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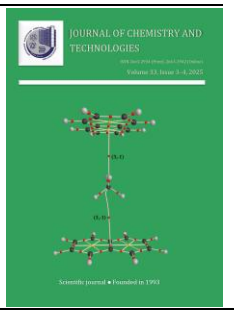

**UDC 662.818**

# MASS TRANSFER DURING THE DISSOLUTION OF SODIUM TETRABORATE IN WATER INTENSIFIED BY MECHANICAL STIRRING

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## Abstract

**The process of mass transfer during the dissolution of sodium tetraborate granules in water was investigated at temperatures ranging from 293 K to 323 K, and with agitator rotation rates of 1.67 to 6.67 $s^{-1}$. The study aimed to determine the dependency of the dissolution rate on the agitator rotation rate and water temperature. An analysis of the experimentally obtained results revealed that the most significant factor influencing the intensification of the dissolution process is the increase in the solution's temperature. The phenomena of external and internal diffusion during the dissolution of sodium tetraborate granules in water under agitation were also examined. A generalized criterion equation was applied to assess the mass transfer process, considering all the investigated factors. A comparison between experimental and theoretically calculated values showed that the maximum absolute relative error did not exceed 6 %. The obtained results are valuable for further research and potential applications in the chemical industry, pharmacology, and cosmetology.**

*Keywords:* sodium tetraborate; borax; dissolution; agitation; diffusion; mass transfer.

# МАСООБМІН ПІД ЧАС РОЗЧИНЕННЯ ТЕТРАБОРАТУ НАТРІЮ У ВОДІ, ІНТЕНСИФІКОВАНИЙ МЕХАНІЧНИМ ПЕРЕМІШУВАННЯМ

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## Анотація

**Процес масообміну під час розчинення гранул тетраборату натрію в воді досліджувався за температур у діапазоні від 293 К до 323 К та за частоти обертання мішалки від 1.67 до 6.67 $с^{-1}$. Метою дослідження було визначити залежність швидкості розчинення від частоти обертання мішалки та температури води. Аналіз експериментально отриманих результатів показав, що найсуттєвішим фактором, який впливає на інтенсифікацію процесу розчинення, є підвищення температури розчину. Також були розглянуті явища зовнішньої та внутрішньої дифузії під час розчинення гранул тетраборату натрію в воді за умови перемішування. Для оцінки процесу масопереносу з урахуванням усіх досліджуваних факторів було застосовано узагальнене критеріальне рівняння. Порівняння експериментальних та теоретично розрахованих значень показало, що максимальна абсолютна відносна похибка не перевищує 6 %. Отримані результати є цінними для подальших досліджень і можливих застосувань у хімічній промисловості, фармакології та косметології.**

*Ключові слова:* тетраборат натрію; бура; розчинення; перемішування; дифузія; масоперенос.

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## Introduction

It is known that the chemical industry utilizes more than 64 products containing boric acid, including sodium tetraborate decahydrate, sodium octaborate tetrahydrate, and zinc borate. These compounds are available in the form of liquids, soluble and emulsified concentrates, granules, powders, tablets, pastes, and crystalline rods.

The industrial applications of borates are quite broad, including their use as insecticides, fungicides, and algicides, with a very low content of herbicides. They are widely employed in the pharmaceutical, agricultural, chemical industries, and cosmetology [1].

Borax (sodium tetraborate) is a naturally occurring alkaline compound. It is used as a preservative, buffer solution, antiseptic, and fungicide in the production of glazes and enamels, as well as for fire-resistant fabrics and wood. Additionally, borax is an ingredient in soap and detergent manufacturing 2]].

Borates have extremely low toxicity for mammals and exhibit a broad spectrum of activity against decay fungi and insects. They are relatively inexpensive and highly soluble in water. In many countries, borates are used as biocides, particularly as diffusion biocides for treating wood products [3].

The solubility of borates plays a crucial role in their industrial applications. Therefore, theoretical and experimental studies of borate dissolution processes remain a relevant research topic.

The physical dissolution of solids is significant in the chemical, food, hydrometallurgical, pharmaceutical, and other industries. It is typically the first stage of technological processes. Given the large-scale production volumes (e.g., potassium mineral fertilizers), dissolution equipment is often bulky, requiring substantial capital and energy investments [4].

