

Coupling experimental and numerical approaches to study adsorption mechanism of stearic acid on iron based surfaces

Clotilde Minfray¹⁾, Sophie Loehle^{1)*}, Christine Matta¹⁾, Thierry Le Mogne¹⁾, Jean-Michel Martin¹⁾,
Raphaele Iovine²⁾, Yukiko Obara³⁾, Ryuji Miura³⁾ and Akira Miyamoto³⁾

1) Laboratory of Tribology and System Dynamics, Ecole Centrale de Lyon, 36 avenue, Guy de Collongue 69134, Ecully Cedex, France

2) TOTAL, Solaize Research Center, BP22 – 69360 Solaize Cedex, France

3) New Industry Creation Hatchery Center, Tohoku University, 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan

*Corresponding author: clotilde.minfray@ec-lyon.fr

1. Introduction

Organic friction modifiers (OFMs) have been used for many years in metallic contacts to reduce friction under mild conditions. The interest for such friction modifiers strongly increased in the recent years because of environmental regulations ask for slightly hazardous lubricant additives. It is proposed here to revisit adsorption and friction behavior of fatty acids by coupling experimental and computational chemistry approaches to better understand their mechanism of action and optimize their performance.

2. Methodology

In this paper stearic acid was studied as an OFM and was used as pure additive or blended at 1%w in PAO4 synthetic base oil. The surfaces of interest used were iron-based materials (including pure iron and iron oxides) as they can be encountered in a steel/steel contact under mild or severe friction conditions. Adsorption and friction properties of such systems were studied experimentally by friction tests, XPS and PM-IRRAS surface analyses. Computational chemistry (UA-QCMD) was also used to study the adsorption kinetics. In the following, adsorption experiments, computational chemistry simulations and friction test are detailed.

Adsorption experiments of stearic acid vapor on the different surfaces were carried out in an environmentally controlled chamber and this was followed by *in-situ* XPS analyses of the surfaces. Stearic acid vapor was generated in the adsorption chamber in presence of the different substrates at different adsorption time (10 min to 2 hours).

Computational chemistry study was also performed to investigate the *in vacuo* interaction of stearic acid molecules with the different substrate (pure iron, Fe₂O₃, and FeOOH). An ultra-accelerated quantum chemical MD (UA-QCMD) simulator has been used in order to deal with chemical reaction dynamics for large complex systems [1]. Different models have been built starting with one single molecule up to self-assembled

monolayer (SAM) on iron based surfaces at 50 °C. Eventually, friction experiments were carried out with a cylinder-on-flat reciprocating tribometer using AISI 52100 steel cylinders and flats. The lubricant was made from stearic acid blended at 1%w in PAO4, temperature was set to 50°C, 100°C and 150°C and the applied load was 50 N (corresponding to a maximum contact pressure of 320 MPa). After the tribological tests, PM-IRRAS analyses were conducted on the tribofilms.

3. Results and discussion

Adsorption experiments

In-situ XPS analyses results, especially the position of the C1s peak contribution from the carboxylic group, show differences depending on the nature of surfaces.

Computational chemistry

As said before, different models have been built starting with one single molecule up to self-assembled monolayer (SAM) on iron based surfaces at 50 °C. We found differences in adsorption energy (physisorption or chemisorption) depending on the kind of surface (pure iron, Fe₂O₃ and FeOOH) and regarding the presence of one single molecule or a SAM.

Friction tests

After the tribological tests, PM-IRRAS analyses were conducted on the tribofilms. Results show the presence of stearic acid in the wear track as well as carboxylate forms.

The experimental and simulation results for both adsorption and friction experiments will be discussed for a better understanding of stearic acid complex interactions with iron-based surfaces,

[1] Md. Korshed Alam et al. Catalysis Today 164 (2011) 9–15