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# Rheological and tribological behavior of polyacrylamide-base solutions for artificial synovial fluid

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*Purpose*: This paper presents tribological and rheological analysis results of artificial synovial fluid base solutions. Special attention was paid to polyacrylamide preparations with different molecular weights and concentrations. *Methods*: Tribological tests were conducted using the $Al_2O_3$–CoCrMo friction pair in the presence of investigated lubricants. Confocal microscopy was used to analyze and assess of volume, depth, and width of wear traces. Moreover, the viscosity and viscoelasticity tests of analyzed solutions were carried out. The rheological measurements were focused on the oscillatory tests, which allowed us to determine the elasticity modulus (G′) and viscosity (G″). *Results*: Viscoelastic nature of the tested preparations depends on the strain rate. It has been shown that elastic properties dominate at higher frequencies. The molecular weight of the polymer has a particular influence on these properties. The most promising results were obtained for 6% and 8 % high molecular weight polyacrylamide compositions. *Conclusions*: However, all tested polyacrylamide solutions show better rheological and tribological characteristics than commercial preparation based on hyaluronic acid.

*Key words*: *artificial synovial fluid, rheology, viscoelasticity, friction, wear*

## 1. Introduction

All tribosystems, both in technology and nature, consist of at least two contacting bodies moving toward each other. The system's operation is mainly responsible for the movement and load transfer between the components. Various lubricants are commonly used to reduce resistance to motion and wear [17]. It is crucial in the case of complex biotribological systems, such as human joints, which cannot function properly without proper lubrication. The function of a lubricant is performed by a synovial fluid, which keeps the friction coefficient at a very low level. In the case of the knee joint, the resistances to motion reach values 0.001–0.03 [17], [35]. Because of the complex structure of the joints, the natural lubrication mechanisms are complicated to determine. There are many theories of joint lubrication. However, hydrodynamic and boundary film lubrication mechanism is fundamental [23], [27]. Proper lubrication is responsible for the optimal functioning of the entire human musculoskeletal system. Physicochemical and rheological properties of the synovial fluid significantly impact the quality of joint lubrication [24], [25]. Synovial fluid has viscous and elastic properties. The dominance of one over the other depends mainly on the shear rate. Therefore, during joint exploitation, these properties may change [16], [39]. The viscous properties of the synovial fluid are responsible for dissipating mechanical energy in the form of heat. Elastic properties, on the other hand, are responsible for the storage of mechanical energy [6]. Disorders caused by joint injuries or diseases often result in undesirable changes in chemical composition and secretion of synovial fluid. This often leads to disturbances in the biotribological system and limits biofunctionality [15], [21]. The solution to this problem is viscosupplementation treatments, which inject a synovial fluid replacement directly into the articular cavity. Artificial

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synovial fluids are designed to support and restore damaged joints' proper functioning [10]. For this purpose, substances with high biocompatibility are usually used. An example is hyaluronic acid, a component of almost all popular compositions of artificial synovial fluid. Most commercial preparations very often differ only in the concentration, molecular weight of the acid, and type of functional additives. Usually, the reliability of such substitutes is short-term and is associated with frequent repetition of the viscosupplementation treatment [1], [11], [19]. However, in recent years, more and more attention has been paid to compounds based on liquid crystals, especially cholesterol esters, which are characterized by very good anti-wear properties [14], [37]. There are also literature reports on synthetic polymers used in joint injections. Polyacrylamide is an example of such a biopolymer. Due to its beneficial utility properties and high lifetime prediction, it can adequately replace the most common commercial preparations based on hyaluronic acid [29], [38]. Polyacrylamide hydrogels are increasingly used in modern treatments of osteoarthritis in animal [8], [36] and human therapies [4], [34].

In this context, our study is aimed to analyze the viscoelastic and tribological characteristics of polyacrylamide hydrogels as a base for a composition of artificial synovial fluid.

## 2. Materials and methods

Rheological and tribological tests were carried out in the environment of polyacrylamide-base synovial fluid preparations. For comparative purposes, Hyalgan, which is one of the most commonly used hyaluronic acid-based commercial artificial synovial fluid preparation, was also used for the tests. Compositions of polyacrylamide hydrogels are presented in Table 1.

All compositions were prepared one day before the test and stored in a laboratory incubator at 37 °C to ensure conditions similar to the human body. pH values were determined using a Clarytrode 120 electrode (Hamilton, Giarmata, Romania) in conjunction with the multifunctional S80-K SevenMulti ionic conductivity meter (Mettler Toledo, Greifensee, Switzerland). pH measurements were performed on the compositions prepared in larger quantities, which were divided into smaller samples for tribological and rheological tests. The pH of the preparations was constant as confirmed by triplicating.

### 2.1. Rheological tests

The viscoelastic properties of polyacrylamide preparations were carried out using Rheostress 6000 rheometer (Thermo Fisher Scientific, Waltham, MA, USA) at a temperature of 37 ± 1 °C. The measurements were realized in a parallel plate system (Fig. 1) where the diameter of the upper plate was 35 mm and the gap between plates was 0.8 mm.

The rheological measurements were focused on the oscillatory tests, which allowed to determine the elasticity modulus ($G'$) and viscosity ($G''$) at a constant shear strain value $\gamma = 0.01$. The RheoWin computer software was used to register the measurements.