Currently, there is a growing interest in granulating powdered materials. Granulation involves increasing the size of fine particles, resulting in spherical or cylindrical agglomerates. Granules offer several advantages over powdered materials: they do not cake or clump, maintain a narrow particle size distribution, and exhibit uniform characteristics [5].

Considering that the chemical industry employs reactors with volumes exceeding 10 $m^3$, compressed pellets with a diameter of 20 mm were used for the experimental study of sodium tetraborate (ST) dissolution.

There are numerous methodologies, calculation dependencies, and various theories for studying mass transfer processes in a solid-liquid system. In particular, the theory [6; 7] and the slip velocity theory [8] have been proposed for modeling mass transfer from particles to stirred liquids. However, all proposed theories and calculation dependencies are based on certain assumptions and corresponding simplifications. Therefore, several authors have focused on verifying the accuracy of these theories for predicting dissolution processes in industrial-scale stirred reactors [9] and identifying the relationship between impeller diameter and reactor diameter [10].

Newnham and Miles (1978) [10] and Pangarkar et al. (2002) [11] demonstrated that the theory leads to inaccurate predictions of the power influence per unit mass of particle dissolution. Newnham and Miles (1978) [10] argue that slip velocity theory better predicts experimental results compared to theory. Meanwhile, Pangarkar et al. (2002) [11] highlight that the lack of data on turbulence characteristics, especially for large reactors, and the difficulty of solving nonlinear equations for calculating slip velocity in turbulent flows limit the applicability of this approach.

Due to the reasons described above, several authors propose determining the mass transfer coefficient using semi-empirical equations or a modified theory [12–15]. Additionally, there are calculation dependencies that include the impeller rotation speed in conditions where solid particles are in a suspended state [16]. Recently, many authors have emphasized the importance of in-depth analysis of the mixing process during dissolution in pharmaceutical and chemical processes [6; 17–22].

Ukrainian researchers have devoted considerable attention to studying the dissolution of single particles and dissolution in a stationary layer, particularly in [23]. Works [24; 25] examine the physical dissolution process of a polydisperse mixture of benzoic acid in a gas-liquid flow. Based on the isotropic turbulence theory, the theoretical mass transfer coefficient was determined.

In [26], the dissolution of solid spherical particles under vacuum conditions was studied, creating conditions for liquid boiling, and mass transfer coefficients were determined. The research results were generalized into a criterion-based dependency describing chemical interaction processes accompanied by gas formation.

All the mentioned studies were conducted either with a single sphere or a stationary layer of substance. The uniqueness and novelty of our research lie in the fact that the spheres move freely within the reactor, interact with each other, and contact the liquid. Thus, this study most accurately reflects the dissolution process during mechanical stirring under industrial conditions.

It is also worth noting the work [27], which presents a mathematical model of mass transfer based on Sherwood, Reynolds, and Schmidt numbers. This model can be used to predict dissolution processes of various solid substances and mass transfer in porous media. The determination of the mass transfer coefficient during mixing in a solid-liquid system was performed by Joshi et al. [28] using an artificial neural network. This approach allowed describing the mass transfer process during mixing based on dimensionless complexes with relatively low error. The source analysis conducted in this study indicates the feasibility of using dimensionless complexes to describe dissolution processes. The correlation found using the neural network further confirms the relevance of using Sherwood, Reynolds, and Schmidt numbers.

Summarizing the literature review, we note the relevance of studying mass transfer processes in a solid-liquid system, predicting ways to intensify this process, and utilizing models based on dimensionless complexes.

## Experimental section

*Experimental Setup*

The schematic diagram of the experimental setup is shown in Fig. 1 [29]. An asynchronous motor (1) with a three-blade impeller (2) stirs distilled water in a heat-resistant beaker (3), in which sodium tetraborate (ST) granules (4) are placed. The temperature of the ST solution in water was measured using a thermocouple (5), which was connected to a computer-integrated system (CIS) (9). The CIS was connected to the heating element (8) and the electric motor (1) to maintain the required solution temperature and impeller rotation speed.

