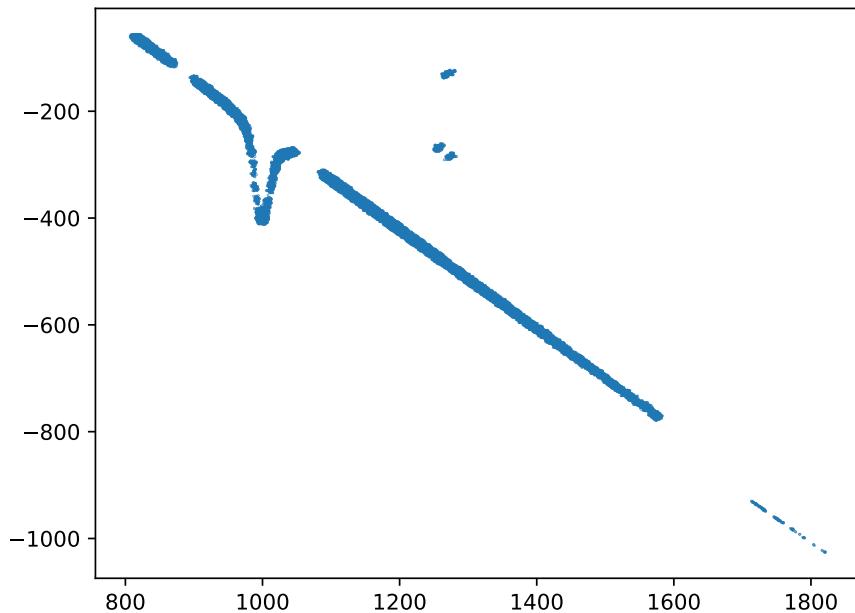


Figure 4: The screenshot after 60 s of NaCl, plotted in pixels.

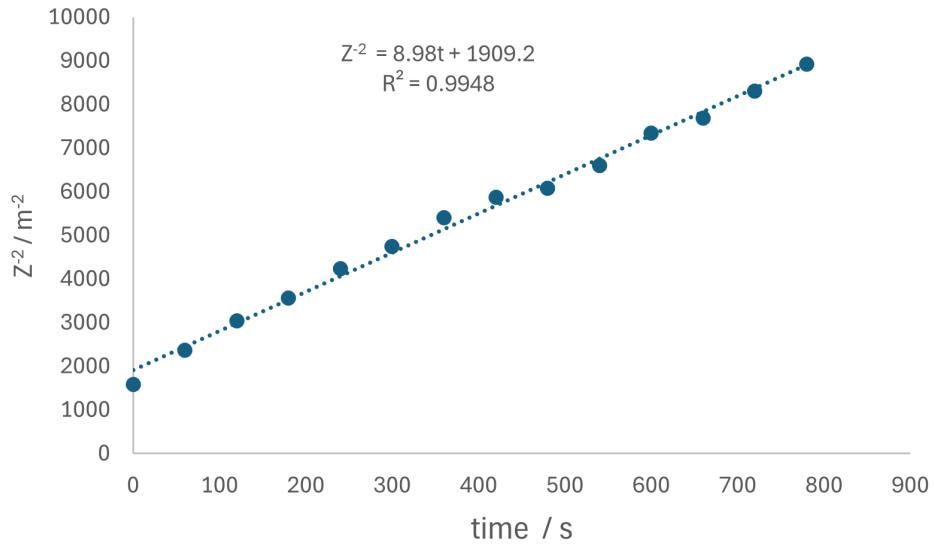
The depth of the amplitude $Z_{m,\text{NaCl}}$ was determined via python and converted into a real length using Equation 11.

$$Z_{m,\text{NaCl},10s} = \Delta y_{p,\text{NaCl},10s} \cdot F = 147.7 \text{ pixel} \cdot 0.139 \frac{\text{mm}}{\text{pixel}} = 20.5 \text{ mm} = 0.0205 \text{ m}$$

$Z_{m,\text{NaCl}}$ is calculated for every screenshot that was made in a 60 s time interval over the course of a 15 min video. To determine the diffusion coefficient D the amplitude $Z_{m,\text{NaCl}}$ negative squared is plotted over the time t as shown in Figure 5. The values, that were used for the plot are listed in Table 2.

