Some considerations concerning four-ball machine testing of the polyacrylamide solutions

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Abstract. Polyacrylamide (PAM) is one of the most widely used and technically important water-soluble polymers. Polyacrylamide (PAM) is usually obtained by free radical polymerization of acrylamide (AM) and in its partially hydrolysed form is a synthetic straight-chain polymer of acrylamide monomers, some of which have been hydrolysed. The structure of HPAM molecule is a flexible chain. This kind of structure is known as a random coil in polymer chemistry. Due to the hydrolysed groups contained in its molecule, HPAM has multiple charges distributed along the chain that make it a polyelectrolyte. The paper presents the experimental results concerning the lubricant solutions based on polyacrylamide behaviour when were tested on the four ball machine. It has to be mentioned that this kind of polymer was not used until now in lubrication and the studies concerning its tribological behaviour are at the beginning.

1. Introduction

The synthetic lubricants utilization, especially of those liquids, was developed in the last two decades because of the new fields development such as aerospace technique, nanotechnology, cryogenic technologies, medical technique etc. In the same time the liquid lubricants based on the mineral oils showed theirs performance limits. This fact required new researches for theirs substitution.

Generally the synthetic lubricants have a good oxidation stability, a wide service temperature range, a high viscosity index and a good plastic compatibility, so that this type of products are possible alternatives for lubrication of machine elements.

Polyacrylamide (PAM) is one of the most widely used and technically important water soluble polymers. Polyacrylamide (PAM) is usually obtained by free radical polymerization of acrylamide (AM) and in its partially hydrolysed form is a synthetic straight-chain polymer of acrylamide monomers, some of which have been hydrolysed (figure 1) [1,2,3,4].

The normally available PAM often contains about 1 to 3 mole % carboxylate groups as an impurity. Because of this fact, for the researches presented in this paper it was considered HPAM, with different hydrolysis levels [2,5,6].

Some physical properties of HPAM such as polymer adsorption, shear stability and thermal stability are influenced by the hydrolysis degree (HD). However, although commercial polymers are supplied with a stated degree of hydrolysis, it is well known that at elevated temperatures the hydrolysis of the amide will continue [5,7].
The primary chain structure of polyacrylamide (PAM) and partially hydrolysed polyacrylamide (HPAM) and its corresponding sodium salt.

Figure 1. The primary chain structure of polyacrylamide (PAM) and partially hydrolysed polyacrylamide (HPAM) and its corresponding sodium salt.

The structure of HPAM molecule is a flexible chain. This kind of structure is known as a random coil in polymer chemistry. Due to the hydrolysed groups contained in its molecule, HPAM has multiple charges distributed along the chain that make it a polyelectrolyte. Generally, the polyelectrolytes are distinguished from non-ionic polymers by the large effects, which change in salt concentration, and pH has on their viscosities. These changes in viscosity, respectively its decrease with salt concentration increase, appear due to the interactions between the fixed charges along the chain and the mobile ions in solution. Because the necessity of lubrication can occur in different conditions, we considered opportune to take into account this behaviour. Also, the polyelectrolyte character of HPAM makes it to interact quite strongly with the metallic surfaces [6,7,8].

The paper presents the experimental results concerning the lubricant solutions based on polyacrylamide behavior when were tested on the four ball machine. It has to be mentioned that this kind of polymer was not used until now in lubrication and the studies concerning its tribological behavior are at the beginning [8,9,10].

2. Experimental conditions
There were tested six lubricant solutions based on polyacrylamide. The lubricant solutions were made using four polymers which had the main characteristics presented in table 1.

Table 1. The characteristics of the studied polymers.

| Polymer type | Characteristics | Molecular weight, MW, [g/mol] | Hydrolysis degree, HD [%] |
|--------------|-----------------|-------------------------------|--------------------------|
| P1           |                 | 3·10^6                        | 8.3                      |
| P2           |                 | 4·10^6                        | 8.6                      |
| P3           |                 | 7·10^6                        | 8.7                      |
| P4           |                 | 10·10^6                       | 8.6                      |

The lubricant solutions were made using different solvents based on tap water and NaCl salt. The dynamic viscosity of the lubricant solutions was determined with the Brookfield viscometer. The characteristics of used lubricant solutions are presented in table 2, and the chemical composition of the used tap water are showed in table 3.