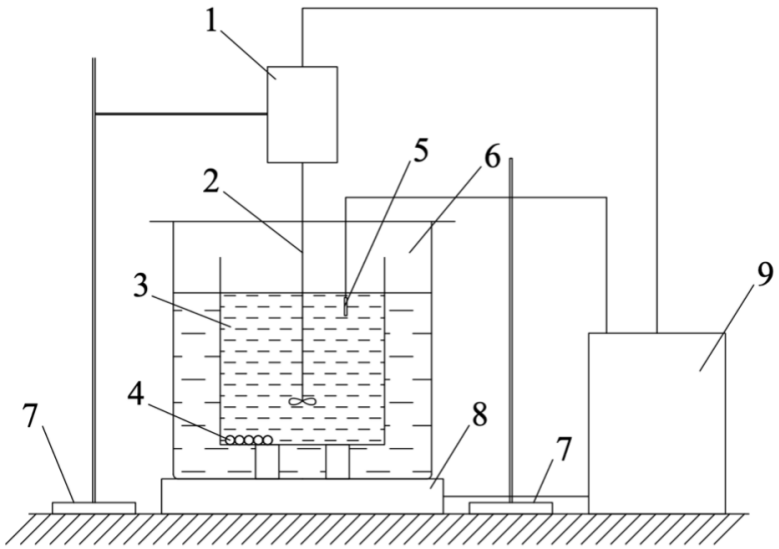


**Fig. 1. Schematic diagram of the experimental setup:**
**1 – asynchronous motor, 2 – three-blade impeller, 3 – heat-resistant beaker, 4 – sodium tetraborate granules, 5 – thermocouple, 6 – water bath, 7 – support, 8 – heating element, 9 – CIS**

*Experimental Methods*

Chemically pure sodium tetraborate decahydrate of Turkish origin was used in the study. The experiment was conducted in a cylindrical vessel with a height of 0.21 m and a diameter of 0.16 m. The temperature was controlled using a SESTOS D1s-Vr-220 regulator and a UNI-T UT-T03 thermocouple with an accuracy of 0.5 K. The mass of the granules was measured using an AD3000 electronic scale with an accuracy of 0.01 g. The diameter of the granules was measured with a YT-72300 micrometer with an accuracy of $0.1 \times 10^{-4}$ m.

Sodium tetraborate decahydrate powder was loaded into a mold and pressed into granules with a diameter of 20 mm under a pressing force of 4 MPa. Before the experiment, the ST granules were placed in a vessel with a saturated sodium tetraborate solution. At specific time intervals, the granules were removed, the surface moisture was

removed using filter paper, and their diameter and mass were measured. The experiment continued until a constant mass was achieved.

To study the dissolution kinetics of ST experimentally, the heating element was turned on to heat the distilled water in the vessel to the specified temperature. The three-blade impeller was set to rotate at a predetermined frequency, and five ST granules were simultaneously introduced. Every 180 seconds, the granules were removed from the vessel, the surface moisture was removed using filter paper, and their diameter and mass were measured. Each experiment was repeated at least three times until stable values were obtained. The change in sodium tetraborate concentration was calculated based on the change in ST mass, and the surface area of ST was determined by measuring its diameter.

Experiments were conducted at temperatures of 293, 303, 313, and 323 K and impeller rotation frequencies of 1.67, 3.33, 5, and 6.67 $s^{-1}$. The physical characteristics of ST and the sodium tetraborate solution are presented in Table 1 and Table 3.

*Table 1*

**Physical characteristics of sodium tetraborate granules** Ошибка! Источник ссылки не найден.**]**

| Diameter $d \cdot 10^3$, m | Mass, $M_0 \cdot 10^3$, kg | Porosity, % | Volume, $V \cdot 10^6$, m³ | Surface area, $F \cdot 10^3$, m² | Molar mass, kg/kmol |
|---|---|---|---|---|---|
| 20 | 6.5 | 12 | 4.19 | 1.256 | 381.37 |

According to reference data [30], the saturation concentration of sodium tetraborate in distilled water is presented in Table 2.