### 2.2. Tribological tests

Tribological tests were carried out using a UMT TriboLab tribometer (Bruker, Billerica, MA, USA), with a ball-on-disc system, presented in Fig. 2.

Ball specimens with a diameter of 6 mm were made of corundum ($Al_2O_3$). Cylindrical samples with a diameter of 8 mm and thickness of 5 mm were made of a CoCrMo metal rod and subjected to mechanical grinding with sandpaper grades 1500, 2000 and 2500. Next, the specimens were polished with $Al_2O_3$ powder until obtained a mirror-like surface. Before tribological measurements, disc and ball specimens were rinsed for 10 minutes in water and ethanol in an ultrasonic cleaner. The chemical composition of metal specimens is presented in Table 2.

Table 1. Chemical composition of polyacrylamide hydrogels

| Composition | Dissolved substance | Concentration [wt %] | Solvent |
|---|---|---|---|
| PAML 4% | Low molecular weight polyacrylamide average Mn 150.000 (SIGMA ALDRICH, Product number 749222) | 4 | deionized water |
| PAML 6% | | 6 | |
| PAML 8% | | 8 | |
| PAMH 4% | High molecular weight polyacrylamide average Mn 6 000 000 (SIGMA ALDRICH Product number 92560) | 4 | deionized water |
| PAMH 6% | | 6 | |
| PAMH 8 % | | 8 | |

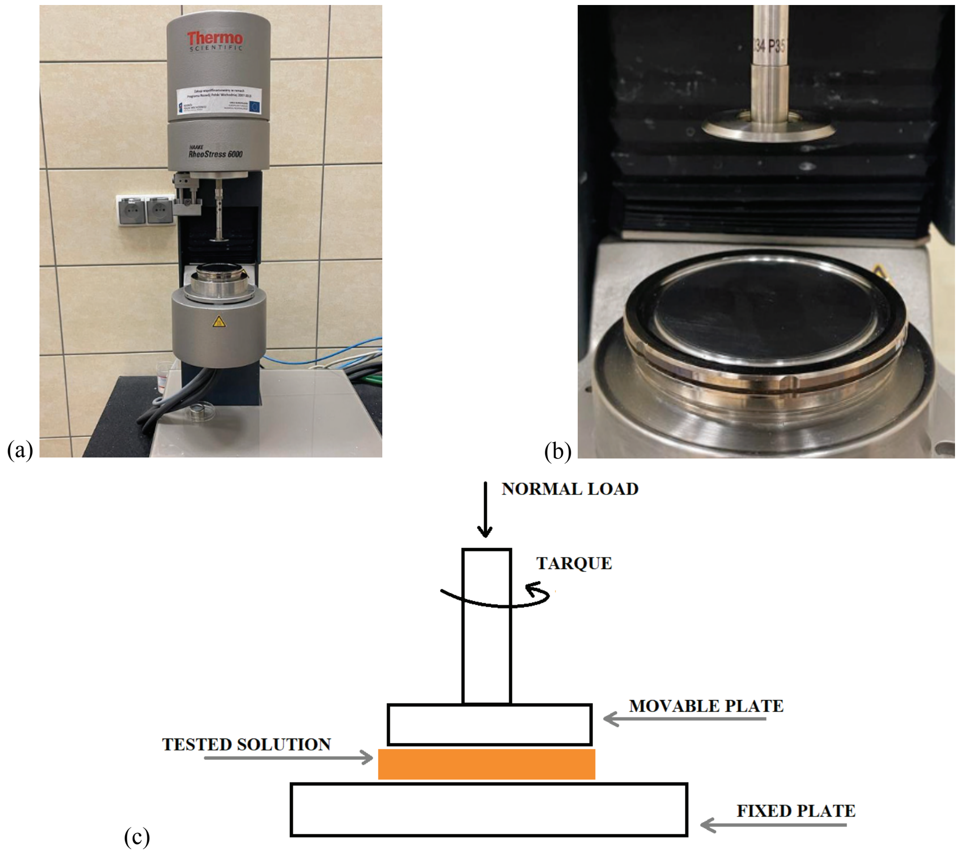


Fig. 1. (a) Rheometer Rheostress 6000, (b) parallel plate system, (c) scheme of rheological measurement system

