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Mechanical properties and tribological behavior of a silica or/and alumina coating prepared by sol-gel route on stainless steel

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ABSTRACT

The increasing importance of developments in the mechanical industry requires a constant evolution of skills, particularly in the area of functionalization and protection of metallic alloy surfaces. The wear of materials is one of the causes of loss of profitability. This study aims to develop solutions to extend the lifetime of stainless steel 304 L. In this work, we have achieved to optimize protective coatings on stainless steel against wear, using sol-gel method associated with dip-coating technique. Three routes have been proposed to achieve this type of coating, a single sol precursor of silica or alumina and a mixture of sols precursors of both oxides. The results of tribological tests show that silica coating does not provide a performance gain towards the stainless steel protection. Alumina coating even as thin film is very efficient to resist against wear. Wear track widths on the sample is reduced by a factor 2, and the wear volume of the counterface is decreased by a factor 30, corresponding to a total wear volume reduction of a factor 7. The combination of both oxides seems to be a very promising way for such kind of application. With silica/alumina coating, we have obtained a reduction of widths wear track of a factor 1.5 and a decrease of friction coefficient as compared to alumina coating.

1. Introduction

The main objective in this study is to increase the durability of 304-L stainless steels with a protecting thin ceramic film. Thanks to their high hardness values, ceramic coatings would present the best properties to decrease wear. Apart from the conventional deposition ways such as PVD [1,2], CVD [3] or plasma sprayed [4–7], the sol-gel method is a new alternative process to develop coatings in order to improve the tribological behavior of the whole systems [8–16]. Indeed, many fields of material engineering use this method due to the advantages it offers. The sol-gel process allows the composition of the coating to be adjusted according to the intended application. For example, currently, the applications for sol-gel coatings are in the enhancement of adhesion, in anti-corrosion coatings, optical sensor and more recently, in the research developments for applications such as thermal barriers or high electrochemical performance films [17–21]. Here, the objective is to optimize the deposition parameters, and thus the nature and the structure of the coating to reduce wear. These parameters are dependent on the precursor concentration in the primary solution. It has also been proven that the final heat treatment conditions (temperature, atmosphere) play an important role on the nature and properties of the oxide film. Selected materials were silica- and alumina-based compounds, which are well known to be good candidates to tackle wear because of their mechanical behavior [12,22–24]. The aim of this study is the optimization of the sol-gel synthesis of silica and/or alumina coatings on stainless steel 304 L, to determine the best candidate materials in terms of tribological behavior and mechanical properties.

2. Materials and methods

2.1. Materials

AISI 304-L stainless steel disks of 30 mm in diameter and 5 mm in thickness were used as substrates. Pre-treatment of the surfaces of the samples was first done by grinding in order to obtain an average roughness (R_a) of 0.6 μm, followed by alkaline degreasing, acid pickling and finally nitric acid passivation of the surface.

2.2. Coating processing on stainless steel substrates AISI 304 L

The preparation of silica and alumina sol-based coatings was performed as described below:

2.2.1. Silica sol

For the silica coating, the precursor used was an alkoxide, tetraethyl orthosilicate (TEOS). The sol was prepared [13] by mixing acetylacetone (AcAc) and water in ethanol at room temperature, followed by adding the required amount of TEOS and hydrochloric acid under stirring to obtain the proper concentration of TEOS. The volume fractions of TEOS,

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ethanol, hydrochloric acid, AcAc and water in the prepared solution are 7/89.5/0.5/2/1, respectively. After stirring of the mixed solution at room temperature for 0.5 h, a transparent solution was obtained, which was then aged for 24 h and used for film preparation. The viscosity of silica sol is around 7 MPa · s during deposition. The silica coatings were shaped by a dip-coating method with a withdrawal speed of 300 mm min^{-1}. The films were dried for 2 h at 80 °C and then densified by thermal treatment under air for 25 min at 500 °C with heating rate of 100 °C/h. A scanning of the TEOS concentration in the sol was performed in the range of 0.17 to 1.66 mol L^{-1}.

2.2.2. Alumina sol

The alumina precursor is a metal salt. An excess of ammonia solution (5 mol L^{-1}) was firstly dripped into an aqueous aluminum chloride solution (0.13 mol L^{-1}) at room temperature until a pH value of 9 was reached. The obtained compound was then filtered and washed with hot distilled water. The cleaned hydrated precipitate was aged for 72 h in a drying oven at 85 °C. After drying, the precipitate consists of boehmite (Fig. 1), and a known amount of the boehmite was dispersed in distilled water. The solution was peptized with a 12.5 vol% of acetic acid (100%) and then stirred for 24 h to obtain a colloidal dispersion. Next, an addition of PEG (Molecular Weight = 400 g mol^{-1}) with a volume fraction of 1.5% was done and a new maturation of 24 h with magnetic stirring was necessary. A deposition was made by dip coating with a withdrawal of 300 mm min^{-1}. The films were then dried and densified under the same conditions as for silica films. XRD studies on alumina xerogel have shown a phase transformation from boehmite to gamma alumina. By adjusting the amount of boehmite in the present medium, a scanning of the boehmite concentration was performed in the range of 0.17 to 1.60 mol L^{-1}.

