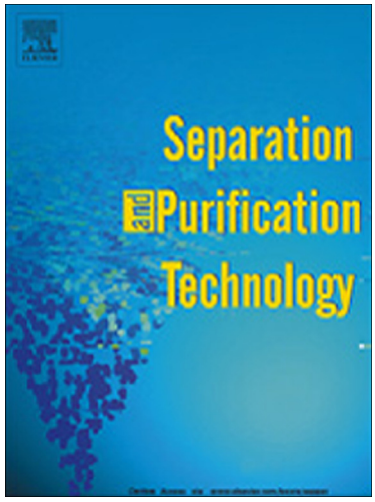

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Rotation Generator of Hydrodynamic Cavitation for Water Treatment

Martin Petkovšek[a]*, Mojca Zupanc[b,c,d], Matevž Dular[a], Tina Kosjek[b], Ester Heath[b,c], Boris Kompare[c], Brane Širok[a]

[a]Laboratory for water and Turbine Machines, Faculty of Mechanical engineering, University of Ljubljana, Aškerčeva 6, 1000 Ljubljana SI-Slovenia
[b]Department of Environmental Sciences, Jožef Stefan Institut, Jamova 39, 1000 Ljubljana, SI-Slovenia
[c]Jožef Stefan International Postgraduate School, Jamova 39, 1000 Ljubljana, SI-Slovenia
[d]Institute for Ecological Engineering , Ljubljanska 9, 2000 Maribor, SI-Slovenia
[e]Faculty of Civil and Geodetic Engineering, University of Ljubljana, Hajdrihova 28, 1000 Ljubljana SI-Slovenia
*corresponding author (martin.petkovsek@fs.uni-lj.si)

Abstract
Nowadays, due to lack of freshwater resources a sufficient wastewater management is an environmental concern. This global issue is resulting in the rapid growth of technologies for wastewater treatment. In this study a novel rotation generator of hydrodynamic cavitation is presented, which is used as a tool for pharmaceuticals removal in water. Cavitation is a major concern in the design of turbomachines due to the performance degradation, vibrations and cavitation erosion. The primary task for engineers is to avoid hydraulic machines to work in cavitation conditions, but in the present case, we are using cavitation as a tool for water treatment. On presented machine analysis of hydrodynamics is made, where the extent and aggressiveness of cavitation is evaluated. The study has shown, that for a sufficient treatment, hydrodynamic cavitation with combination of hydrogen peroxide is needed. Parameter which is also very important is the temperature during the process. The experimental results show that hydrodynamic cavitation has a good potential for efficient removal of pharmaceuticals what suggests to continue with research in this field and to consider an appropriate design for a commercial use.

Keywords: Hydrodynamic cavitation, Hydrogen peroxide, Pharmaceuticals removal, Optimal operating conditions

1. Introduction

Awareness of a drinking water shortage and influence of wastewater on the environment has brought the scientific community to start researching the presence of pharmaceuticals in the environment. Significant effort of understanding what kind of influence pharmaceuticals have on the environment and living creatures, has been made. For wastewater treatment many techniques, with the help of cavitation, have been developed.
Cavitation as a phenomenon is characterized by a formation, growth and collapse of bubbles within a liquid. There are four types of cavitation: hydrodynamic, acoustic, optical and particle induced. In hydrodynamic cavitation the geometry of a system is the reason for velocity fluctuations in a liquid flow, which can cause local drop of pressure. Vaporous cavity can form anywhere in a flowing liquid where the local static pressure is reduced to the liquid vapour saturation pressure. The primary parameter for determining the presence and intensity of cavitation is the cavitation number:

$$\sigma = \frac{p_\infty - p_v(T_\infty)}{\frac{1}{2}\rho v^2} \quad (1)$$

Cavitation number is a non-dimensional number, which is defined as the difference between the system and vapour pressure (at the system temperature) divided by the dynamic pressure. Decreasing the cavitation number results in a higher probability of cavitation occurrence or in an increase in the magnitude of the already present cavitation.

The cavitation bubble by its formation collects the energy from the surrounding fluid and this energy can mainly be released in two ways: i) at a symmetrical bubble collapse pressure impulse in an order of several hundred bars can be released [1], ii) at an asymmetrical collapse in the vicinity of a solid boundary a so called micro yet is formed, which can reach the velocities in the order of 100 m/s [2]. It is also known, that during bubble collapse very high temperatures of several thousand K occur [3], but these last for very short time - in one μs the temperature drops to the temperature of the surrounding fluid. Such extreme conditions are adequate to rupture or kill biological structures or they can cause water molecules to dissociate into OH and H radicals.

Different organic compounds demand different techniques and methods for treatment. The destruction of organic compound using cavitation is usually described by two methods, with free radicals and pyrolysis. Which mechanism is prevailing depends on organic compound and on cavitation intensity [4].

Till now many studies with ultrasonic cavitation were made, but there are only few studies on the applications of hydrodynamic cavitation to disintegrate water pollutants [4,5]. Sawant et al. [6] reported on high efficiency, more than 80 percent, of killing the zooplankton present in the sea water, with hydrodynamic cavitation. Sivakumar and Pandit [7] studied wastewater treatment with hydrodynamic cavitation using multiple hole orifice plates. Their conclusion was that hydrodynamic cavitation is more effective than ultrasonic cavitation by treating textile wastewater (rhodamine B solution). Results of Wang and Zhang [8] by treating alachlor with hydrodynamic cavitation showed that decreasing the cavitation number, which leads to an increase of cavitation events, increases the degradation rate of alachlor.

Also different hybrid techniques based on cavitation have been researched [9,10,11,12], mostly hydrodynamic cavitation in combination with ultrasonic cavitation, UV radiation or by adding chemicals. Zupanc et al. [13] investigated the removal of pharmaceutical with combination of hydrodynamic cavitation and hydrogen peroxide in a simple Venturi section. They reported that the average removal of selected pharmaceutical reached over 60%.

Till now no appropriate design of a hydrodynamic cavitation generator for a commercial use is available. Researchers mostly used multiple hole orifice plates [4,5,6,7,11] or simple Venturi restriction [13] on lab scale, where the flow restrictions causes considerable pressure losses in a system. Innovative design was presented by Kumar and Pandit [14]. They used a high-speed homogenizer, which consist of an impeller inside a cage-like stator with numerous slots, where cavitation generates.

In the present study the presented cavitation generator has an advantage of low pressure losses and easy scale up for an installation on a real system. In order to determinate the applicability of the hydrodynamic cavitation for wastewater treatment, experiments in deionized water were performed.

2. Experimental setup

For the purpose of investigating hydrodynamic cavitation effect on pharmaceuticals, a special cavitation generator (CG), shown in Fig. 1, was built at the Laboratory for Water and Turbine Machines (Faculty of Mechanical Engineering, University of Ljubljana).

The CG is based on two facing rotors with special radial grooves, where each one is spinning in the opposite direction. The rotors are driven by electrical motors with power of 0.37 kW each. They have special geometry which causes periodically repeating pressure drops. The rotating frequency is approximately 2800 rpm and their diameter is 90 mm, which means, that local velocities reach up to 26 m/s. The housing is made out of transparent acrylic glass due to visualization measurements. The type of cavitation, which is forming inside the CG is the so called shear cavitation where cavitation structures are formed due to the the opposite movement of the two shear layers [2].

The CG (1) was installed in an open loop, Fig. 2, where centrifugal pump (2) was used for fluid circulation. The cooling system (3) was necessary for keeping the fluid at a constant temperature. The temperature of the fluid was monitored by resistance temperature detector (RTD probe) (4) in a reservoir (5), while the static pressure inside the CG was monitored by the pressure transmitter (6). The static pressure inside the CG could be varied by installing the centrifugal pump upstream or downstream the CG, while the valves (7), upstream and downstream CG, were used for minor flow rate and pressure adjustment. The flow rate was held constant at 3 L/min during the experiments.
An open loop was chosen to simulate actual conditions of a real water treatment system. The cooling system in this experiment was necessary for holding the temperature at the desired value, because the volume of the sample was relatively small, 2.5 L.

To evaluate the extent and aggressiveness of cavitation from the hydrodynamic point of view we measured pressure oscillations by a hydrophone and in addition observed cavitation by a high speed camera (Fig. 3).

Pressure was measured with a hydrophone Reson TC4013 with usable frequency range 1 Hz to 170 kHz and receiving sensitivity of -211 dB ± 3 dB re 1V/µPa. Fastec Imaging HiSpec4 2G mono high-speed camera (CMOS sensor 1696×1710 pixels, pixel size 8×8 µm, up to 523 fps at full resolution, up to 300000 fps at reduced resolution) was used to capture the cavitation structures between the two rotors. For the present experiment the camera was recording at 8000 fps at a reduced resolution. The exposure time was set to 20 µs.

The quality of water (the gas and impurity content) can significantly influence the cavitation extent and aggressiveness [2,15]. In order to assure repeatable measurements the quantity of dissolved oxygen was monitored. Before the cavitation exposure, 9.5 mg of dissolved oxygen was constantly measured in 1 L sample.