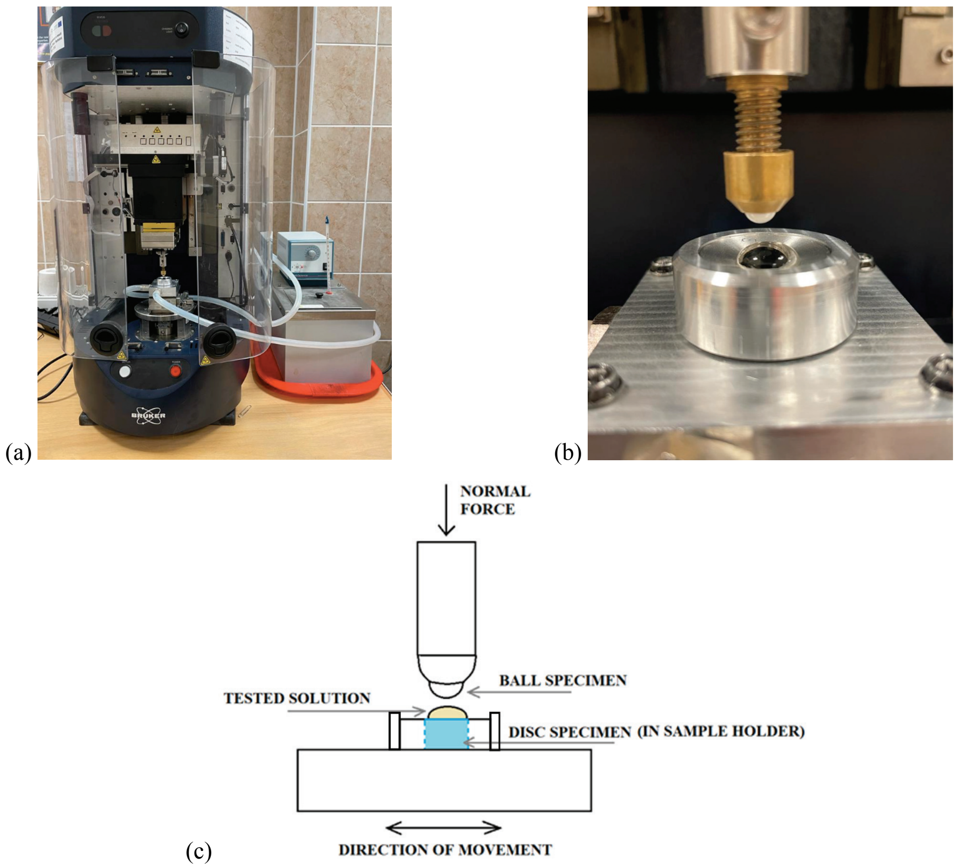


Fig. 2. (a) Tribometer Bruker UMT, (b) friction node, (c) tribological system scheme

Table 2. Chemical composition of CoCrMo alloy (maximum content as a wt. %)

| Co | Cr | Mo | Mn | Si | C | Al | Zr | Ti |
|---|---|---|---|---|---|---|---|---|
| Remainder | 27.720 | 5.780 | 0.650 | 0.370 | 0.036 | <0.020 | <0.010 | <0.010 |

The wear test was conducted in various polyacrylamide-base synovial fluid preparations (Table 1) at room temperature (21 ± 1 °C) at a constant frequency of 1.0 Hz for 3600 cycles. The system displacement amplitude was 500 μm. The ball specimen was pressed against the disc's surface at normal force $F = 5N$. For statistical purposes, every test was repeated three times.

The wear tracks formed after tribological tests were subjected to microscopic observations using the LEXT OLS4000 confocal microscope (CLSM, Olympus, Tokyo, Japan). The 3D imaging feature of CLSM allows us to measure the volume, depth, and width of wear tracks. The laser in the microscope scans horizontally, but it does so layer by layer. The scan step pitch was 0.05 μm. The size of the scanned surface including the place of friction was 1400 × 480 μm. The images were taken without any filters. The details of the measurement method were described in our previous publication [9].

# 3. Results

pH measurement studies have shown that low molecular weight polyacrylamide compositions have a more acidic pH in the range of 4.1–4.3. On the other hand, compositions based on high molecular weight polyacrylamide show more neutral pH values around 7.2–7.4, which is much closer to the commercial preparation, where the pH value is equal to 7.47 (Fig. 3).

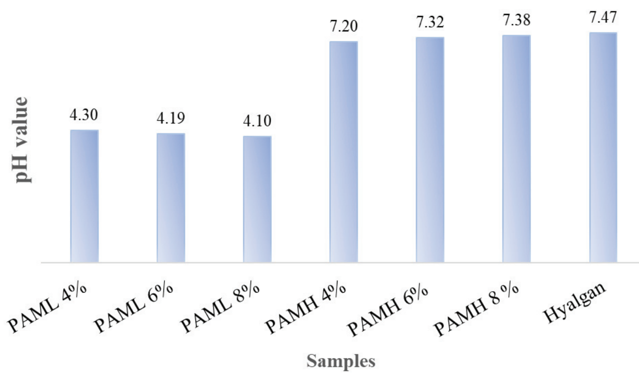


Fig. 3. pH values of tested compositions

## 3.1. Rheological test

The lowest viscosity (Fig. 4a) was observed for the lower molecular weight compositions, particularly PAML 4%. On the other hand, for preparations with a high molecular weight, a higher viscosity was obtained (Fig. 4b). However, both types of preparations differ from each other. Preparations based on low-molecular polyacrylamide show more stable characteristics, while the decrease in viscosity is much more rapid in the case of high-molecular polyacrylamide.

