INFLUENCE OF pH LUBRICANT (WATER) IN THE FRICTION COEFFICIENT IN SLIDING OF Si_3N_4–Al_2O_3

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Abstract. *In order to evaluate the effect of the pH of the lubricant (distilled and deionized water) in the friction coefficient between the Si_3N_4 and Al_2O_3 ceramics in running-in period and during the steady state regime of ULFC (ultra low friction coefficient, $\mu < 0.01$), tribological tests were performed with ball-on-disk setup, with Si_3N_4 balls and Al_2O_3 disks, with temperature of (22 ± 2)°C, sliding speed of (1.00 ± 0.03) m/s and load of (54.25 ± 0.17) N. The values of pH´s used in the tests were 3, 4, 5, 6, 7, 8.5, 10 and 12. When the pH values of the lubricant stayed above 10 or below 4, the ULFC regime was not reached. At pH equal to 12 the running-in time drops considerably (from nearly 60 minutes to approximately 25 minutes), but the friction coefficient lay about 0.05 (above ULFC regime). The wear on the disk always remained below the value obtained on the ball, when the pH was 12 the wear on disk and the ball were very small, and when the pH was 3 the wear on disk and the ball were highest. The explanation for this phenomenon comes from the fact the pH variation changes the interaction of the electric double layer surrounding the silica particle, generated in the tribochemical reactions, with the water and the interfaces, changing the lubrication regime (mixed, pH between 4 and 10, to hydrodynamic, pH below 3 and above 12, as proposed in the literature). At pH below 3 the electrical double layer is very thin, making it not occur the boundary lubrication, acting only the hydrodynamic lubrication. At pH above 10 the generated silica turns into other compounds, which do not allow a boundary lubrication, not allowing a mixed lubrication.*

Keywords: *Friction coefficient, Silicon Nitride, Alumina, Water, Tribology.*

1. INTRODUCTION

Since a very low friction coefficient (≤ 0,002) was observed, the tribological behavior of ceramics under water lubrication has been investigated by several authors. Tomizawa and Fischer[1] discovered that the friction coefficient of silicon nitride sliding against itself, with water as lubricant, starts with values of $\mu = 0.7$ and reach values as low as $\mu = 0.002$. They attributed the very low values of the friction coefficient to the formation of a highly polished surfaces between the bodies. They observed also that a similar phenomenon occurred with SiC-SiC pair.

Xu and Kato[2] attributed this phenomenon to the occurrence of tribochemical reactions in the process, according to these authors, led to the formation of a tribofilm composed of silicon dioxide with low shear strength, suggesting that the charged particles of colloidal silica provided a mixed lubrication.

Anderson[3] observed for alumina sliding on itself, the kinetic friction coefficient is $\mu = 0.2$ in steady state. He also observed for silicon carbide $\mu = 0.02$ after a running-in period.

Recently, Ferreira (2008)[4] discovered that ULFC can be obtained for the dissimilar ceramic pair Si_3N_4–Al_2O_3 sliding in water. In his work, Ferreira showed that decrease in surface roughness decreases the time of running-in, which was in agreement with the result Tomizawa and Fischer.

However, the mechanism for ULFC occurrence is not well understood, several explanations was presented. Some authors attribute the phenomenon to the hydrodynamic lubrication[1,4,5,6,7], others considered that a triboreaction is necessary to establish the ULFC regime and that the lubrication occurred in the mixed regime [2,8]. All those studies were performed with SiC-SiC ou Si_3N_4–Si_3N_4 pair. Ferreira (2008) adopted the later mechanisms to explain the ULFC regime to the Si_3N_4-Al_2O_3 sliding under water with friction coefficient in the range from 0.002 to 0.004.

It was attributed in all studies above mentioned the lubricant (water) plays a key role. The effect of pH water was studied for SiC-SiC and Al_2O_3-Al_2O_3 pairs under sliding conditions[10]. In this previous work no significant difference was observed, with the variation of the pH solution on SiC-SiC and Al_2O_3-Al_2O_3. This paper studies the dissimilar Si_3N_4-Al_2O_3 ceramic pair in sliding lubricated by controlled pH water, on the tribological behavior.