In our study we examined the removal of four pharmaceuticals (ibuprofen, ketoprofen, carbamazepine and diclofenac) using a combination of hydrodynamic cavitation and hydrogen peroxide. The amount of pharmaceuticals was 1 µg of each pharmaceutical per one litre of sample. In addition 10 mL of 30 percent solution of hydrogen peroxide was introduced per one litre of sample. The experiments were performed in deionized water under different operating conditions. Duration of each experiment was 15 minutes. The water temperature could be held constant, ± 1 °C, from 20 °C (limitation due to the tap water temperature in the cooling system) up to 68 °C (limitation due to operating temperature of the acrylic glass).

2.1 Rotating disc design

Two rotating disc pair designs were investigated. Both designs had the same number of teeth and grooves - one disc had 11 grooves and the second 12 grooves, to avoid the resonance. Also the distance between the two facing rotors was the same for both designs (0.8 mm).

In the first design the teeth on the rotors were right angled and were separated by 7 mm deep and 10 mm wide grooves (Fig. 4a). As the two grooves of the opposing rotors pass each other a low static pressure region is established - if the pressure is low enough, the cavitation forms.

In the second design we modified the teeth of one rotor so that they exhibit an inclination at an 8° angle (we did not modify the second rotor). In this version when the teeth are aligned, the gap between them resembles the Venturi nozzle geometry (Fig. 4b).

3. Results of hydrodynamic analysis

The extent and aggressiveness of cavitation was observed by visualization and by means of pressure oscillations. Two conditions for each rotor design were investigated - high static pressure (150 kPa) inside CG (pump installed upstream of CG) and low static pressure (pump installed downstream of CG). Figure 5 shows main regions of cavitation occurrence for both rotor designs.

Cavitation is present in three different regions (Fig. 5). (1) notes the gap between the rotor and the housing, where attached cavitation forms on the leading edge of the teeth. Bubbles shed from the attached cavitation can also be seen here. When the two grooves are aligned, cavitation forms in the gap between the rotors (2). Small cavitation clouds can also form in the gap between the aligned teeth - (3) (this is characteristic of the 8° rotor design).

Figure 6 again shows the described cavitation patterns inside the CG. The left image shows attached cavitation on the leading edge of the right tooth - also small bubbles which are shed from attached cavitation can be seen. In the middle image the cavitation cloud between the aligned grooves is shown. A small cavitation cloud which formed in the slot between the two facing teeth is shown in the right image.

3.1 Right angled rotor design

Figure 7 shows cavitation between teeth on two opposite rotors. In first frame the two facing teeth are aligned. Later on the left one moves upward, and the right one downward. When grooves on opposing rotors align a low pressure region forms between them - in this moment cavitation potential reaches its peak.
In the first frame, where the teeth are aligned, cavitation between the grooves can be seen. The volume of cavitation cloud starts to reduce. In the right groove it collapses entirely in frame (4), while some bubbles are still present in the left groove. Between frame (6) and (9) the next pair of grooves is aligning, yet the cavitation extent is smaller than the preceding alignment. Cavitation is again bigger in the next alignment (between the frames (14) and (16)).
One can see that cavitation extent varies significantly between alignments, which is probably a result of very complex pressure field dynamics.

One can see, that in case of lower static pressure (Fig. 7), the extent of cavitation is much bigger than in case of higher static pressure (Fig. 8), where the cavitation is hardly seen. In the first frame no cavitation is noticeable. It starts to form in the left groove (frame (3) and (4)) and it reaches its maximum extent in frames (8) and (9).

To investigate the pressure oscillations within the CG, hydrophone was used. Results for the two absolute static pressures (100 and 150 kPa) are shown in Fig. 9.

One can see that both the amplitudes and gradients are smaller in the case of lower static pressure (Fig. 9), what implies, that despite its smaller extent, the cavitation is more aggressive at higher static pressure.

3.2 8° rotor design

In this design, when the teeth are aligned, the gap between them resembles the Venturi nozzle geometry. Figures 10 and 11 show cavitation inside CG after the modification of its teeth.

The Venturi shape geometry of the teeth causes a larger low pressure zone. One can see that the extent of cavitation is much bigger than in the original CG (Figs. 7, 8). We can expect that cavitation in this geometry will be more aggressive. This was again investigated by hydrophone measurements (Fig. 12).

The amplitude of pressure oscillations in both cases (Fig. 12) are bigger than in the original design (right angled teeth, Fig. 9). Very rapidly changing pressure field in the case of higher static pressure (Fig. 12), points to more aggressive cavitation process - it is expected that this design will be the most successful for pharmaceuticals removal.

4. The selection of the operating conditions

Based on pure hydrodynamic analysis one can conclude that the 8° rotor design is more suitable for further investigation with chemical analysis (direct measurements of pharmaceutical removal). For comparison we also performed one set of measurements in the right angled rotor design.
Preliminary chemical analysis on the removal of four selected pharmaceuticals was made for the four cases described in the previous section. The experimental conditions were the same for all tests - temperature 20 °C, time of exposure 15 min and amount of added 30 % solution of hydrogen peroxide was 10 mL per litre of sample.

Figure 13 shows preliminary chemical results of chemical analysis for preliminary measurements. One can see that the removal of pharmaceutical were significantly better in the case of 8° rotor design. The results are also better for the case of lower pressure inside the CG. This implies that it is the cavitation extent and not its aggressiveness that accelerates the pharmaceutical removal rate.

5. Results of removal of pharmaceuticals

Our preliminary tests showed, that the combination of 8° rotor design and low pressure inside the CG is the most successful at pharmaceuticals removal. Hence analysis of the influence of the temperature, duration of exposure and the amount of H_2O_2, were made for this combination only.
The removal of pharmaceuticals (%) was determined as a difference between the concentration of an individual pharmaceutical before and after cavitation experiments, by using solid phase extraction and derivatisation with MTBSTFA (N-(t-butyldimetylsilyl)-N-methyltrifluoroacetamid) prior to analysis with gas chromatography coupled to mass spectrometric detection. Detailed sample preparation and removal validation is presented in related work of Zupanc et al. [13].

Temperature, duration of exposure and the amount of hydrogen peroxide were systematically varied. All the results are given as the average removal of 3 samples - error bars note the discrepancies between the tests. They present the average removal of four selected pharmaceuticals (ibuprofen, ketoprofen, carbamazepine and diclofenac).

5.1 Influence of the temperature

Investigation on temperature dependence was made in the range from 20 °C up to 60 °C. Other variables remained constant (100 kPa of static pressure, 10 mL of hydrogen peroxide per 1 litre of sample and 15 minutes of exposure to cavitation).

It is clearly seen (Fig. 14), that the temperature has a significant effect on the removal of pharmaceuticals. The removal increases almost linear with temperature.

We tested, whether the temperature itself (without the presence of cavitation) and addition of H_2O_2 (10 mL 30% H_2O_2 per 1L of sample) could reduce the concentration of pharmaceuticals. Heating the sample up to 68 °C in 15 minutes resulted in only 23% removal of pharmaceuticals (compared to about 80 %, when cavitation was present).

A possible explanation of the temperature influence can be drawn from the definition of cavitation number, Eq. 1. Figure 15 shows how the cavitation number depends on absolute pressure and temperature.

The vapour pressure increases with the temperature, what leads to the decrease of the cavitation number. This implies, that the cavitation extent will be greater at a higher temperature - this relates well with results of preliminary measurement, where it was concluded, that the cavitation extent plays significant role in pharmaceuticals removal efficiency.

5.2 Influence of the duration of exposure

In the influence of duration of exposure to cavitation was investigated at temperature 50 °C, 100 kPa of static pressure and 10 mL of hydrogen peroxide per 1 litre of sample. Unexpectedly the removal does not increase with time of exposure - it oscillates (Fig. 16). Similar results were reported by Thompson [16] for the concentration of Na^+ and ultrasonic cavitation.

Reasons for achieved results are yet unknown and it will be an object of further inquiry.

5.4 Influence of the amount of added hydrogen peroxide

Finally we varied the amount of hydrogen peroxide addition. Other variables were kept constant (temperature of 50 °C, 100 kPa of static pressure and 15 minutes of time exposure of cavitation). According to Arrojo [17] and Gogate [4], free radicals can be formed in combination with cavitation and the presence of hydrogen peroxide (H_2O_2). One can see (Fig. 17) that the presence and the amount of H_2O_2 significantly influences the removal of pharmaceuticals.

The highest removal was obtained when 5 to 10 mL 30% H_2O_2 per 1 L of sample was added. Higher concentrations showed a negative effect on the removal - probably because the excess H_2O_2 can act as a radical scavenger for hydroxyl radicals [4]. On the other side the removal without added H_2O_2 is insufficient.
Similarly as the case of temperature influence, we tested the influence of H_2O_2 without the presence of cavitation. This was achieved by driving both rotors in the same direction (all other conditions remained unchanged, the amount of H_2O_2 was 10 mL per 1L of samle). Only 17% of pharmaceuticals were removed. This suggests that the main cause for pharmaceuticals removal are the free radicals formed from H_2O_2 during cavitation exposure. Results imply that the best combination is limited by addition of H_2O_2.