*Table 2*

**Saturation concentration of sodium tetraborate at different temperatures**

| T, K | 293 | 303 | 313 | 323 |
|---|---|---|---|---|
| Cs, kg/m³ | 47 | 72 | 112.20 | 179.1 |

*Table 3*

**Dynamic viscosity and density of the solvent at different temperatures**

| T, K | $\mu \cdot 10^4, Pa \cdot s$ | $\rho$, kg/m³ |
|---|---|---|
| 293 | 10.04 | 998.20 |
| 303 | 7.97 | 995.60 |
| 313 | 6.58 | 992.24 |

*Theoretical part*

The dissolution of solid substances is calculated using a system of differential material balance equations and the kinetic mass transfer equation.

$$\begin{cases} M_0 - M = W_C \cdot \overline{C_\tau} & (1) \\ \dfrac{dM}{d\tau} = \beta \cdot F \cdot (C_s - \overline{C_\tau}) \ , & (2) \end{cases}$$

where $M_0$ – initial mass of the granules, kg;
$M$ – current mass of the granule during dissolution at the measurement moment, kg;
$\overline{C_\tau}$ – average concentration of sodium tetraborate in the solution at the measurement moment, kg/m³;
$\tau$ – residence time of the ST granules in the apparatus, s;
$\beta$ – mass transfer coefficient during dissolution, m/s;
$F$ – surface area of ST granules, m²;
$W_c$ – volume of liquid in the vessel, m³;
$C_s$ – saturation concentration at the current solution temperature, kg/m³.

The difference $C_s - \overline{C_\tau}$ represents the driving force of the dissolution process. By integrating equation (2) over the time interval from 0 to $\tau$, was obtained the dependence of the mass transfer coefficient on time:

$$\beta = \frac{M_0 - M}{\overline{F} \cdot (C_s - \overline{C_\tau}) \cdot \tau} \quad (3)$$

The surface area of the granule $\overline{F_i}$ during dissolution decreases, so the average value of this quantity was used for calculations:

$$\overline{F_i} = \frac{\sum_1^5 \frac{\pi \cdot \left(d_i^2 + d_{i+1}^2\right)}{2}}{5} \quad (4)$$

where $d_i$ is the granule diameter at time $\tau$, $i$ is the sample number (i = 1, 2, 3, … n).

The concentration of sodium tetraborate in the solution during the time interval $\tau$ is determined by the formula:

$$C_{\tau_i} = \frac{M_i - M_{i+1}}{W} \quad (5)$$

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The average concentration of sodium tetraborate $\overline{C_{\tau_i}}$ in the solution is calculated by the following equation:

$$\overline{C_{\tau_i}} = \sum \frac{C_{i+1} + C_i}{2} \quad (6)$$

where $C_i$ – the current concentration of sodium tetraborate in the solution($C_0 = 0, \tau = 0$), kg/m$^3$.

In mass transfer theory, kinetic, hydrodynamic, and geometric variables are represented by dimensionless complexes. In dimensionless complexes, the mass transfer coefficient β is expressed as a Sherwood number:

$$Sh = \frac{\bar{\beta} \cdot \bar{d}}{D} \quad (7)$$

where $\bar{\beta}$ – average value of the mass transfer coefficient;
$\bar{d}$ – average diameter of the sodium tetraborate granule, m;
$D$ – diffusion coefficient of sodium tetraborate in distilled water, m$^2$/s.

The Sherwood number is a function of several parameters, the main ones being the Reynolds number (Re), the Schmidt number (Sc), and the geometric simplex (G):

$$Sh = f(Re, Sc, G)$$

The empirical equation for determining the Sherwood number of the studied system is:

$$Sh = A \cdot Re^x \cdot Sc^y \cdot G \quad (8)$$

where $A$ – proportionality constant determined from experimental data.

The Reynolds number, which characterizes the hydrodynamics of the system, was calculated as:

$$Re = \frac{\rho \cdot n \cdot d_m^2}{\mu}, \quad (9)$$

where $\rho$ – density of the liquid, kg/m$^3$;
$n$ – impeller rotation frequency, s$^{-1}$;
$d_m$ – diameter of the three-blade impeller, m;
$\mu$ – dynamic viscosity of the liquid, Pa·s.

The Schmidt number, which characterizes the physical parameters of the system, is calculated as:

$$Sc = \frac{\nu}{D}, \quad (10)$$

where $\nu$ – kinematic viscosity of the solution, m$^2$/s.

The geometric simplex value is determined as the ratio of the particle diameter to the apparatus diameter:

$$G = \frac{d_g}{D_a} \quad (11)$$

where $d_g$ – diameter of the sodium tetraborate granule, m;
$D_a$ – diameter of the apparatus, m.