2. EXPERIMENTAL

A high purity alumina powder with 750 ppm MgO (AKS-3030A, Sumitomo Chemical Co., Japan) was uniaxially pressed to form discs in a cylindrical die (diameter of 52 mm) at 45 MPa. The discs were then pressed at 200 MPa in a cold isostatic press and afterward sintered at 1650 ºC for 1h in air using an electric furnace.

The flat surfaces of discs were machined and then a hole in the center of each disc was drilled to fix it at the wear equipment. Before the test, the flat surfaces were lapped using copper lapping discs and a 15 µm monocrystalline diamond suspension. The value of RMS roughness was 299 nm, evaluated by contact profilometer (Kosaka, Surfcorder

- 1700α).

Lubricated sliding wear tests were conducted using a ball-on-disc setup in the Plint Microcontoled TE67 PIN ON DISC MACHINE. Commercial Si_3N_4 bearing balls with 11.11 mm (7/16") in diameter were used in the tests. During the test, the prepared Al_2O_3 discs were fixed and rotated with a tangential speed of 1.0 m/s. The Si_3N_4 ball was pressed against the Al_2O_3 disc through a dead weight system with a normal load of 54.25 ± 0.17 N that resulted in a mean Hertzian pressure of 900 MPa. The tests were performed at ambient temperature (22 ± 2 ºC) with a flux of filtered, distilled and deionized water, that covered the contact interface between ball and disc. To vary the pH, was added a solution of sodium hydroxide (0.1 M) or a hydrochloric acid solution (0.1 M) to the water(12). It was used a pHmeter Gehaka model PG2000 to perform the measurement of pH value. The values of pH's used in the tests Were 3, 4, 5, 6, 7, 8.5, 10 and 12. The friction force was monitored with a cell load with an accuracy of 0.10 N at a frequency of 1 Hz. The mean friction force (and friction coefficient) was measured up to achieve at least 10 min at the ultra low friction regime. Some tests were carried out to confirm the reproducibility of the results.

Before and after the tests the specimens were cleaned in an ultrasound bath (20 min in acetone). The roughness profiles were measured before and after the tests without filtering for comparative purposes. The worn surfaces of tested ball and disc were analyzed by optical microscopy.

Table 1 shows the measured physical and mechanical properties of tested materials.

Table 1: Physical and mechanical properties of Al_2O_3 disc and Si_3N_4 ball

Material	Alumina	Silicon nitride
Density, ρ [$g.cm^{-3}$]	3.908 ± 0.010	3.183 ± 0.015
Vickers Hardness, HV_{10} [GPa]	16.32 ± 0,16	14.79 ± 0.10
Young Modulus, E [GPa]	445.28 ± 16.17	288.0 ± 13.7
Fracture Toughness, K_{Ic} [$MPa.m^{1/2}$]	4.4 ± 0.4	6.0 ± 1.3

3. RESULTS AND DISCUSSION

A typical plot of tribological tests for this pair, showing the friction coefficient as a function of sliding time is shown in Fig. 1.

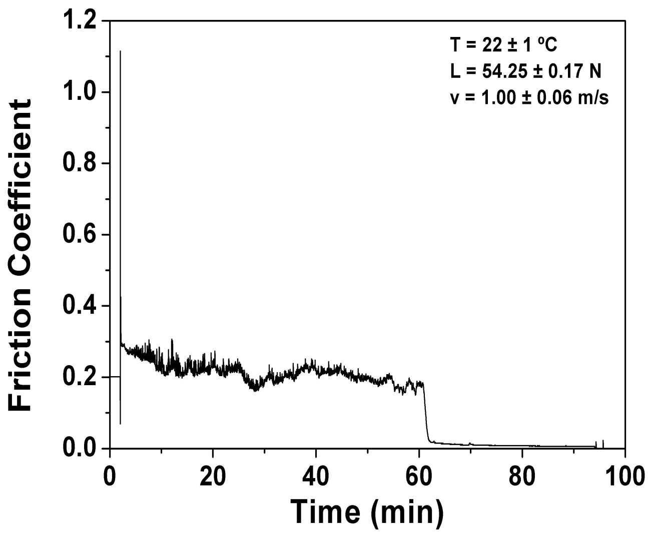

Figure 1: Si_3N_4-Al_2O_3 friction coefficient lubricated with water. T - water temperature, L - applied load, v - sliding speed.