Table 2: Measured values of Δy , Z , and Z^{-2} as a function of time of the NaCl sample.

Time [s]	Δy	Z [mm]	Z [m]	Z^{-2} [m^{-2}]
0	180.6246	25.1068	0.0251068	1586.4143
60	147.7368	20.5354	0.0205354	2371.3348
120	130.5184	18.1421	0.0181421	3038.2730
180	120.4435	16.7416	0.0167416	3567.8270
240	110.5652	15.3686	0.0153686	4233.8307
300	104.4336	14.5163	0.0145163	4745.5833
360	97.8275	13.5980	0.0135980	5408.1442
420	93.8439	13.0443	0.0130443	5877.0411
480	92.2600	12.8241	0.0128241	6080.5605
540	88.5696	12.3112	0.0123112	6597.8315
600	83.9791	11.6731	0.0116731	7338.8412
660	82.0591	11.4062	0.0114062	7686.2952
720	78.9158	10.9693	0.0109693	8310.7991
780	76.1865	10.5899	0.0105899	8916.9066

Figure 5: The values of the negative squared amplitude $Z_{m,\text{NaCl}}$ plotted over the time t .

To evaluate Figure 5, Equation 10 is rearranged to Z^{-2} to get Equation 13, which shows the connection between the slope m and the diffusion coefficient.

$$Z^{-2} = \frac{4\pi D n_0^2}{(n_1 - n_2)^2 L^2 K^2} \cdot t \quad (12)$$

$$= m \cdot t \quad (13)$$

The linear fit is described by the equation

$$y = m \cdot t + b,$$

where the slope is $m = 8.98 \text{ m}^{-2} \text{ s}^{-1}$ and the intercept is $b = 1909.2 \text{ m}^{-2}$. The slope of the line is defined as

$$m = \frac{4\pi D n_0^2}{(n_1 - n_2)^2 L^2 K^2}. \quad (13)$$

The parameters are given as $K = 0.01 \text{ m}$, $L = 0.268 \text{ m}$, $n_{2,\text{NaCl}} = 1.3541$, $n_1 = 1.3375$, and $n_0 = 1$. Using the experimentally determined slope, the diffusion coefficient of sodium chloride can be calculated as

$$D_{\text{NaCl}} = m \cdot \frac{(n_1 - n_2)^2 L^2 K^2}{4\pi n_0^2}. \quad (14)$$

Using the numerical values yields the diffusion coefficient of sodium chloride as

$$D_{\text{NaCl}} = \frac{(1.3541 - 1.3375)^2 (0.268 \text{ m})^2 (0.01 \text{ m})^2 \cdot 8.98 \text{ m}^{-2} \text{ s}^{-1}}{4\pi \cdot 1^2} = 1.414 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}. \quad (15)$$

3.2.2 KCl

The same steps that were discussed for the NaCl sample were performed on the values of the KCl sample. The values are listed in Table 3 and plotted in Figure 6.

Table 3: Measured values of Δy , Z , and Z^{-2} as a function of time of the KCl sample.

Time [s]	Δy	Z [mm]	Z [m]	Z^{-2} [m^{-2}]
0	112.462	15.632	0.016	4092.247
30	102.618	14.264	0.014	4915.006
60	97.166	13.506	0.014	5482.051
90	87.700	12.190	0.012	6729.334
120	84.963	11.810	0.012	7169.810
150	82.487	11.466	0.011	7606.694
180	76.077	10.575	0.011	8942.683
210	73.859	10.266	0.010	9487.739
240	71.108	9.884	0.010	10236.224
270	68.714	9.551	0.010	10961.623
300	69.556	9.668	0.010	10698.023
330	64.574	8.976	0.009	12412.225
360	65.976	9.171	0.009	11890.370
420	59.690	8.297	0.008	14526.705
480	57.616	8.009	0.008	15591.461
540	56.105	7.799	0.008	16442.663
600	51.561	7.167	0.007	19468.170
660	50.408	7.007	0.007	20369.523
720	47.818	6.647	0.007	22635.090
780	47.298	6.574	0.007	23136.261
840	45.039	6.260	0.006	25515.384
900	41.436	5.760	0.006	30145.349