Considering that the physical parameters of the solution change within a narrow range, according to the recommendations of many authors, the exponent near the Schmidt number was taken as 0.33. Thus, the empirical equation becomes:

$$Sh = A \cdot Re^x \cdot Sc^{0,33} \quad (12)$$

The diffusion coefficient was estimated using the Wilke-Chang method from the following equation [31]:

$$D = 7.4 \cdot 10^{-8} \cdot \left[ \frac{(\phi \cdot M_p)^{0,5} \cdot T}{\mu_p \cdot V_1^{0,6}} \right] \quad (13)$$

where $D$ – mutual diffusion coefficient at very low concentration of the dissolved substance, m$^2$/s;
$M_p$ – molecular mass of the solvent;
$T$ – temperature, K;
$\mu_p$ – viscosity of the solvent, mPa·s;
$V_1$ – molar volume of the solution at boiling temperature, m$^3$/mol;
ϕ – "association parameter" of the solvent (for water, (ϕ = 2.6)) [31].

## Results and discussion

The results of the experimental studies on the change in the diameter of sodium tetraborate ($NaB_4O_7$) granules (STG) are presented in Figure **2**. As a result of the dissolution of the STG, the diameter of the granules decreases. Therefore, to determine the mass transfer surface area, the average surface area of the granules was calculated during the experimental studies at different temperatures and stirring speeds using equation (4). The results of these studies are shown in Figure 2.

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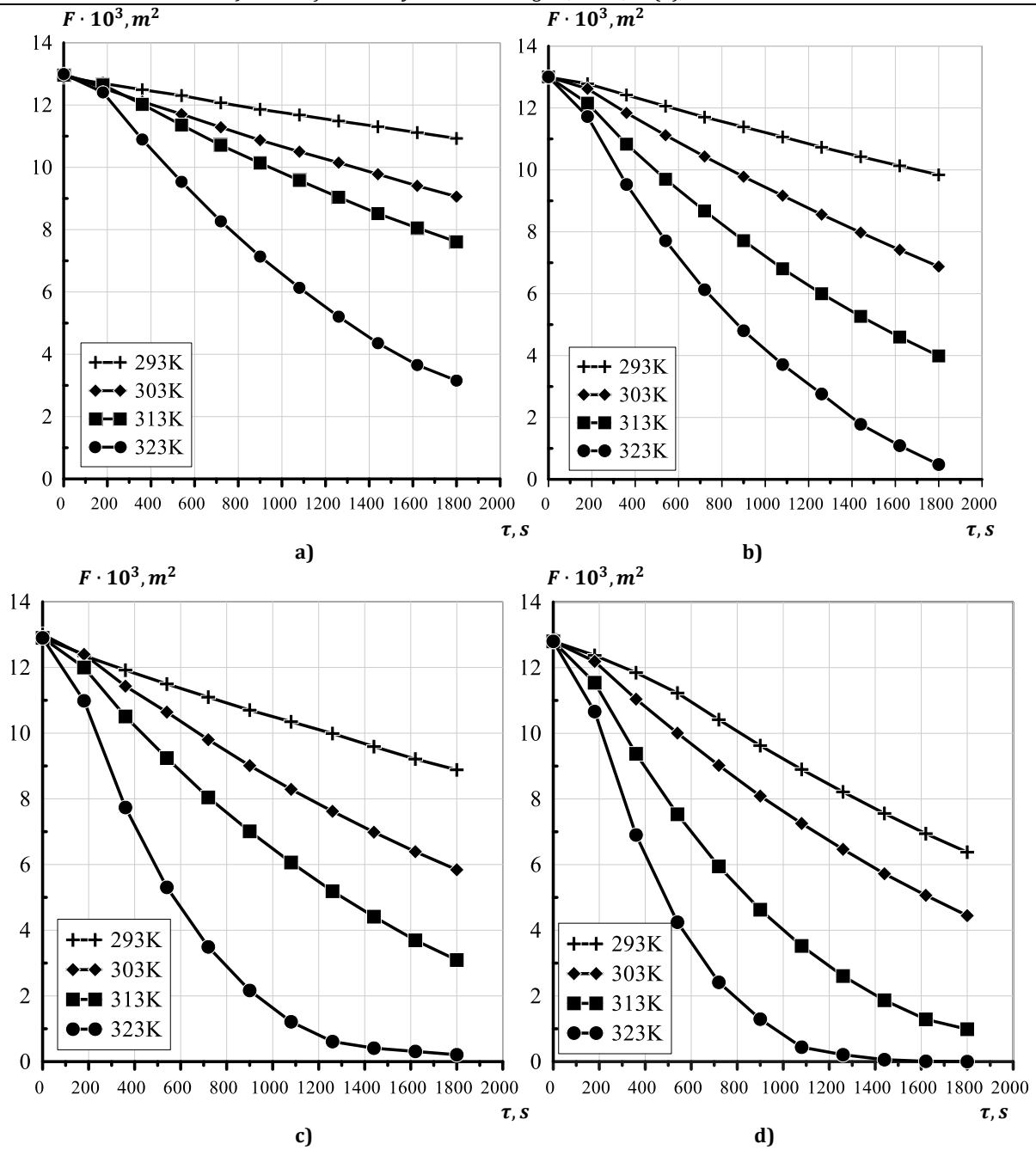