At the beginning of the test, the friction coefficient reached 1.5 and after a short period, decreased to 0.3. Then it was followed a transient regime where the friction coefficient variation was large. During this period the mean value and the amplitude of the friction coefficient decreased and after one hour the friction coefficient reached values of $\mu<0.01$ remaining in this regime until the test was ended. This friction coefficient range (<0.01) were named Ultra Low Friction Coefficient (UFLC) range.

Figure 2 shows the friction coefficient versus time for the pair alumina-silicon nitride sliding in water, with pH controlled.

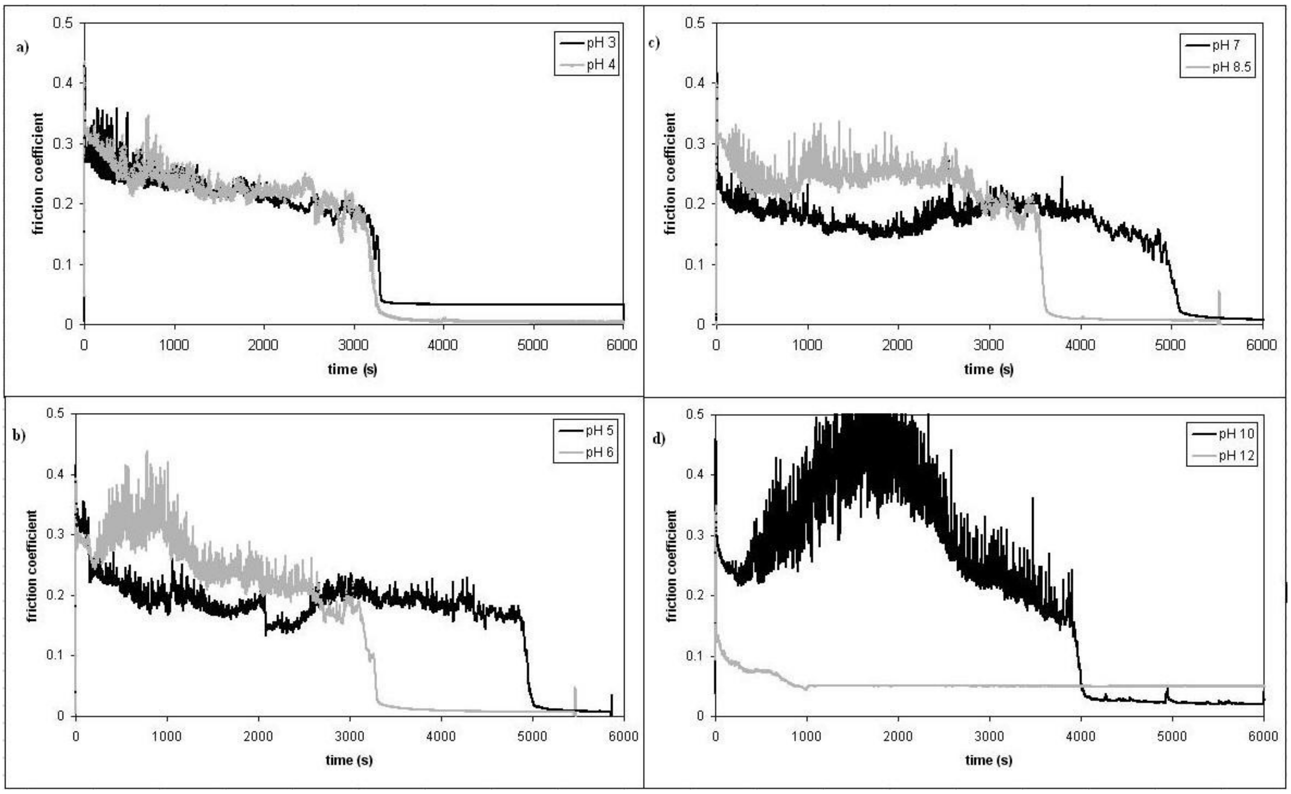

Figure 2: friction coefficient versus time for the pair alumina-silicon nitride sliding in water, with pH controlled. (a) pH 3 and 4 (b) pH 5 and 6 (c) pH 7 and 8.5 (d) pH 10 and 12