5.4 Gathered results from chemical analysis
Table 1 presents the experimental conditions for three sets of measurements. By the first set, temperature was varied from 20 to 60 °C, by the second set, time exposure to cavitation varied from 5 to 30 minutes and by the third set of experiments, the amount of hydrogen peroxide from 0 to 20 mL per 1L of sample was varied.
One can see that for the best reduction of pharmaceuticals, one needs a combinations of limited addition of H_2O_2 and high temperature of the medium. The temperature of the medium could be related to a larger cavitation extent, what cannot be tested in the present configuration. One can also see, that the removal rate between 5 and 30 minutes does not change. This speaks in favor of energy consumptions and time needed for removal by possible real water treatment system.

6. Conclusions

A new machine for generation of hydrodynamic cavitation is presented. Based on hydrodynamic analysis (visualization and dynamic pressure measurements) cavitation inside the CG was evaluated. Chemical evaluation of the removal of ibuprofen, ketoprofen, carbamazepine and diclofenac in deionized water was conducted.
Relation between chemical and hydrodynamic analysis of removal of pharmaceuticals revealed the following conclusions:

- Hydrodynamic cavitation with combination of hydrogen peroxide could be an efficient tool for removal of pharmaceuticals .
- Cavitation or hydrogen peroxide itself do not have sufficient effect on removal.
- Temperature (possibly only the extent of cavitation) and the amount of hydrogen peroxide are important for removal process, while unexpectedly the removal does not increase with prolonged time of exposure to cavitation.

- The generation of free radicals from hydrogen peroxide is catalyzed by cavitation process.
- The formation of free radicals during the interaction between the cavitation and H_2O_2 is probably the main cause for successful pharmaceuticals removal.

7. Acknowledgements

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Table captions:
Table 1: Results from chemical analysis by selected conditions.

Figure captions:

Figure 1: Scheme of cavitation generator (CG).

Figure 2: Schematic diagram of the experimental setup, 1: Cavitation generator (CG), 2: Pump,3: Cooling system, 4: RTD probe, 5: Reservoir, 6: Pressure gauge, 7: Control valve.

Figure 3: Placement of hydrophone, high-speed camera and illumination in CG.

Figure 4: Design of an original rotor (a) and redesigned rotor (b).

Figure 5: Model of a right angled teeth pair (left), and 8° teeth pair (right), with noted cavitation regions.

Figure 6: Attached cavitation with travelling bubbles (left), cavitation cloud between the grooves (middle), cavitation cloud in the slot (right).

Figure 7: Visualizations of the two right angled teeth rotors with static pressure of 100kPa (time step between frames is 1/8 ms).

Figure 8: Visualizations of the two right angled teeth rotors with static pressure of 150kPa (time step between frames is 1/8 ms).

Figure 9: Pressure oscillations in case of right angled rotor, with static pressure inside the CG of 100 kPa (upper diagram) and with static pressure inside the CG of 150 kPa (lower diagram).

Figure 10: Visualizations of the generated cavitation between right angled and 8° angled teeth rotor with static pressure of 100 kPa (time step between frames is 1/8 ms).

Figure 11: Visualizations of the generated cavitation between right angled and 8° angled teeth rotor with static pressure of 150 kPa (time step between frames is 1/8 ms).

Figure 12: Pressure oscillations in case of 8° angled rotor, with static pressure inside the CG of 100 kPa (upper diagram and with static pressure inside the CG of 150 kPa(lower diagram).

Figure 13: Removal (%) of pharmaceuticals in case of static pressure variations for right angled and 8° angled rotor design.

Figure 14: Removal (%) of pharmaceuticals in case of temperature variations.

Figure 15: Cavitation number corresponding to temperature and absolute pressure.

Figure 16: Removal (%) of pharmaceuticals in case of time dependence.

Figure 17: Removal (%) of pharmaceuticals in case of H_2O_2 dependence.

Table 1

Experimental set #	Rotors	Pressure [kPa]	Temperature [°C]	Time [min]	H_2O_2 [mL/L]	Removal [%]
1.1	8° + 0°	100	20	15	10	36
1.2	8° + 0°	100	30	15	10	42
1.3	8° + 0°	100	40	15	10	41
1.4	8° + 0°	100	50	15	10	70
1.5	8° + 0°	100	60	15	10	**82**
2.1	8° + 0°	100	50	5	10	66
2.2	8° + 0°	100	50	10	10	58
2.3	8° + 0°	100	50	15	10	**70**
2.4	8° + 0°	100	50	20	10	55
2.5	8° + 0°	100	50	25	10	50
2.6	8° + 0°	100	50	30	10	65
3.1	8° + 0°	100	50	15	0	16
3.2	8° + 0°	100	50	15	1	63
3.3	8° + 0°	100	50	15	5	**70**
3.4	8° + 0°	100	50	15	10	**70**
3.5	8° + 0°	100	50	15	20	56

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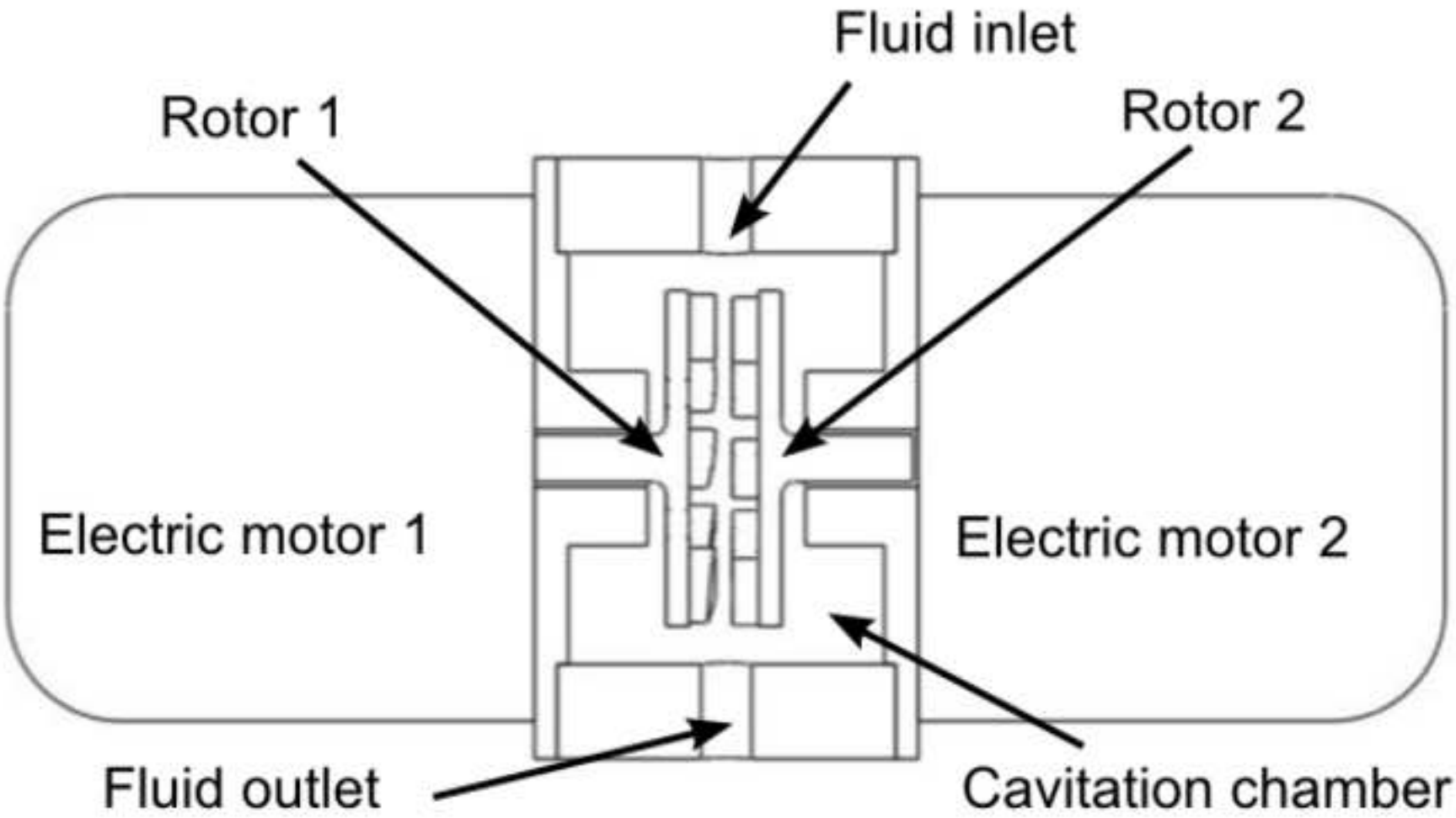