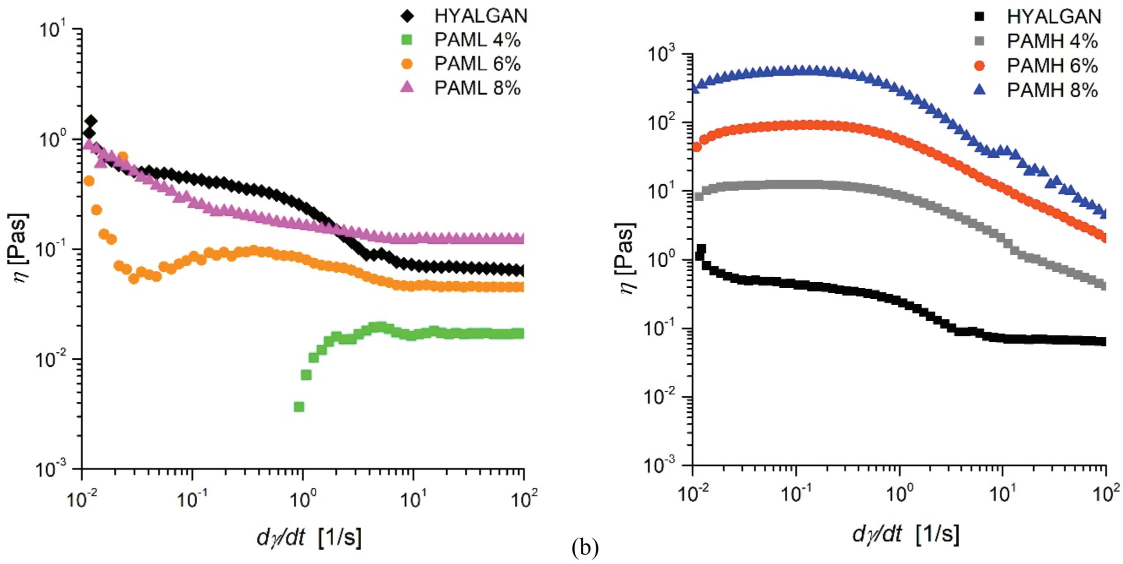


Fig. 4. Viscosity of HYALGAN and solutions based on it: (a) PAML, (b) PAMH

Nevertheless, for all formulations, the same relationship was obtained, that the viscosity decreases with increasing shear rate.

The oscillatory tests were also performed. Obtained results show the relationship between the storage modulus G′ and the loss modulus G″ as a function of frequency. In all tested solutions, the advantage of the elastic modulus G′ over the viscous modulus G″ can be seen (Figs. 5, 6).

The oscillatory test results are presented in two distinct areas. The first area (yellow) shows the characteristics at low frequency. The second area (blue) represents the higher frequency range. A sharp *G*′ value jump occurs between both regions, mainly for PAML and hyalgan preparations. The $a_1$ and $a_2$ are parameters of the curve's slope in a given area. Higher values of the "a" parameter result in a more rapid change in the modulus of elasticity. Obtained results show that curves for the preparations based on low molecular weight polyacrylamide and the commercial preparation of Hyalgan are similar (Fig. 5). However, the high-molecular polyacrylamide curves are smooth, which proves that preparations are more stable at tested frequencies (Fig. 6). PAMH structure is more bound, which ensures greater elasticity and viscosity of solutions. All analyzed compositions show a similar tendency that with

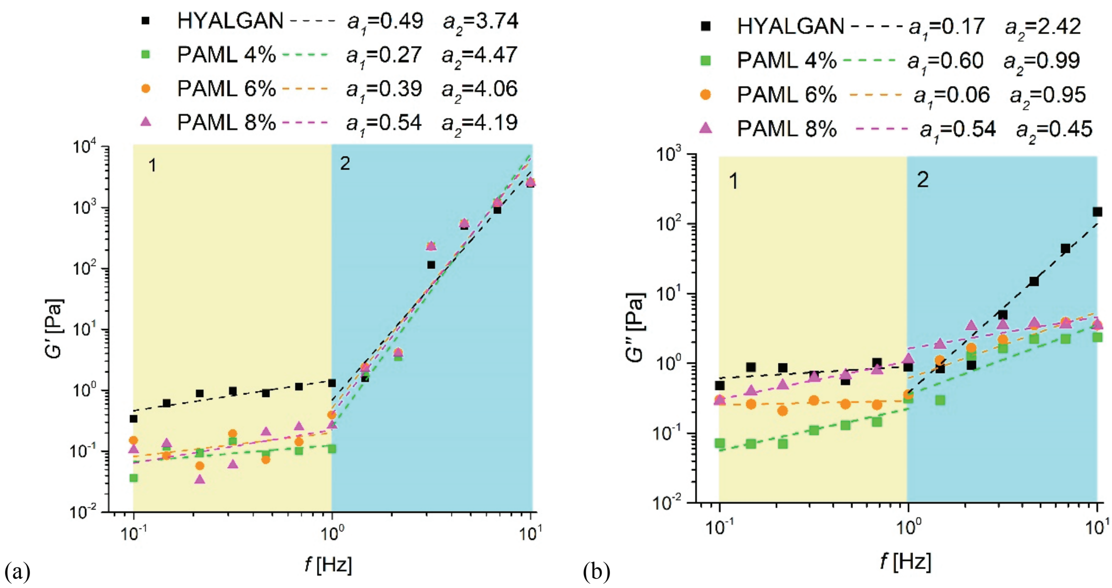


Fig. 5. a) storage modulus *G*′ and b) loss modulus *G*″ as a function of the oscillating frequency *f* for preparations PAML and HYALGAN, $a_1$ and $a_2$ – slope parameters