When the tests were performed with pH between 4 and 10 we had a total friction coefficient less than $1x10^{-2}$. In this pH range, the colloidal silica formed by a tribochemical reaction, adsorb water on its surface, which alters the electrostatic properties of the particle surface, generating a tribofilm electrically charged. When in contact with other particles it supports the materials allowing a boundary lubrication, resulting in a mixed film (2), as shown in Figure 3.

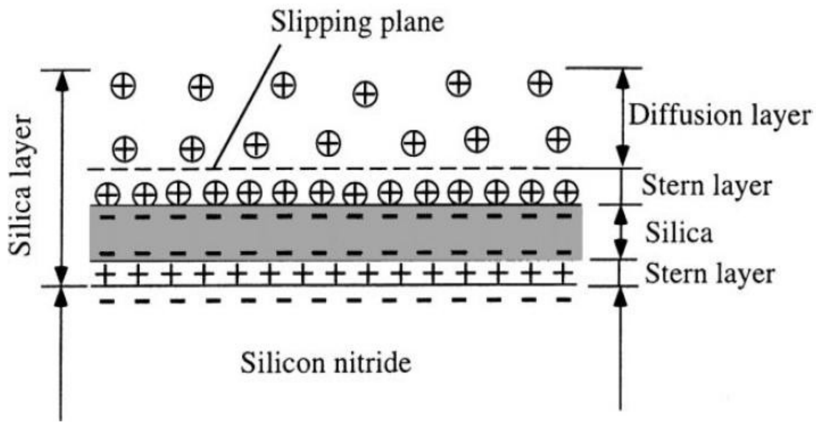

Figure 3: silica layer formed during the test(2).

The tests at pH 3 and pH 12 did not reach a value of ultra low friction coefficient, as noted in Figure 4. At low pH (~ 3, isoelectric point), the repulsive forces that acted upon the colloidal silica particles decreases, causing them to agglomerate, and there is no more support forces. At high pH values (> 10), the solubility of silica increases exponentially (11), making these particles do not form electrostatic interactions with other particles and surfaces.