2.3. Characterization techniques

Several techniques allow the determination of the surface properties of samples. The measurements of contact angles were carried out with a goniometer GBX Digidrop using deionized water (2.5 μL). The roughness of the substrate was measured with a Bruker's NPFLEX™ 3D Surface Metrology white light interferometry. With this technique, it is also possible to determine a wear surface. Ten measurements were performed on each sample to obtain an average wear profile along the track. Then, the total wear volume was determined by multiplying the wear profile by the circumference of the worn track. The wear width was determined by optical microscopy Keyence VHX-1000E, while the wear volume of the ball was obtained by calculation of the spherical cap measured by white light interferometry.

The rheological analysis of sols was made by a viscometer Rheomat RM100 of type Taylor–Couette flow at a shear range of 200–3867 s^{-1}. Structural analyses of the thermal-treated xerogels were performed with X-ray diffractometer BRUKER D4 ENDEAVOR. XRD patterns were collected at room temperature by scanning steps of 0.02° (2θ) over a 10.00° < 2θ < 100.00° angular range and using a Cu Kα radiation (0.15418 nm).

Thickness of films was determined by Scanning Electron Microscopy (SEM) JEOL JSM 6400 on cross-sectional images or by a DEKTAK 3030ST (VEECO) mechanical profilometer. Adhesive properties of films were evaluated by a Nano-Scratch tester (NST, CSM Instruments) to measure critical loads of crack formation or delamination. Scratch tests were performed with a loading rate of 13 mN min^{-1} and a maximum normal load of 100 mN. Three scratches were performed on each sample. Tribological evaluation was carried out by uni-directional sliding test on a pin-on-disk tribometer (CSEM Instruments) under ambient environment. This test is based on the international standard ASTM G99, which simulates the wear issue found in mechanical industry. The pin was a 316-L stainless steel ball with a diameter of 10 mm. Normal loading force was 2 N and sliding distance was 250 m. Tests were made at a sliding velocity of 10 cm s^{-1}, within a circular diameter of 20 mm on the samples. Three tests were systematically done on each sample.

3. Results and discussion

3.1. Surface preparation on AISI 304-L stainless steel

After mechanical grinding of the substrate surface to an average R_a value of 0.6 μm, several chemical treatments were tested in order to enhance the surface wettability (Fig. 2). All the samples were degreased by using alkaline solution, and then two types of picklings (either fluonitric or sulfuric) were applied. In the case of sulfuric pickling, two immersion durations have been tested. The effect of passivation on the surface from these two types of pickling has also been studied. Chemical preparation does not affect the roughness of the substrate which remains at an R_a value of 0.6 μm. Fig. 2 showed that the fluonitric stripping did not confer to satisfactory standards in terms of wettability; on the contrary, sulfuric stripping provided an appropriate surface. For economical reasons, 7 min of stripping was chosen. In view to facilitate easy handling and to avoid excessive surface reactivity, we decided to passivate the surface.

3.2. Silica coating

After the analysis of the surface topography as well as the rheological study of the sol, homogeneous coating is derived from a sol with a molar concentration of 0.88 mol L^{-1}. Profilometry measurements show that the thickness of silica coating was around 1 μm. The tribological behavior of the silica-coated sample has been compared to the one of the pristine substrate without surface preparation and also to one of the substrates which has undergone both surface preparation

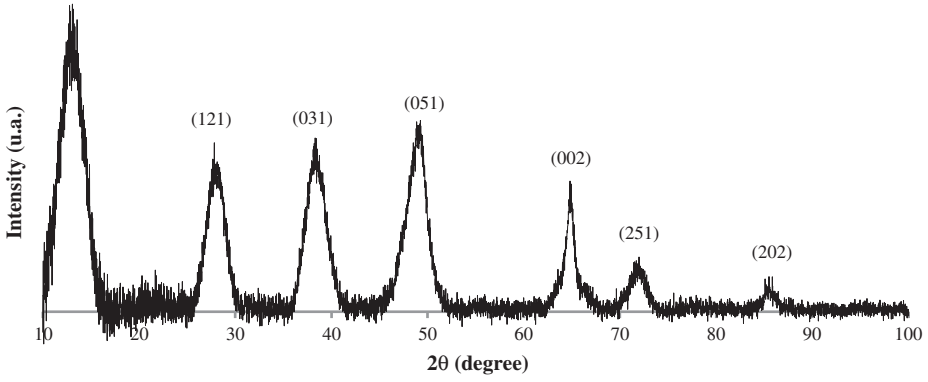

Fig. 1. X-ray pattern of the boehmite after drying at 85 °C (JCPDS card 00-021-1307).