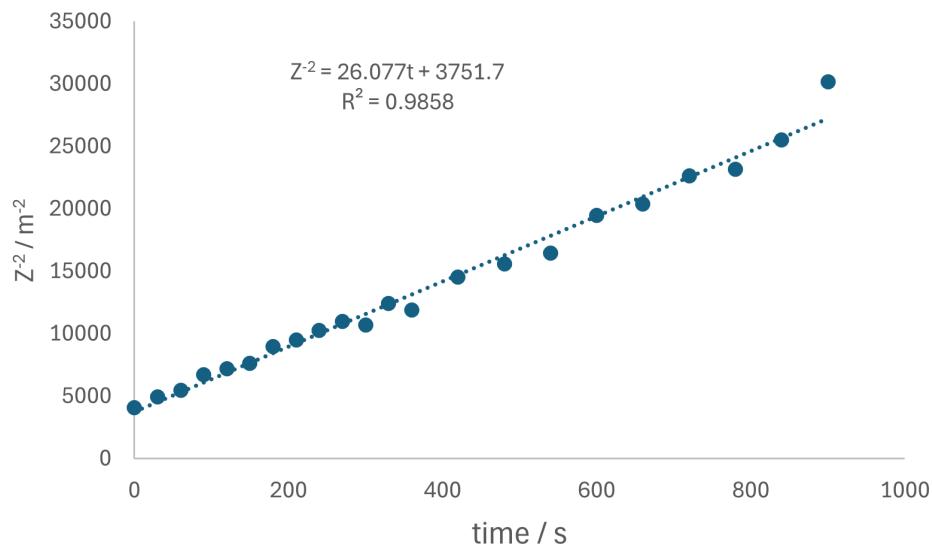


Figure 6: The values of the negative squared amplitude $Z_{m,\text{KCl}}$ plotted over the time t .

The slope m is determined to be

$$m = 26.077 \text{ m}^{-2} \text{ s}^{-1},$$

and the refractive index of the potassium chloride solution is

$$n_{1,\text{KCl}} = 1.3514.$$

Using these values, the diffusion coefficient of potassium chloride can be calculated by inserting them in the corresponding relation to Equation 14, yielding the diffusion coefficient of potassium chloride as

$$D_{\text{KCl}} = 6.636 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}.$$

3.2.3 ZnSO_4

The same steps that were discussed for the NaCl sample were also performed on the values of the ZnSO_4 sample. The values are listed in Table 4 and plotted in Figure 7.

Table 4: Measured values of Δy , Z , and Z^{-2} as a function of time of the ZnSO_4 sample.

Time [s]	Δy	Z [mm]	Z [m]	Z^{-2} [m^{-2}]
0	262.698	36.515	0.037	749.993
30	255.813	35.558	0.036	790.910
60	252.318	35.072	0.035	812.968
90	249.452	34.674	0.035	831.757
120	245.314	34.099	0.034	860.052
150	239.136	33.240	0.033	905.068
180	233.178	32.412	0.032	951.907
210	233.421	32.446	0.032	949.926
240	402.783	55.987	0.056	319.028
270	428.880	59.614	0.060	281.384
300	438.306	60.925	0.061	269.411
330	416.072	57.834	0.058	298.974
360	464.484	64.563	0.065	239.899
390	460.666	64.033	0.064	243.892
420	458.976	63.798	0.064	245.692
480	469.945	65.322	0.065	234.356
540	463.595	64.440	0.064	240.820
600	446.640	62.083	0.062	259.451
660	446.322	62.039	0.062	259.821
720	420.690	58.476	0.058	292.446
780	413.144	57.427	0.057	303.227
840	407.494	56.642	0.057	311.693
900	398.984	55.459	0.055	325.133
960	381.365	53.010	0.053	355.868
1020	370.073	51.440	0.051	377.916
1080	361.588	50.261	0.050	395.862
1140	345.601	48.039	0.048	433.331
1200	334.485	46.493	0.046	462.612
1260	323.186	44.923	0.045	495.523
1320	312.769	43.475	0.043	529.083
1380	302.859	42.097	0.042	564.275
1440	292.118	40.604	0.041	606.534
1500	283.425	39.396	0.039	644.310
1560	284.104	39.491	0.039	641.233
1620	277.021	38.506	0.039	674.442
1680	272.474	37.874	0.038	697.143
1740	267.477	37.179	0.037	723.433
1800	229.074	31.841	0.032	986.322