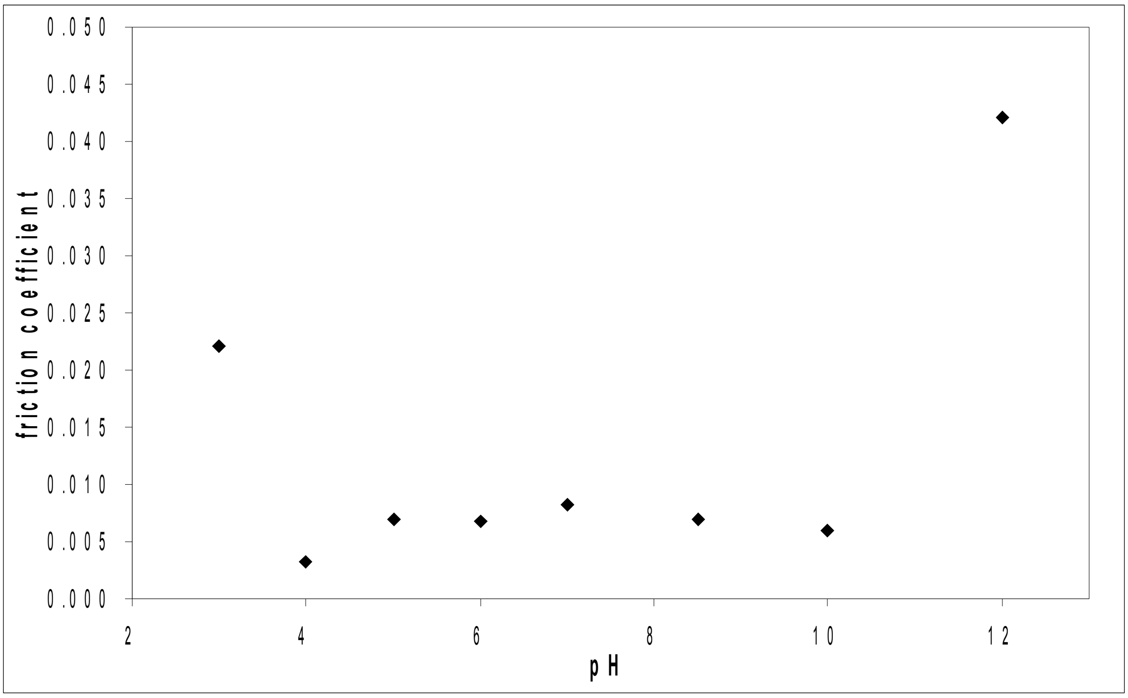

Figure 4: average friction coefficient during the regime UFLC versus pH.

In Figure 5 we can see that the running-in period occurs after a tiime between 3500 and 5500 seconds in the tests, except when the pH is 12 in this case the running-in time fall to 1500 seconds.

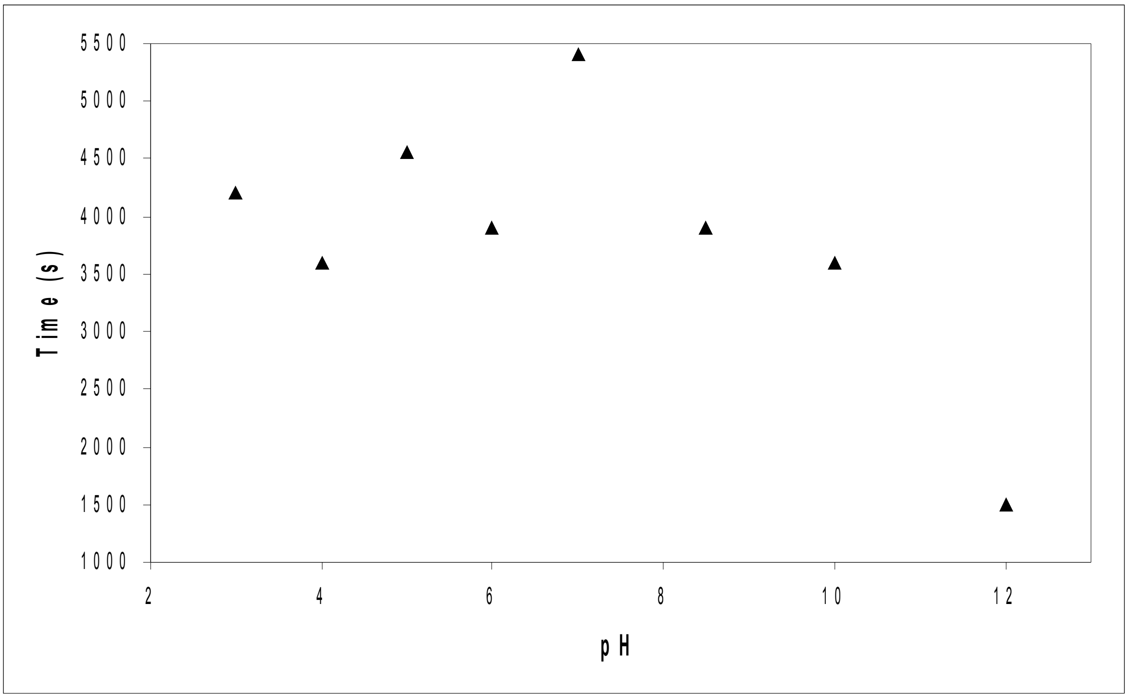

Figure 5: Running-in time versus pH.