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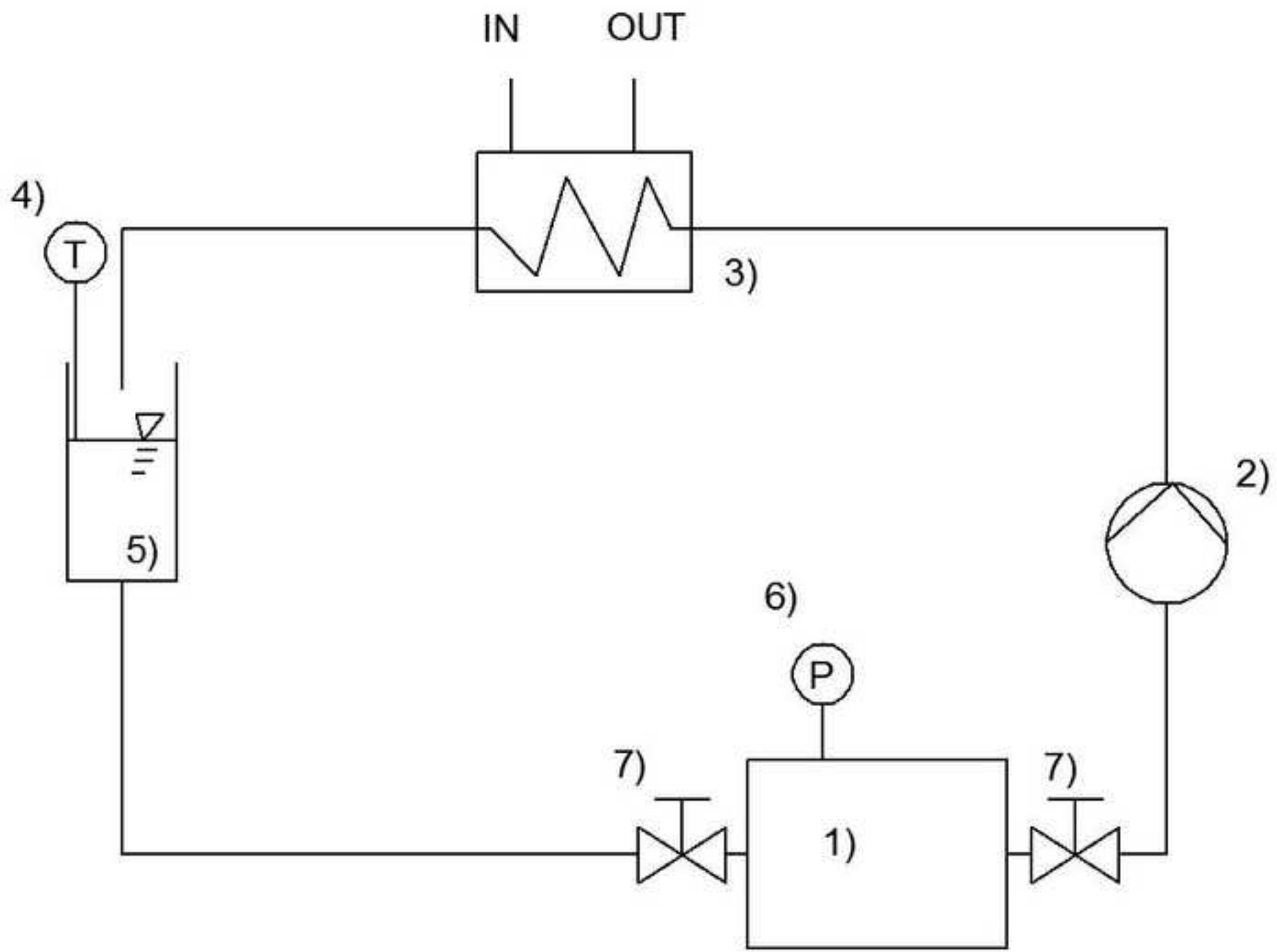

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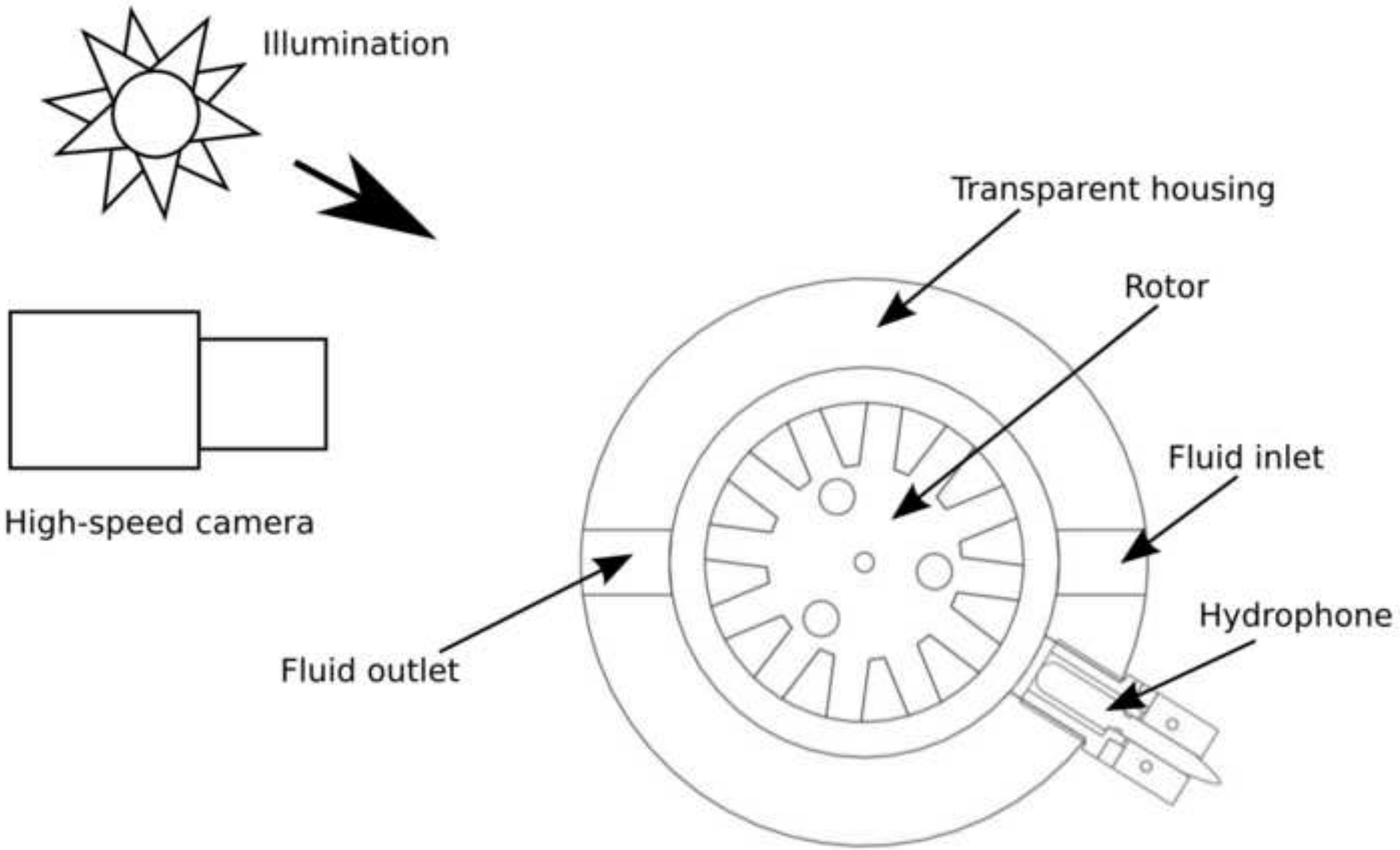