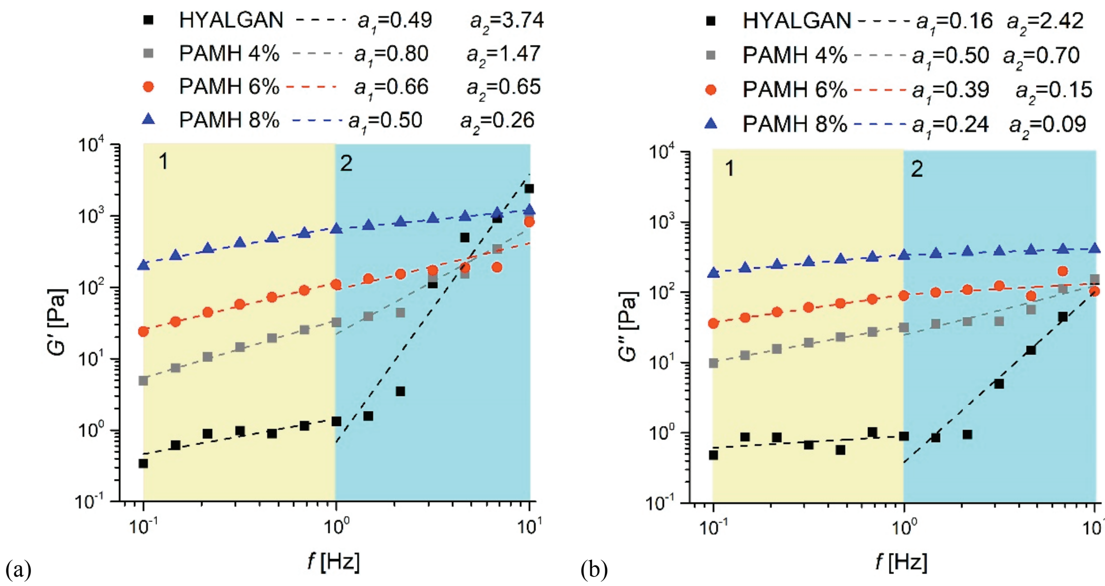


Fig. 6. a) storage modulus *G*′ and b) loss modulus *G*″ as a function of the oscillating frequency *f* for preparations PAMH and HYALGAN, $a_1$ and $a_2$ – slope parameters

increasing frequency (shear rate), the elasticity of the solutions also increases. At low-frequency values, there are fluctuations where the elastic modulus alternates with the viscous modulus. However, the $G''$ modulus dominates over the $G'$ modulus, which indicates viscous properties. The opposite situation can be observed at higher frequencies.

## 3.2. Tribological test

The tribological test made it possible to determine the coefficient of friction (COF) for kinematic pairs in the presence of lubricants based on PAML, PAMH, and commercial preparation Hyalgan. The results of friction tests are shown in Fig. 7.

Results obtained for tribological systems where a lubricant was used show that the resistance to motion values oscillate within the range of 0.2–0.3. The proposed compositions of artificial synovial fluid based on polyacrylamide show slightly more favorable lubricant properties compared to the commercial preparation Hyalgan. Furthermore, the tendency of the curves for the different concentrations of PAML (Fig. 7a) and PAMH (Fig. 7b) is very similar. The lowest resistance to motion occurred for 6% solutions, which seem to be the most promising compositions. Nevertheless, the differences in the obtained values are minor. It can be seen that the most unstable resistances to motion occur in conditions of dry friction (Fig 7c). In this case, the coefficient of friction is also the largest. At the beginning of the test, the coefficient of dry friction reaches a value of 0.4, while over time increases rapidly and reaches a maximum value of 0.7. All tested lubricants significantly reduce friction, but the most satisfactory results were obtained for polyacrylamide preparations.

Confocal images of wear tracks have been compared and particularly analyzed to evaluate wear. Selected images of wear tracks are presented in Fig. 8.

The most pronounced traces were observed for dry friction (Fig. 8a). Moreover, darker spots can be seen on the friction marks in the environment of high molecular weight polyacrylamide. Rinsing of wear products accumulated at the edges of wear tracks is more difficult because of the higher viscosity of lubricant