The tests have been carried-out on a four ball machine with a classical construction configuration. The tests were performed using balls made from rolling contact bearing steel RUL1-V (AISI 52100) type. The main characteristics of these balls are: diameter – 12.7 mm; surface roughness Ra – 0.02 μm; hardness HRc - 58.

The test balls are so arranged that their centers form a pyramid with a nominally equilateral base. Each of the lower balls forms three conjunctions – one with the top ball and two with the ball pot. The top ball forms three conjunctions also, one with each of the three lower balls. The upper ball was
rotated with a constant speed of 1500 rpm (0.998 m·s\(^{-1}\) sliding speed). For each test, the four-ball machine ran 60 seconds, and was used new balls and new lubricant sample.

**Table 2.** The characteristics of the lubricant solutions.

| Solution type | Polymer type | Polymer concentration [wt.%] | Dynamic viscosity [Ns/m\(^2\) x10\(^{-3}\)] | Solvent          |
|---------------|--------------|-------------------------------|--------------------------------------------|------------------|
| S1            | P1           | 1.0                           | 240                                        | tap water        |
| S2            | P2           | 1.0                           | 320                                        | tap water        |
| S3            | P3           | 1.0                           | 530                                        | tap water        |
| S4            | P4           | 1.0                           | 1450                                       | tap water        |
| S5            | P3           | 1.0                           | 250                                        | tap water + 2% NaCl |
| S6            | P3           | 1.0                           | 190                                        | tap water + 5% NaCl |

**Table 3.** Chemical composition of the tap water used as solvent.

| Chemical element | NO\(_2^-\) | SO\(_4^{2-}\) | Cl\(^-\) | K | Ca | Mg | Na |
|------------------|-----------|---------------|---------|---|----|----|----|
| Concentration [mg/l] | 18 | 40 | 98 | 1.6 | 83 | 10 | 57 |

Before and after each test the four-ball device elements and the testing balls were cleaned with water and dried in a warm air draught. A constant quantity of 20 milliliters of lubricant solution was used for each test, so all the balls were completely covered by lubricant. The bulk temperature of the lubricant solution sample was measured with a thermocouple, mounted at the bottom side of the balls pot. The lubricant solution samples temperature during all tests was maintained at about 30…40°C, no important changes being observed in the lubricant bulk temperature, due to the short time tests. The wear spot diameter of the balls was measured using a laboratory microscope.

During each test there were measured and determined the following parameters:
- the axial load applied to the upper ball;
- the friction force that appears between the upper ball and the lower balls;
- the wear spot diameters of the lower balls determined before the welding;
- the axial load which corresponds to the balls welding;
- the friction force value before the welding.

3. Results and discussions

Regarding the influence of the molecular weight and the solvent on the dynamic viscosity of the lubricant solutions it can be observed in figure 2 and figure 3 that dynamic viscosity increases with the molecular weight increase and decreases with the NaCl content increase.

The behavior of the lubricant solutions during the tests is presented in figure 4 and figure 5. It can be observed that the friction force has different variations as a function of lubricant solution composition. For solutions S1, S2, S3 and S4 the friction force increases with the increase of the axial load in the range of 100…300 N. The same tendency can be remarked for solutions S1, S2, S4 and axial load range of 400…1100 N where for solution S3 the friction force decreases with axial force increase. Also, from figure 4, it can be observed that the friction force increases with the increase of polymer molecular weight, except solution S3 for which the friction force has a minimum value.
**Figure 2.** Lubricant solutions dynamic viscosity vs. molecular weight (solutions S1, S2, S3, S4).