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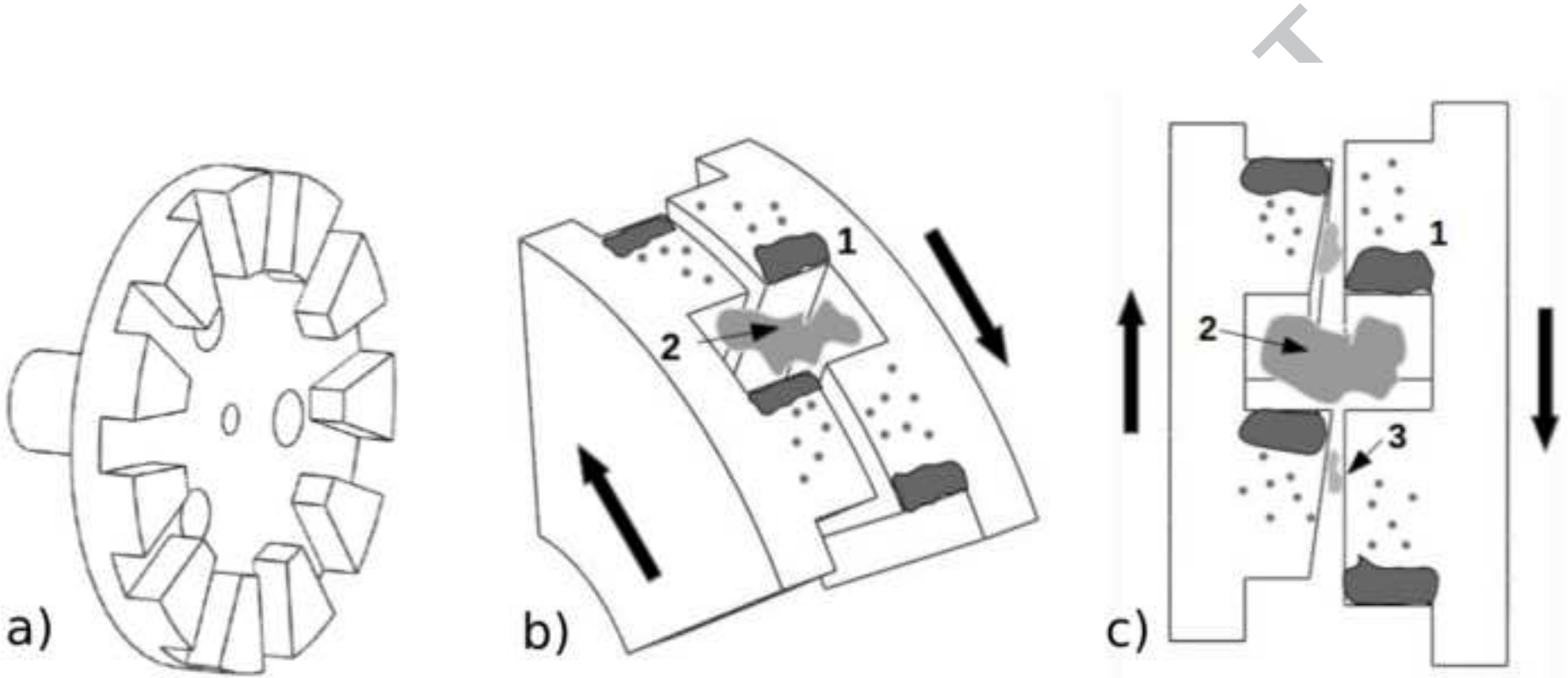

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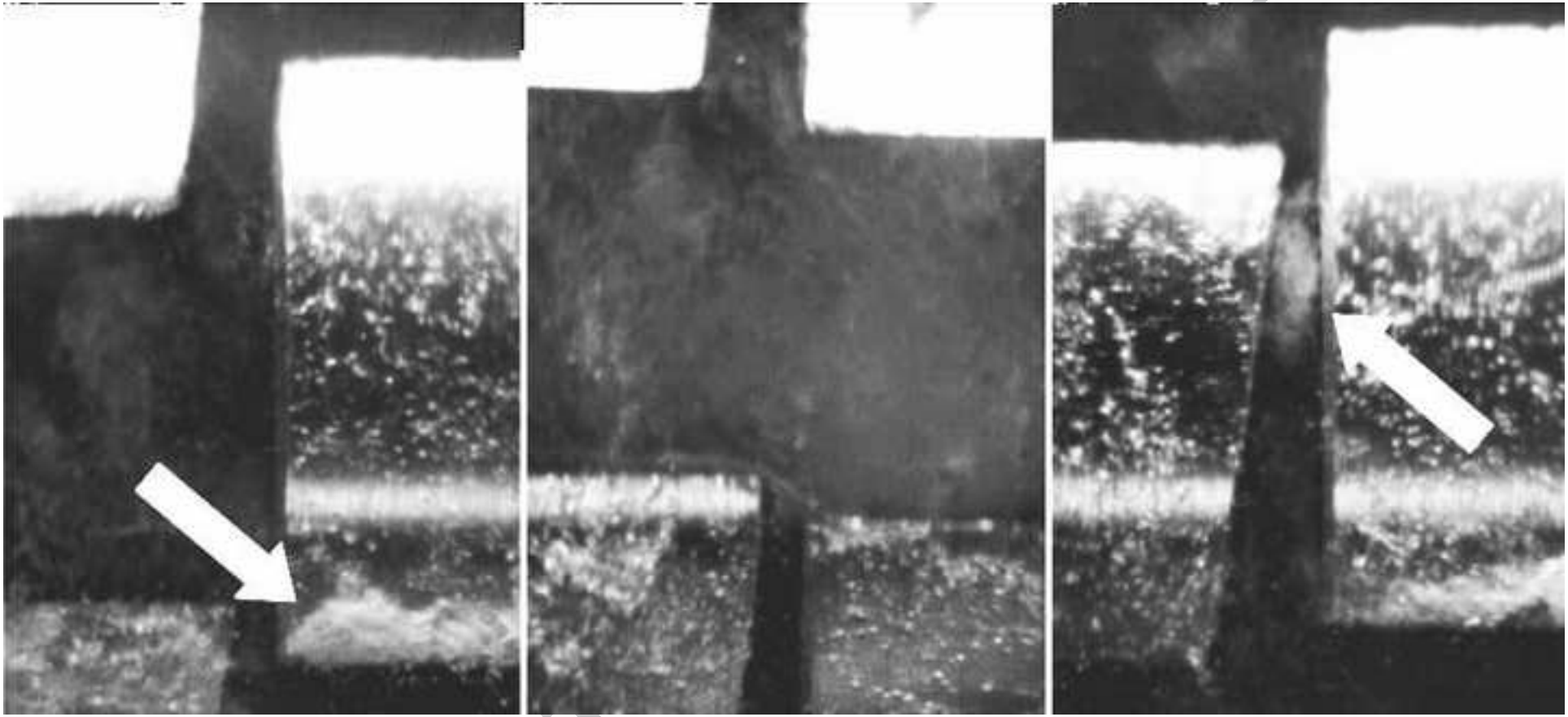

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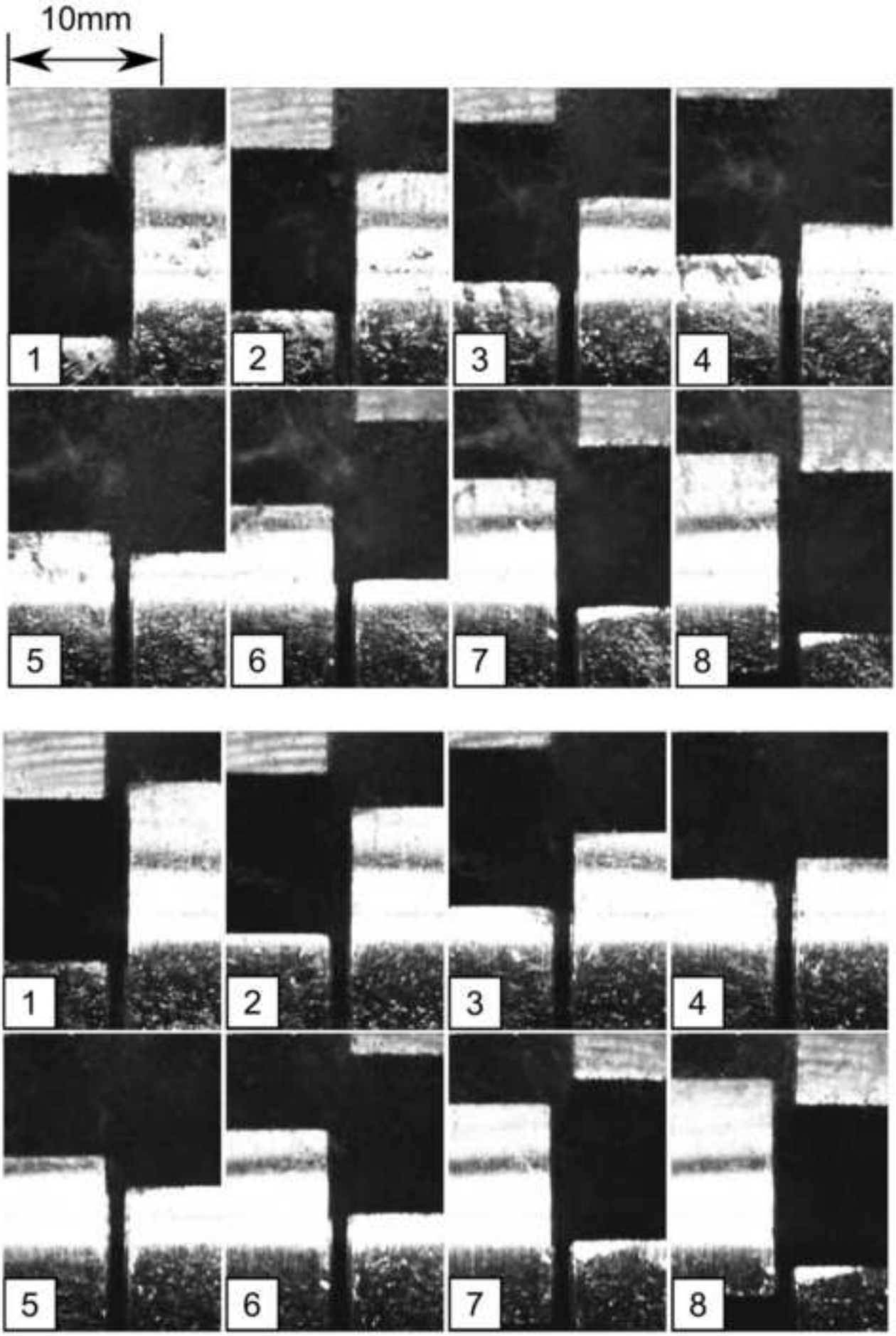