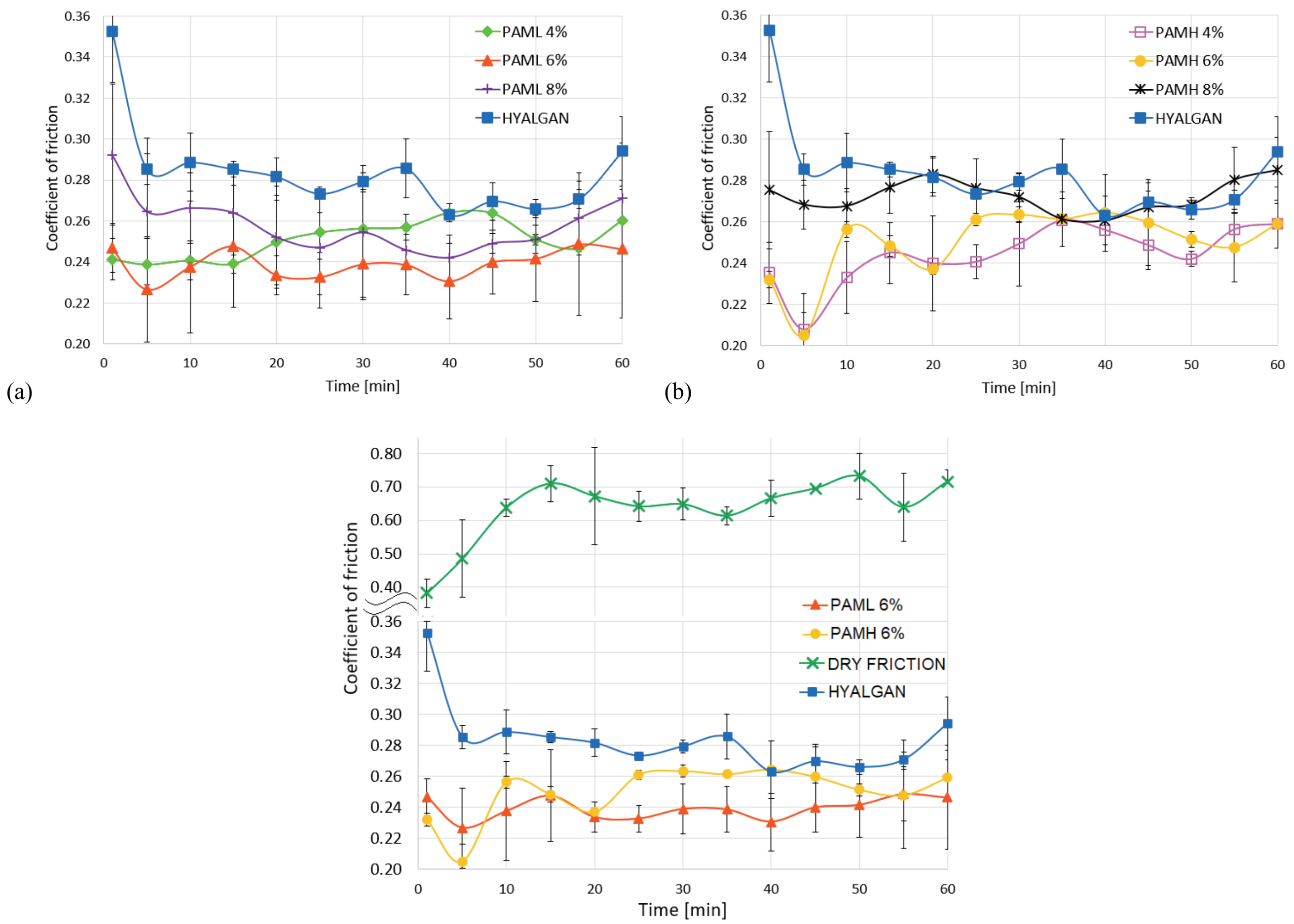


Fig. 7. Coefficient of friction as a function of time for: (a) PAML, (b) PAMH, (c) 6% composition

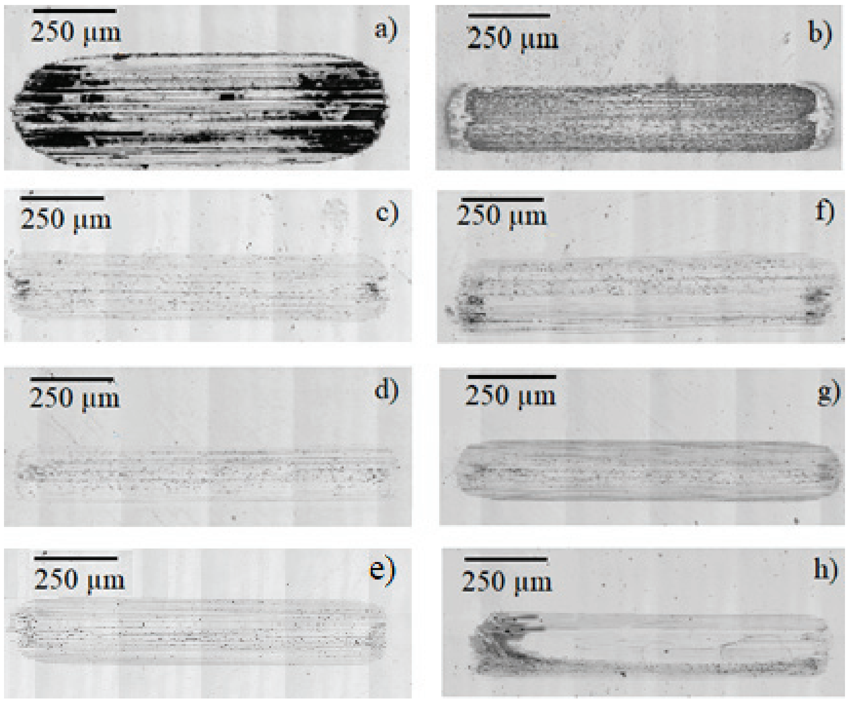


Fig. 8. Wear tracks for a) dry friction, b) Hyalgan, c) PAML 4%, d) PAML 6%, e) PAML 8%, f) PAMH 4%, g) PAMH 6%, h) PAMH 8%

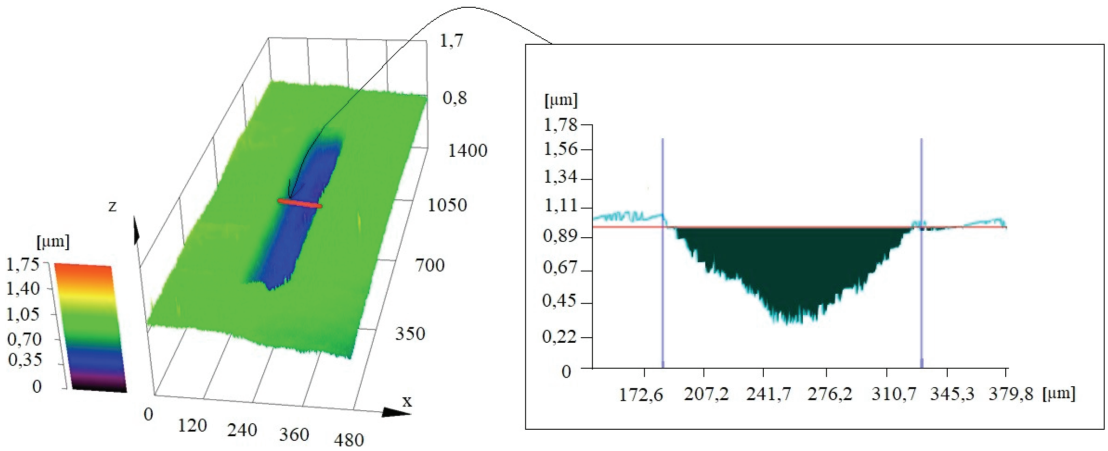


Fig. 9. Profile of the CoCrMo surface after wear test in the environment of PAML 4%