**Figure 3.** Lubricant solutions dynamic viscosity vs. NaCl content in solvent (solutions S3, S5, S6).

**Figure 4.** The dependence between the friction force and the axial load for the tested lubricant solutions (solvent – tap water).

**Figure 5.** The dependence between the friction force and the axial load for the tested lubricant solutions (solvent – tap water + NaCl).
In figure 5 it is represented the influence of NaCl on the friction force, for different axial load values and for the same polymer type. For solution S3 (solvent - tap water) the lowest values of the friction force are registered for the highest values of the axial load, while for the solutions S5 and S6 the influence of NaCl content and the axial load does not influence significantly the friction force. For solution S6 (solvent - tap water + 5wt.%NaCl) the friction force values are a bit smaller compared to those registered for solution S5 (solvent - tap water + 2wt.%NaCl), and they have a decrease tendency with axial load increase. Also, it can be remarked for all the lubricant solutions that the scuffing appearance was started at about 1000 N axial load.

Figure 6. The dependence between axial load before welding and the molecular weight (solvent – tap water, polymer concentration - 1.0 wt. %).

Figure 7. The dependence between axial load before welding and the NaCl solvent content (polymer molecular weight - 7·10^6 g/mol, polymer concentration - 1.0 wt. %).

For a better understanding of the influence of the solutions characteristics on their lubrication properties in figures 6 and 7 were presented the influences of molecular weight and the NaCl content on the axial load value registered before welding. Thus in figure 6 it can be observed that the axial force before welding increases with the polymer molecular weight increase for the lubricant solutions made with tap water solvent and 1.0 wt.% polymer concentration.

The influence of the solvent composition on the axial load before loading is presented in figure 7 for the same polymer (P3) and polymer concentration (1.0 wt.%). From figure 7 results that the increase of the NaCl solution content implies the axial load before welding increase. The maximum value of axial load before welding was determined for the solution S6, even if for this some of the biggest values of the friction force were registered.

To determine the optimum characteristics of the lubricant solution based on polyacrylamide it has to do the correlations between the wear and the solutions characteristics and also between the wear and the loadings. Thus, in figure 8 it is represented the dependence between the wear spot diameter (calculated like the average of the wear spot diameters of the three lower balls) and the molecular weight of the polymers used for the lubricant solutions made with tap water solvent. From figure 8 it can be observed that the minimum value of the wear spot diameter appears for the solution S3 that is made with polymer P1. In figure 9 it is showed the influence of the NaCl content in solution on the wear spot diameter. It can be remarked that the lowest value of the wear spot was registered for the solution S5 for which was determined some of the biggest friction force values (figures 4 and 5).
In figure 10 there are presented the values of the wear spot diameter registered for each tested lubricant solution and the axial load before welding which corresponds of these wear values. It can be observed that the lower values for the wear spot diameter are registered for lubricant solutions S5 (solvent tap water + 2 wt. % NaCl) and S3 (solvent tap water).

4. Conclusions
The HPAM lubricant solutions consists a possible alternative in lubrication. Due to their properties, they can successfully remove other traditional sort of lubricants. The main characteristics (rheological and tribological) of these lubricants were determined and presented by author [7,8,9,10].

The behaviour of these lubricants in elastohydrodynamic contacts was not studied until now, and from this point of view it appears the necessity to determine the influence of their characteristics on the friction force and wear values which are registered during the tests done on four ball machine.
The main conclusions which result from this paper refer to the optimum polymer characteristics values that imply a better behaviour in elastohydrodynamic contacts:
- molecular weight in the range of $6.0\cdot10^6 - 10.0\cdot10^6$ g/mol;
- hydrolysis degree in the range of 8.0 – 9.0 %;
- solvent tap water or tap water with 2wt.% NaCl;

Due to their properties, the HPAM water solutions can be used like lubricants in fields such as:
- the industry lubrication (gears, bearings, rolling bearings, slideways, pumps etc);
- the metal working fluids;
- the drilling fluids;
- the medicine;
- the lubrication in special environments (sea water, brine etc).

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