Figure 6 shows a typical worn surfaces on the Alumina discs. Figure 7 shows the typical plateau formed on the Si_3N_4 ball.

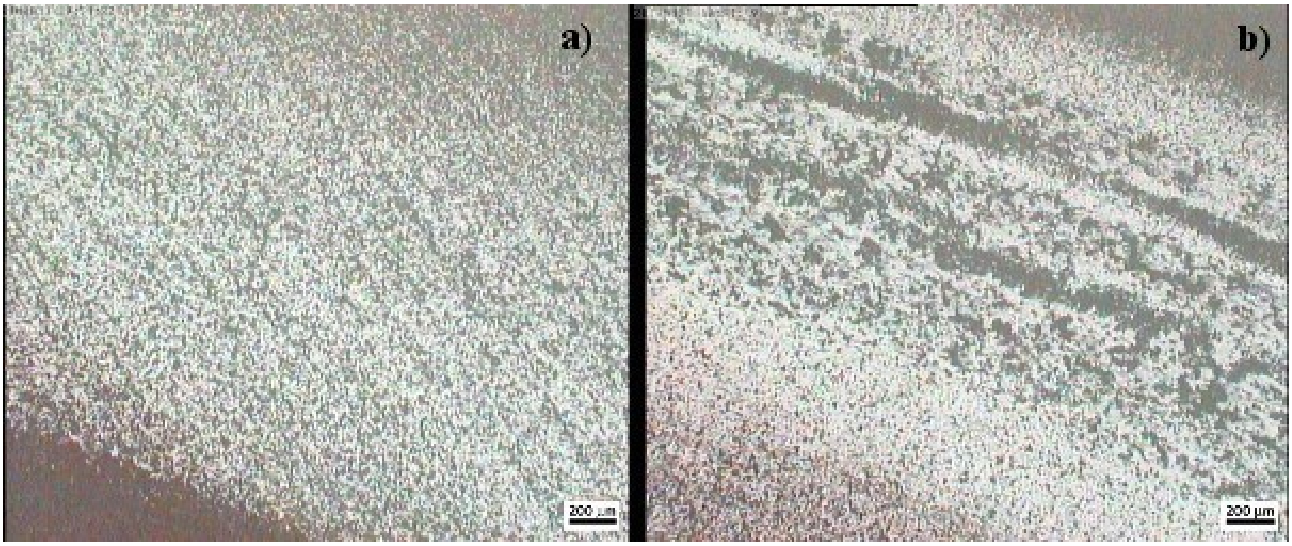

Figure 6: Wear tracks. a) and b) are the deference of the track in the same disc.

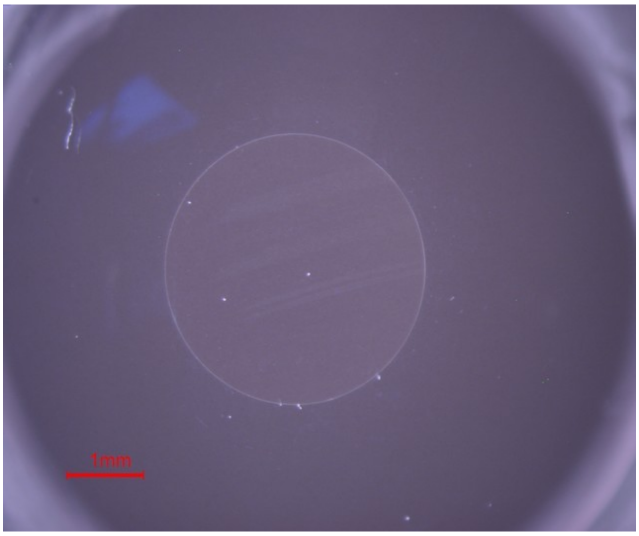

Figure 7: plateau formed on the ball

It was observed the wear track on the discs where not uniform, as shown in figure 6 a and b. In some regions, like in figure 6a, the rms roughness decreased during the tests and in regions like figure 5b the rms roughness reached values about twice the roughness at the beginning of the test. This behavior could not be explained. The ball wear was in all tests uniform and the roughness was always lower than that of the discs.