**Figure 2. Change in the average surface area of $NaB_4O_7$ granules during dissolution: a) 1.67 $s^{-1}$, b) 3.33 $s^{-1}$, c) 5 $s^{-1}$, d) 6.67 $s^{-1}$**

Analysis of the experimental results of the change in the surface area of $NaB_4O_7$ granules, presented in Figure 2, indicates that both the stirring speed and the temperature of the distilled water affect the rate of reduction in the average granule surface area. For example, after 1600 s, at a stirring speed of 1.67 $s^{-1}$ and a temperature of 293 K, the average surface area of the granules decreases from $1.30 \times 10^{-3}$ $m^2$ to $1.11 \times 10^{-3}$ $m^2$, i.e., by a factor of 1.17. At a temperature of 323 K, it decreases to $0.37 \times 10^{-3}$ $m^2$, i.e., by a factor of 3.52. At a stirring speed of 5 $s^{-1}$ and a temperature of 293 K, the average surface area decreases from $1.30 \times 10^{-3}$ $m^2$ to $0.92 \times 10^{-3}$ $m^2$, i.e., by a factor of 1.41, while at 323 K, it decreases to $0.27 \times 10^{-3}$ $m^2$, i.e., by a factor of 4.81. As seen in Fig. **3**, the concentration of the sodium tetraborate solution increases in a similar manner. Specifically, at a stirring speed of 1.67 $s^{-1}$ and an increase in water temperature from 293 to 323 K, the dissolution rate increases by a factor of 3.11 over 1600 s. At 323 K and with an increase in stirring speed from 1.67 $s^{-1}$ to 6.67 $s^{-1}$, the dissolution rate increases by only 1.06 times.