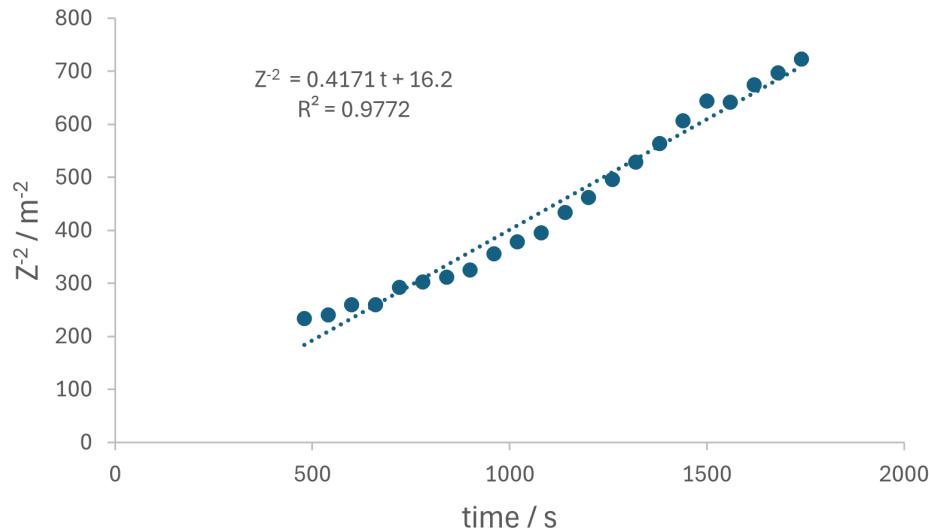


Figure 7: The values of the negative squared amplitude Z_{m,ZnSO_4} plotted over the time t .

Due to poor linearity during the first 540 s, the data from this interval is ignored in the plot. Nevertheless, even without that time period the corelation coefficient shows the lowest of the three accuracies.

The slope m is determined to be

$$m = 0.4171 \text{ m}^{-2} \text{ s}^{-1},$$

and the refractive index of the zinc sulfate solution is

$$n_{1,\text{ZnSO}_4} = 1.3725.$$

Using these values, the diffusion coefficient can be calculated by inserting them in the corresponding relation to Equation 14, yielding the diffusion coefficient of zinc sulfate as

$$D_{\text{ZnSO}_4} = 2.925 \cdot 10^{-10} \text{ m}^2 \text{ s}^{-1}.$$

4 Discussion

The measured refractive indices were $n_{\text{H}_2\text{O}} = 1.3375$, $n_{\text{NaCl}} = 1.3541$, $n_{\text{KCl}} = 1.3514$ and $n_{\text{ZnSO}_4} = 1.3725$, while the literature values were $n_{\text{H}_2\text{O,Lit}} = 1.333^{[2]}$, $n_{\text{NaCl,Lit}} = 1.348^{[3]}$, $n_{\text{KCl,Lit}} = 1.352^{[4]}$ and $n_{\text{ZnSO}_4,\text{Lit}} = 1.36^{[5]}$