(Fig. 4). In the case of the PAML environment, the clustering of wear particles is much lower. 3D computer analysis of microscopic images makes it possible to visualize the morphology of wear areas. An example of a three-dimensional analysis is shown in Fig. 9.

Based on the obtained profiles, the friction marks' depth, width, and volumetric wear were determined. The depth and width were measured at the center of the wear marks, as shown in Fig. 9. The summarized measurement data is shown in Fig. 10.

Trends in all parameters measured are mostly similar, and the volume depends on the wear trace's depth and width. The largest volume of the friction mark occurs under dry friction conditions (approximately $32 \cdot 10^4$ μm$^3$). On the other hand wear of the samples with PAM lubrication was smaller (about $5–6 \cdot 10^4$ μm$^3$). Also, all of the proposed compositions of polyacrylamide gels had better lubricating properties than the commercial preparation based on hyaluronic acid. However, the most promising results were obtained for PAMH 4% and PAMH 6% preparations. Even despite the limited rinsing of wear products from the friction node, the lubricant effectively limited the wear of the samples. The lubricant properties of PAML 6 % are also satisfying, but the pH of PAML solutions is too acidic (Fig. 3) for the human body.

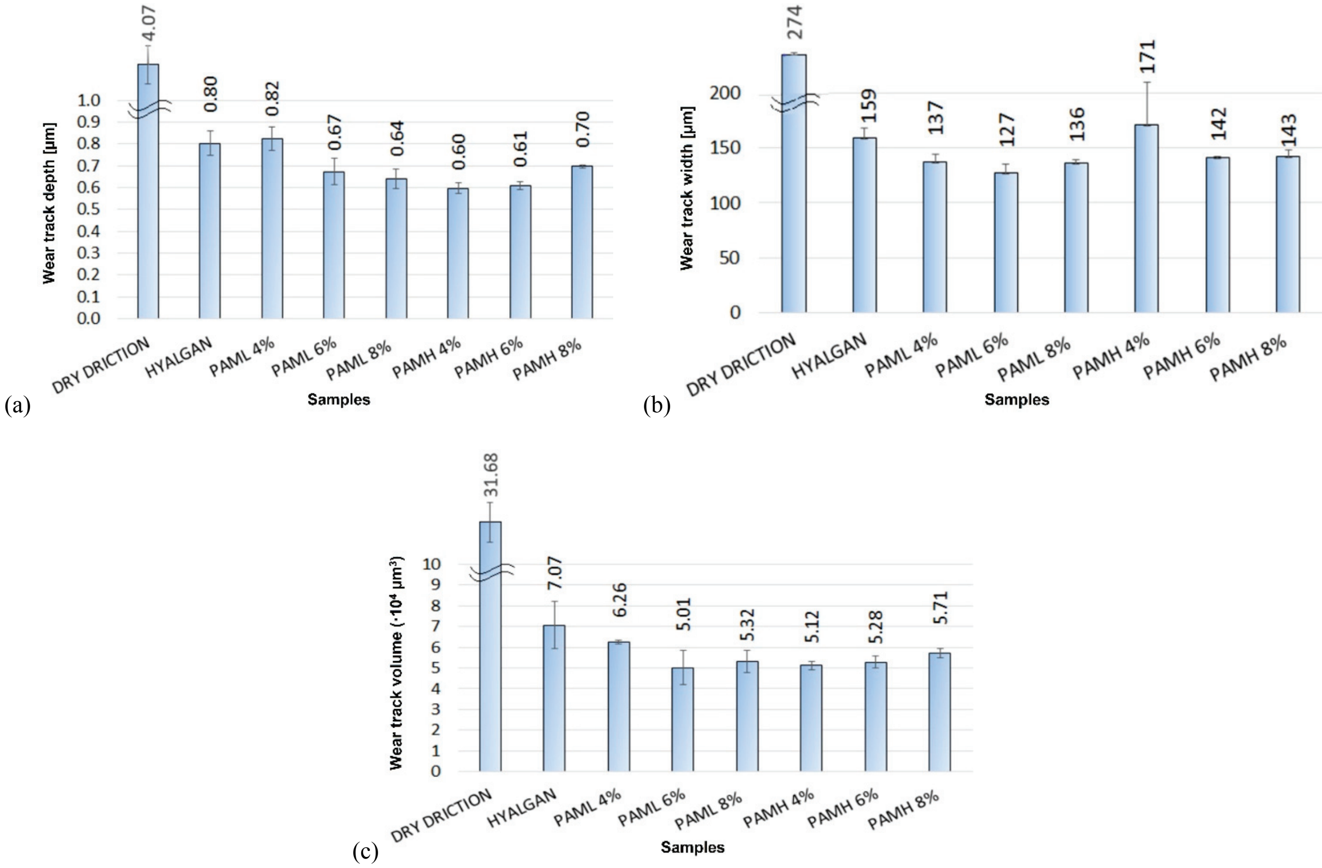


Fig. 10. Parameters of wear tracks: (a) depth, (b) width, (c) wear volume

# 4. Discussion

Literature reports indicate that the natural synovial fluid's pH is 7.31–7.64 [33]. Commercial synovial fluid formulations usually have a neutral or slightly alkaline pH. This is especially important to maintain the overall homeostasis of the body. The tested preparations (Fig. 3) based on high molecular weight polyacrylamide (PAMH) showed a pH close to the natural synovial liquid. In the case of low molecular weight polyacrylamide (PAML), lower pH values of solutions may result from the initiator type and conditions of the polymerization process [5].

The rheological test shows that compositions based on low molecular weight polyacrylamide assume similar viscosity values compared to the commercial preparation (Fig. 4). These values are also close to the viscosity of the natural synovial fluid, which oscillates around 0.5 Pas. [2]. However, the viscosity of synovial fluid depends on many factors like gender, health condition, age, and weight of the human body. All of them influence the chemical composition of body fluids. For this reason, Simou et al. [31] and Jebens et al. [18] in their works presented a much wider range of viscosity values (0.06 to 1.4 Pas). Research results conducted by Drumeanu A.C. [12] also suggest the high viscosity of lubricants based on polyacrylamide**.** In the research conducted, the results most similar to those presented by other scientists were obtained with the PANH 8% preparation. For all preparations, the same relationship was obtained that viscosity decreases with increasing shear rate. This phenomenon is characteristic of non-Newtonian shear-thinning formulations. Similar behavior of fluids is described in the works of Lospichl [20], Rebenda [25], and Bhuanantanondh [3]. Research results conducted by Drumeanu [13] and Sato et al. [30] also confirm that viscosity increases with the molecular weight and concentration of polyacrylamide.