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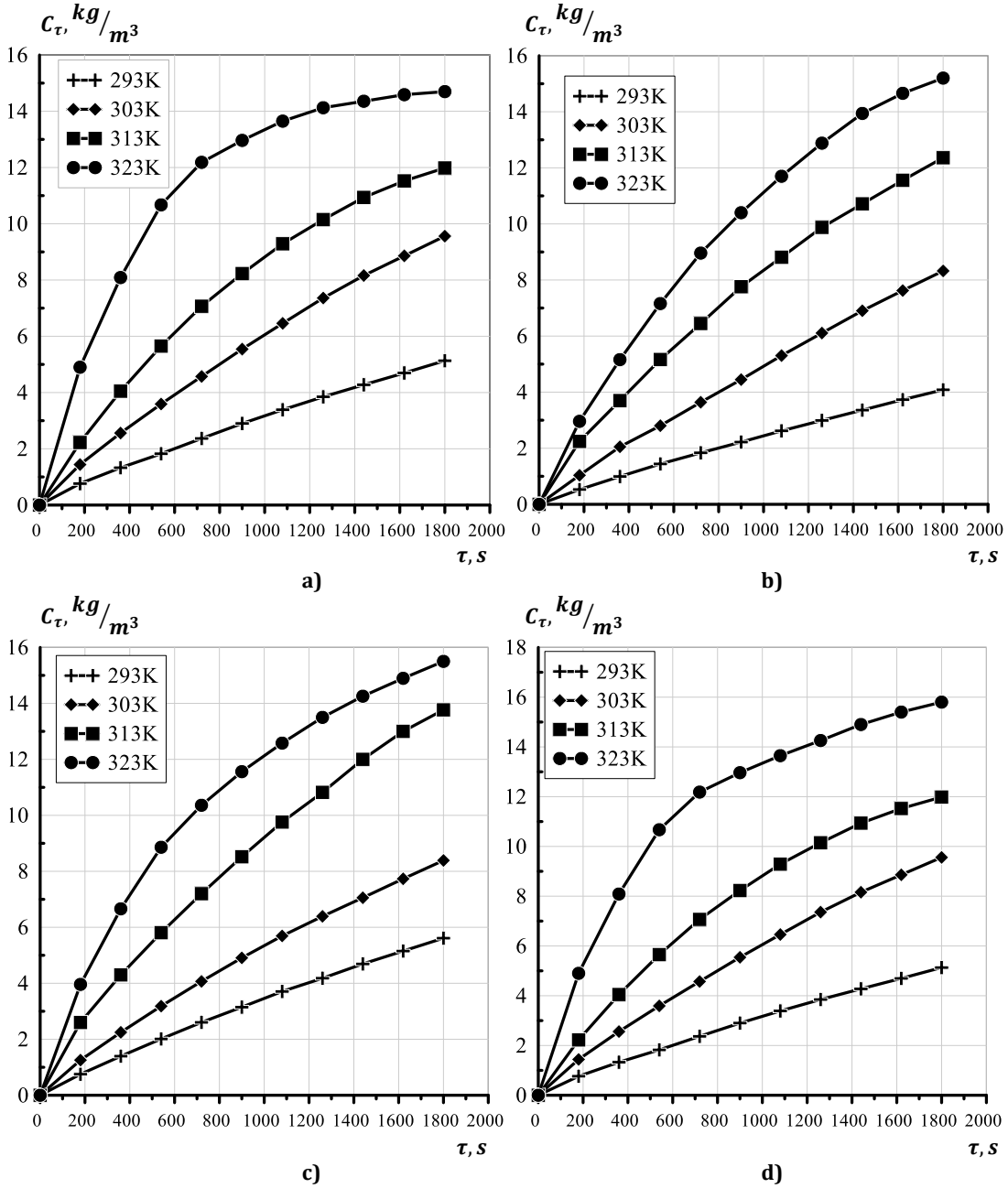


**Fig. 3. Kinetics of dissolution of sodium tetraborate (STG):**
**a) 1.67 $s^{-1}$, b) 3.33 $s^{-1}$, c) 5 $s^{-1}$, d) 6.67 $s^{-1}$**

To generalize the experimental data, it is useful to present them in dimensionless complexes. This representation of experimental results will allow for their application in predicting the dissolution kinetics of STG in industrial reactors of varying volumes. The mass transfer coefficients were calculated based on the experimental data using equation (3), and the calculation results are presented in Table 4.

*Table 4*

**Averaged values of mass transfer coefficients and KTN diameters depending on temperature and stirring frequency over 1800 s**

| $n, s^{-1}$ | $T, K$ | | | | $T, K$ | | | |
|---|---|---|---|---|---|---|---|---|
| | 293 | 303 | 313 | 323 | 293 | 303 | 313 | 323 |
| | $\beta \cdot 10^5, m/s$ | | | | $d_g \cdot 10^2$, m | | | |
| 1.67 | 1.89 | 2.22 | 2.71 | 3.46 | 1.89 | 1.84 | 1.78 | 1.62 |
| 3.33 | 2.57 | 2.92 | 4.14 | 4.71 | 1.74 | 1.87 | 1.53 | 1.61 |
| 5 | 2.81 | 3.83 | 4.91 | 7.66 | 1.94 | 1.67 | 1.61 | 1.21 |
| 6.67 | 3.58 | 4.86 | 7.81 | 8.61 | 1.66 | 1.57 | 1.18 | 1.21 |

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The ratio between the mass transfer coefficients $\frac{\beta_{T+10}}{\beta_T}$ indicates a decrease in the range of 1.20–1.61, which suggests that the process is in a diffusion-controlled region within the specified experimental interval.

The calculated values of the internal diffusion coefficients of sodium tetraborate from the pores of the pellets into the solution and the Schmidt numbers depending on the solution temperature are presented in Table 5.