The measured diffusion constants $D_{\text{NaCl}} = 1.414 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $D_{\text{KCl}} = 6.636 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ and $D_{\text{ZnSO}_4} = 2.925 \cdot 10^{-10} \text{ m}^2 \text{ s}^{-1}$ deviate from the ones obtained from literature sources which were $D_{\text{NaCl,Lit}} = 1.607 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $D_{\text{KCl,Lit}} = 1.994 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ and $D_{\text{ZnSO}_4,\text{Lit}} = 8.536 \cdot 10^{-10} \text{ m}^2 \text{ s}^{-1}$.^[6]

It is assumed that at a concentration of 2M, the refractive index is still proportional to the concentration. However, at higher concentrations, deviations can occur due to strong Coulomb interactions between multivalent ions such as ZnSO_4 , which lead to the formation of ion pairs and a electrostriction effect. This results in a higher density and molar refractivity of the ZnSO_4 solution compared to the other salt solutions, contributing to a deviation of the linear relationship between refractive index and concentration.^[7] The influence of deviations in the refractive indices on the diffusion coefficients calculated with Equation 10 is significant, since the refractive indices appear squared in the denominator^[1], resulting in a deviation of the calculated diffusion coefficients, as observed in the comparison of measured values with literature values.

5 Error Discussion

The standard deviation for the slope of the NaCl plot is $s_m = 0.188 \text{ m}^{-2} \text{ s}^{-1}$, which implies a solid accuracy, the correlation coefficient of $R^2 = 0.995$ as displayed in Figure 5 also implies this solid accuracy. The measurement values for KCl showed a standard deviation of $s_m = 0.701 \text{ m}^{-2} \text{ s}^{-1}$ corresponding to with a corelation coefficient of $R^2 = 0.986$, which is less accurate than for NaCl, but still in an acceptable range. The ZnSO_4 plot showed a standard deviation of $s_m = 0.014 \text{ m}^{-2} \text{ s}^{-1}$ and a correlation coefficient of $R^2 = 0.977$ the accuracy of this measurement shows the poorest accuracy. The standard deviations have to be put in correlation to the slope values to interpret the accuracy of the measurement, which gives $s_m/m = 0.021$ for NaCl, $s_m/m = 0.027$ for KCl and $s_m/m = 0.034$ for ZnSO_4 . Due to the fact that the slope is a linear term in Equation 14, the diffusion constant varies by those values, meaning an uncertainty of 3.4 %.

There were multiple sources for errors in this experiment that have to be considered. The refractive indices of the salt solutions deviate from literature values due to manual adjustment of the borderline in the refractometer, which was based on subjective visual inspection. Especially in the case of ZnSO_4 , the borderline was very blurry, which led to a measurement uncertainty in the refractive index. The measurements of the refractive indices were performed at room temperature without active thermal stabilization, which can lead to deviations as well, since refractive indices are temperature dependent. Another possible source for errors was the lamp that was used to measure the refractive indices, since the different wavelengths of light are refracted differently.

The addition of the salt solutions in the cuvette is also a source for errors because the two layers started mixing during the injection process of the salt solution if it was not done careful enough. The last possible error source was the recording process and the following conversion into data points. The brightness in the recording could have been too low for the python script to be able to convert properly, which leads to missing points in the resulting plot. Additionally the conversion from pixels into milimeters relies on the assumption that the distance between the screen and camera is the exact distance as stated in the instruction.

6 Conclusion

The refractive indices were measured as $n_{\text{H}_2\text{O}} = 1.3375$, $n_{\text{NaCl}} = 1.3541$, $n_{\text{KCl}} = 1.3514$ and $n_{\text{ZnSO}_4} = 1.3725$, which show small deviations from literature values. The diffusion constants of NaCl, KCl and ZnSO₄ were determined as $D_{\text{NaCl}} = 1.414 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$, $D_{\text{KCl}} = 6.636 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ and $D_{\text{ZnSO}_4} = 2.925 \cdot 10^{-10} \text{ m}^2 \text{ s}^{-1}$ by using the Schlieren method. Most deviations from literature values can be explained by mistakes during the conduction of the experiment or errors in the setup.

7 References

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