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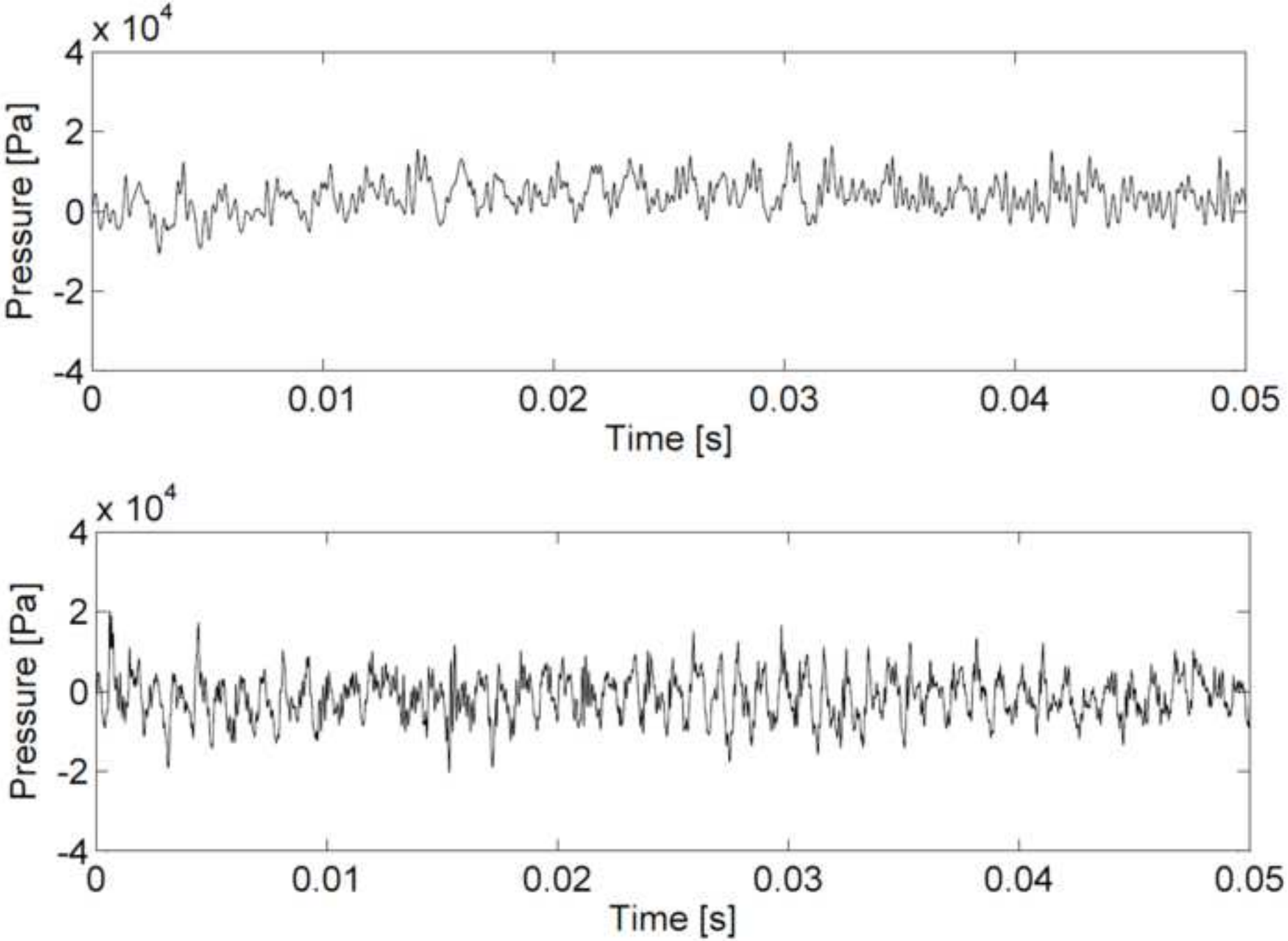

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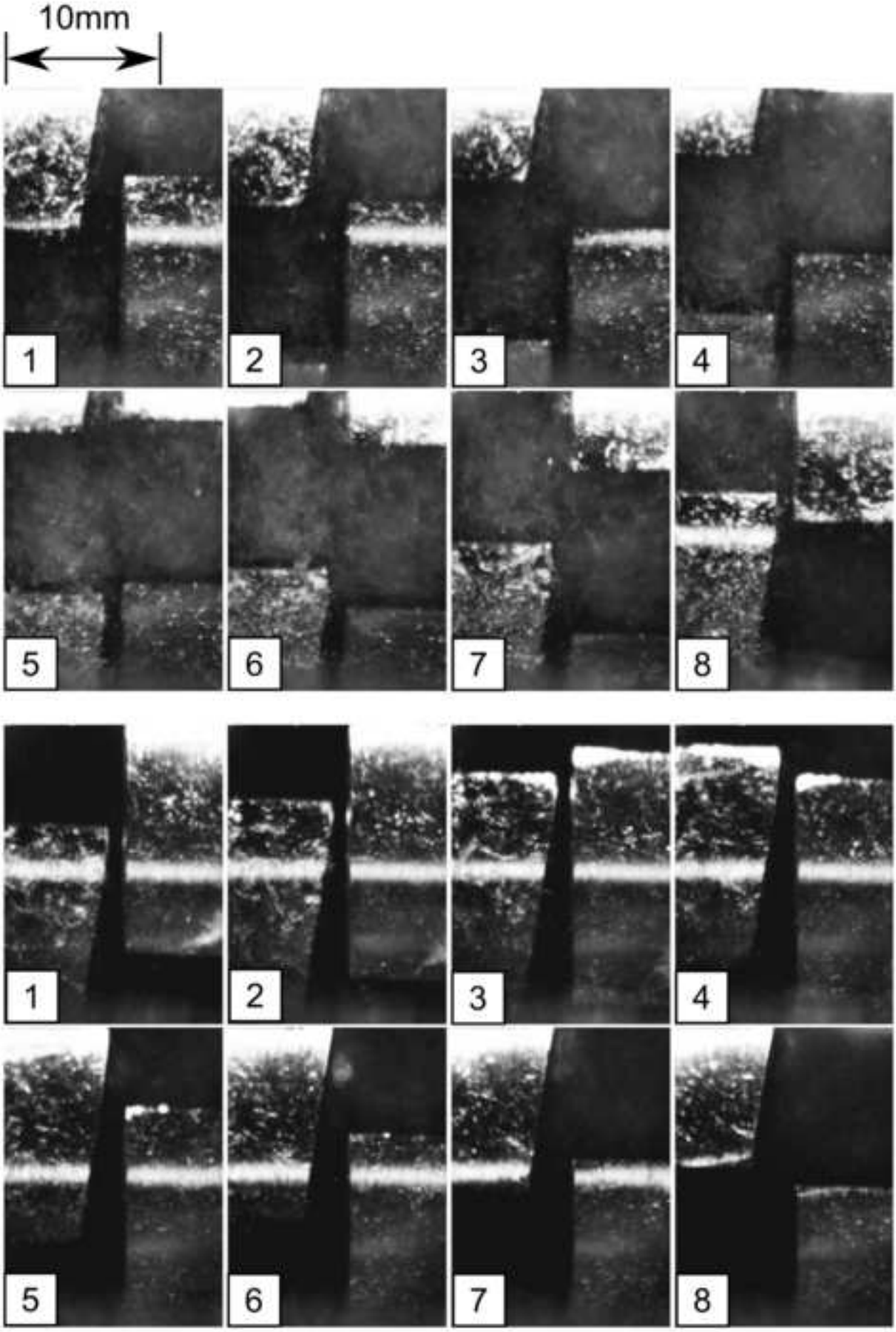