*Table 5*

**Values of the internal diffusion coefficients of sodium tetraborate and Schmidt numbers**

| T, K | D ·10⁸ m²/s | Sc |
|---|---|---|
| 293 | 2.55 | 39.30 |
| 303 | 3.22 | 24.10 |
| 313 | 4.15 | 15.96 |
| 323 | 5.16 | 10.73 |

To determine the unknown coefficients A and x in equation (12), the experimental data were presented as a graphical dependence $Sh/Sc = f(Re)$ in logarithmic coordinates (

Fig. **4**(a)). By approximating the results of the experimental studies with a power function, the calculated values of the coefficient A and the exponent x for the investigated sodium tetraborate solution temperatures were obtained.

$$Sh = 0{,}589 \cdot Re^{0{,}5} \cdot Sc^{0{,}33} \cdot \frac{d_g}{D_a} \qquad (14)$$

A comparison of the calculated Sherwood numbers based on experimental data with the theoretically calculated values based on equation (14) is presented in

Fig. 4(a). As shown in

Fig. 4(b), the maximum absolute relative error does not exceed 6%, which is considered acceptable for practical calculations of technological parameters for the dissolution of granular sodium tetraborate in chemical industry plants.

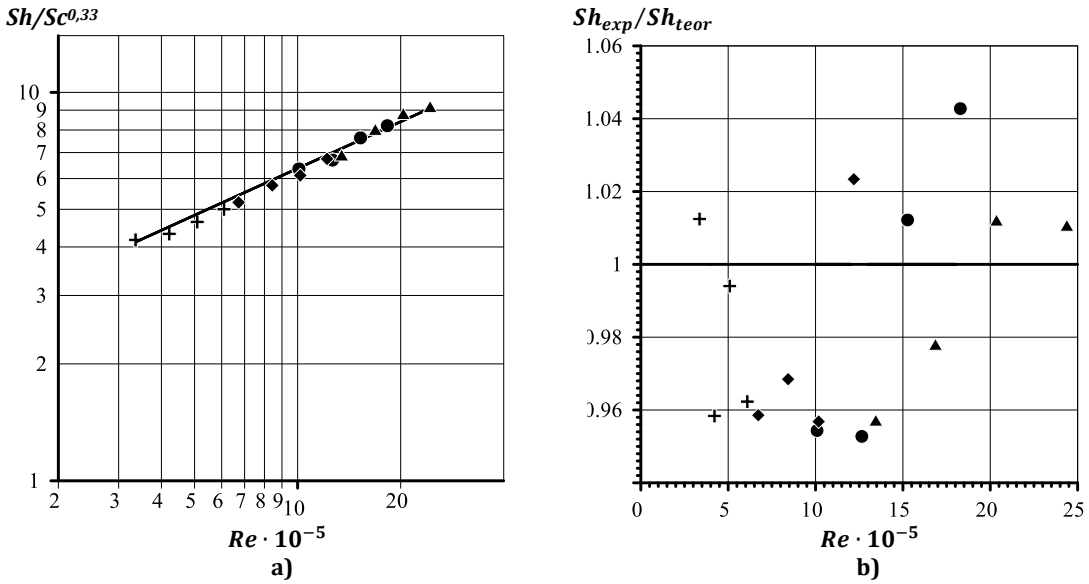


**Fig. 4. consists of two sub-graphs: (a) the graphical dependence $Sh/Sc^{0,33}=f(Re)$ based on experimental data, and (b) the ratio of calculated Sherwood numbers from experimental data to theoretically calculated values based on equation (14), expressed as a function of the Reynolds number $Sh_{exp}/Sh_{teor} = f(Re)$.**

## Conclusions

The dissolution of sodium tetraborate pellets in distilled water was studied at different temperatures and stirring speeds. The results showed that the driving force for the dissolution process depends on both the stirring speed and the temperature of the distilled water. Data analysis revealed that increasing the stirring speed and solution temperature led to a higher dissolution intensity of sodium tetraborate. However, temperature had a more significant effect on the intensification of the dissolution process compared to the stirring speed, although it did not change the process from diffusion-controlled to kinetically controlled. The mass transfer process was assessed using a generalized dimensionless complex that accounted for all the studied factors. Experimental values were

compared with theoretically calculated values, and it was found that the maximum absolute relative error did not exceed 6%. These results indicate that the obtained data are sufficiently accurate for practical calculations of technological parameters during the dissolution of granular sodium tetraborate in the chemical industry.

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