In the study of viscoelasticity the G″ modulus dominates over the G′ modulus, which indicates viscous properties (Figs. 5, 6). The opposite situation can be observed at higher frequencies. The predominance of *G*′ over *G*″ indicates elastic behavior [22], [32]. During the low-frequency, oscillatory motion, where longer time scales occur, the linear molecular chains break down, and strain is reduced. However, at higher frequencies, where the time scales are relatively smaller, chains of polyacrylamide cannot unravel so easily. This proves the advantage of the storage modulus over the loss modulus and improves the elastic properties of the preparations [25]. Rheological tests on polyacrylamide were also performed by Pogoda [6]. Many similarities can be seen, mainly in rheological curves in the oscillation test and analogous relationships related to polyacrylamide concentration. The stiffness and elasticity of the composition increased with the polymer concentration. These properties also depend on the degree of cross-linking of the polymer [6], [7].

Tribological tests also confirm that the use of lubricants improves the conditions in the friction node by reducing the coefficient of friction (Fig. 7). Ruggiero et al. [28] also investigated the lubricant properties of Hyalgan. Obtained results show that the COF increases with time during dry friction conditions, unlike rubbing with lubricant. Assessing wear is very useful in any tribological system. It should be emphasized that the determination of basic wear parameters, i.e., COF, depth and volume of wear tracks, makes it possiblea to evaluate the effectiveness of lubrication in the tested measuring system. In the case of artificial synovial fluids, ensuring good lubrication is particularly important because it directly affects the health and comfort of patients and extends the reliable usage of natural joints. Similar tribological studies on the influence of molecular weight and polyacrylamide concentration were conducted by Drumenau [12]. Rudge et al. [26] confront the tribological properties of gelatin-base hydrogels to polyacrylamide solutions. The subject test confirmed the effectiveness of polyacrylamide, which achieved significantly lower friction coefficient values than gelatin. The validity of using aqueous polyacrylamide solutions was also confirmed by Wang et al. [40]. The obtained results show that polyacrylamide hydrogel fragments can form a lubricating layer between the rubbing surfaces, reducing friction and wear. Effective lubrication is essential for resistance to motion and wear reduction for all tribological systems. However, in biotribological systems, biocompatibility should be an important factor for selecting lubricant components.

# 5. Conclusions

The results of obtained studies confirmed favorable rheological and tribological properties of polyacrylamide-base solutions. The following conclusions can be drawn:

- pH values of preparations based on high molecular weight polyacrylamide were the most similar to natural human synovial fluid, which oscillates between 7.2–7.4;
- the viscoelastic nature of the tested preparations depends on the strain rate. Viscous properties are dominant in larger deformations (frequency up to 1 Hz). However, solutions show more elastic properties at a higher frequency where the deformation is smaller. This may result in the formation of a support layer on the articular surfaces;
- viscosity and viscoelasticity of PAM solutions in most cases increased with polymer concentration. The viscosity of PAMH solutions was much higher and was in the range of 10–200 Pas, while preparations based on PAML reached a viscosity of 0.2–0.8 Pas;
- all of the tested solutions show better tribological and rheological characteristics compared to the commercial preparation based on hyaluronic acid;
- the most promising lubricant properties were obtained by high molecular weight polyacrylamide preparation, especially PAMH 4% and PAMH 6%. That, along with high biocompatibility, makes them a good starting point for future investigation into developing new compositions of artificial synovial fluid.

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