Figure_9

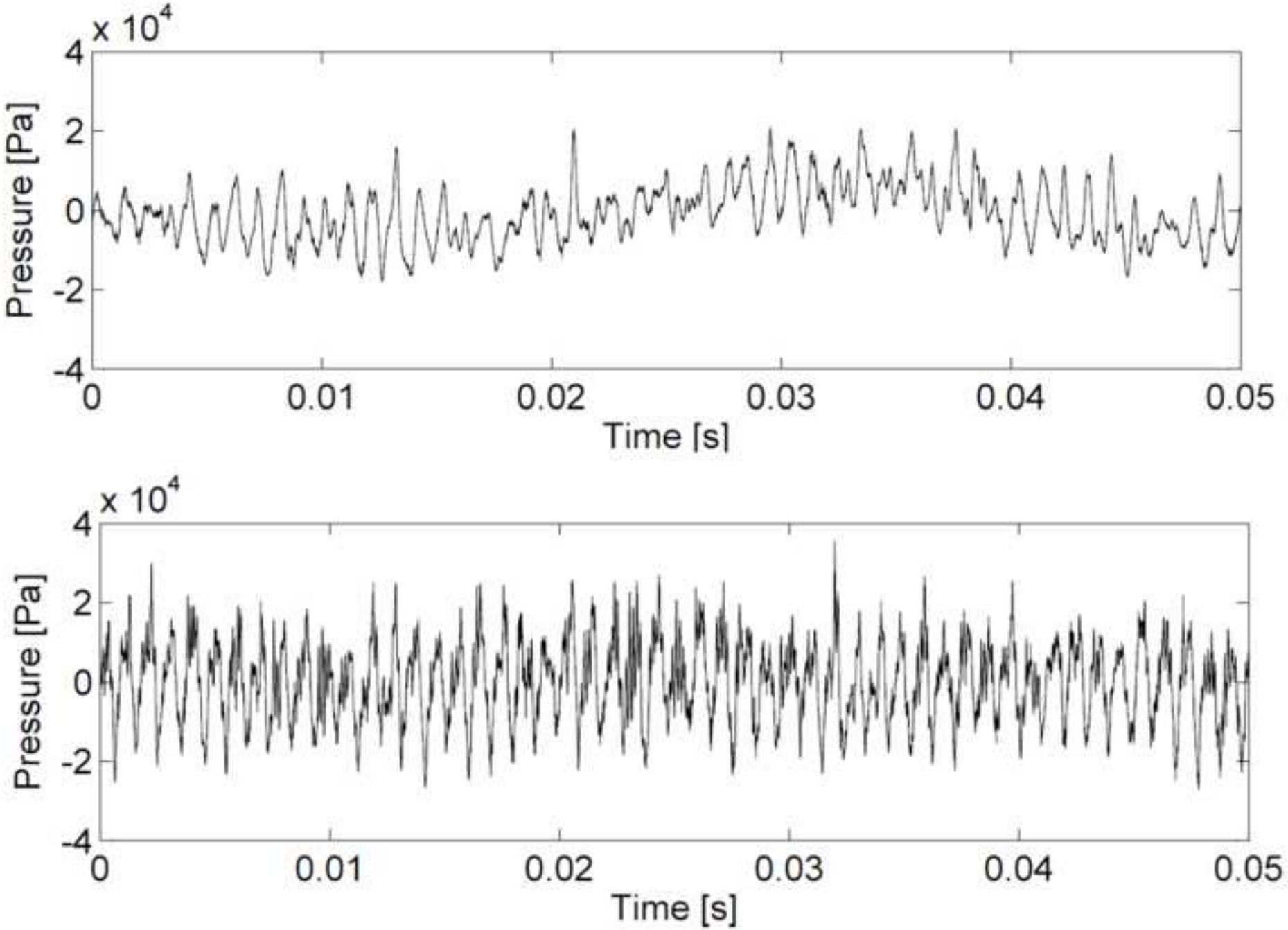

Figure_10

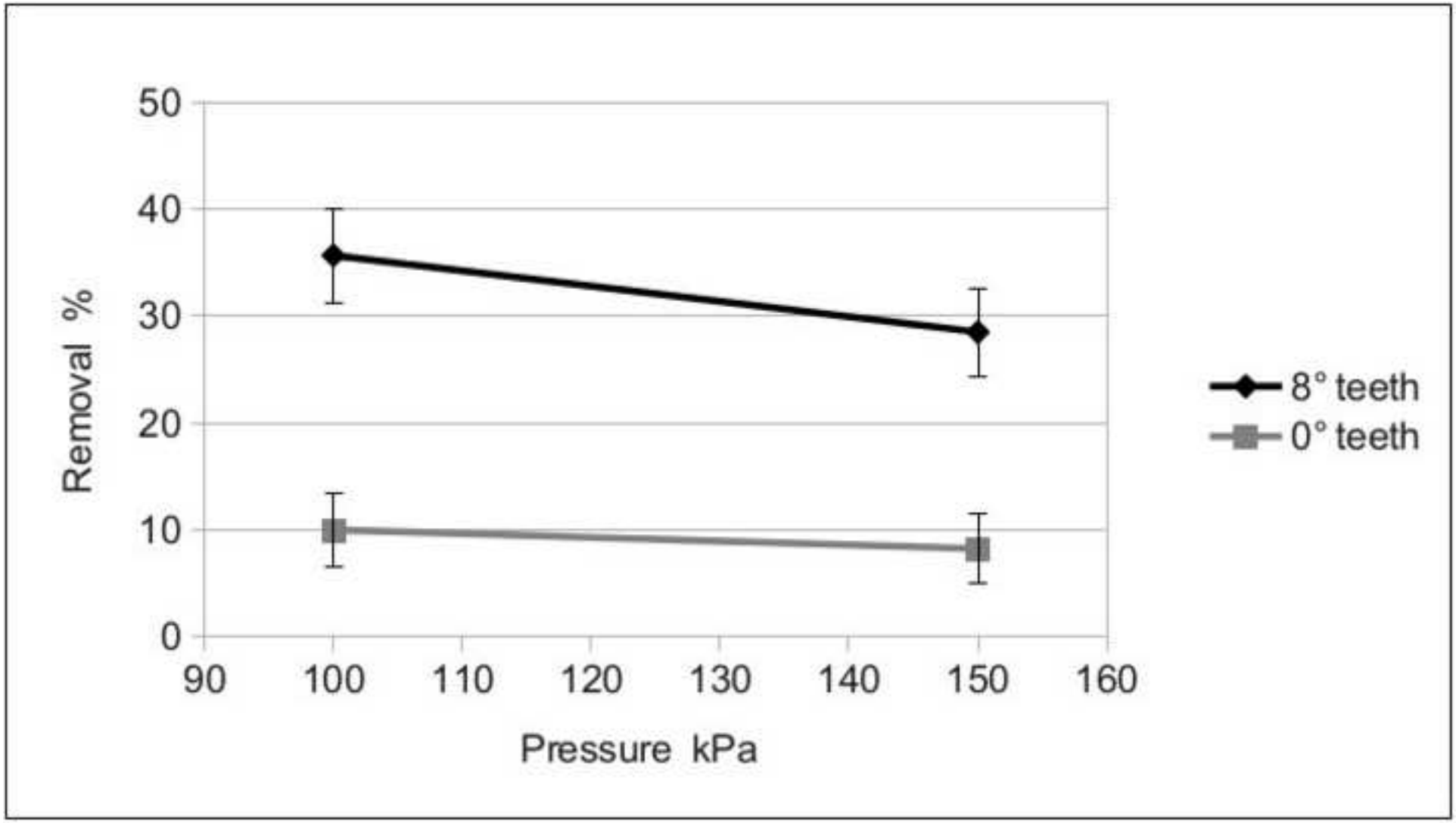

Figure_11