Figure 8 summarizes the wear of the Si_3N_4-Al2O3 pair materials as a function of the water pH. The wear of both materials is highest when the water pH is 3, and lower when the water pH is 12. The wear of the discs was lower than the wear of the balls, once the hardness of the balls is smaller than the discs.

When the pH is 3 the lack of electrostatic support caused by colloidal silica, results in a greater contact, increasing wear. When the pH is 12, the increase of the formation of substances based on silicates, with lubricating properties, reduce the wear and running-in time.

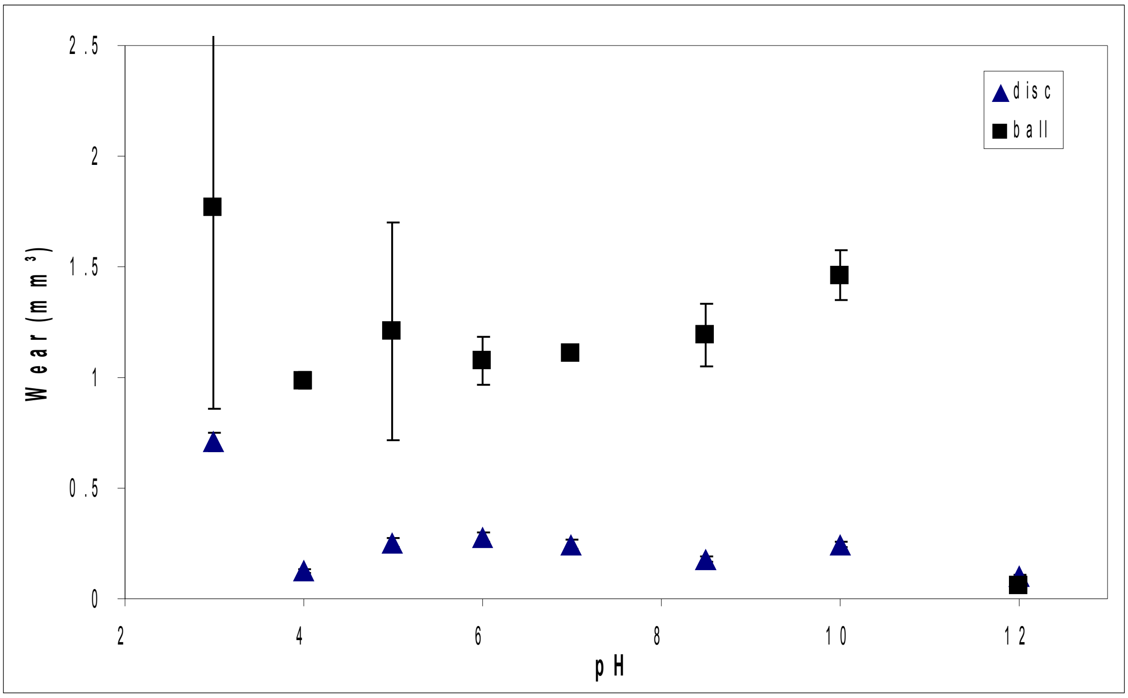

Figure 8: Wear volume.

5. CONCLUSION

Ultra low friction coefficient (ULFC) was reached in an unmated Si_3N_4-Al_2O_3 sliding pair under water. This result showed that the ULFC is not a phenomenon exclusive of Si_3N_4 - Si_3N_4 and SiC-SiC mated pairs as was first observed by Ferreira 2008.

The ULFC was obtained without a very smooth interface as pointed out by the literature.

At pH values too low or too high is not possible to reach the regime of ultra low friction coefficient, so the phenomenon is concentrated at pH's between 4 and 10.

At pH equal to 12 the running-in time drops considerably spite of not reaching a value of ultra low friction coefficient, is 0.05. The wear when the pH is 12 is very low both on disk and on the ball.

When the pH is between 4 and 10 the running-in time fluctuates in a range from 50 to 110 minutes depending on the initial conditions of the test.

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