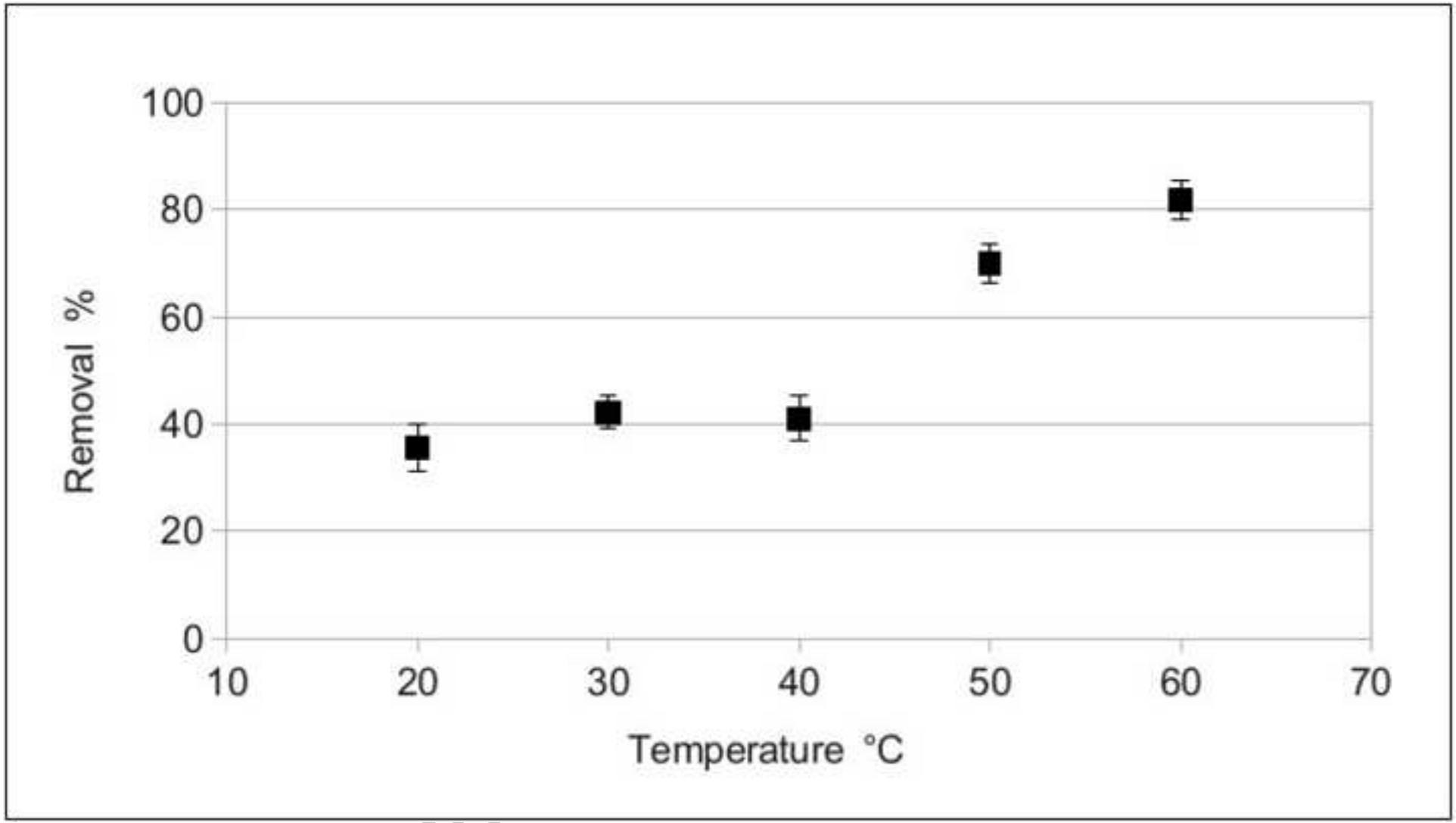

Figure_12

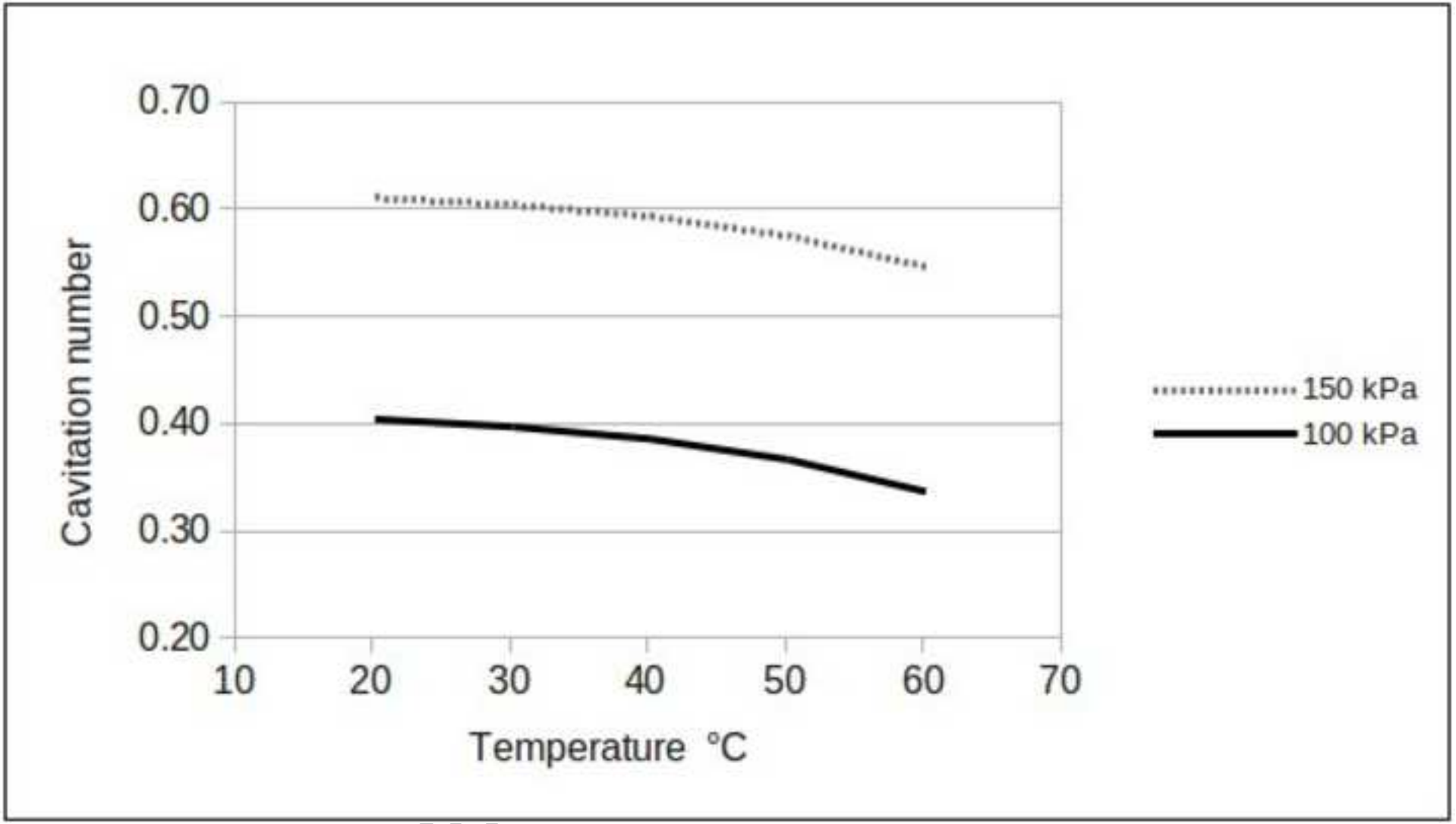

Figure_13

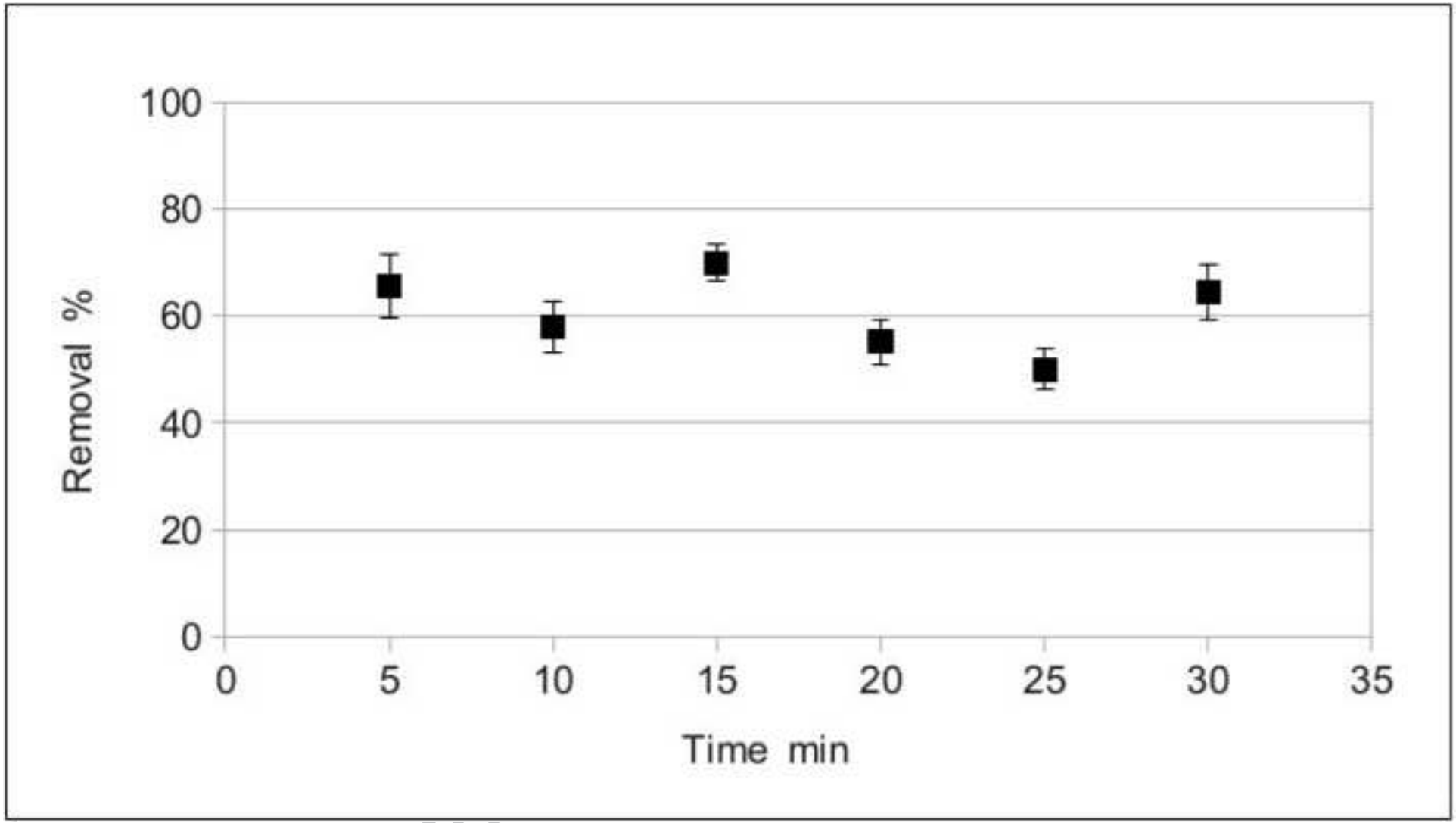

Figure_14