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Multilayered diamond mechanical seal rings under biodiesel lubrication and the full sealing conditions of pressurized water

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ABSTRACT

A mechanical sealing tribosystem based on multilayer microcrystalline/nanocrystalline diamond (MCD/NCD) coatings is proposed. The tribological behaviour was investigated in ring–on–ring self–mated planar contact configuration. Tests were carried out under biodiesel lubrication and pressurized water (2 bar) sealing conditions. A 3D optical confocal profilometer was used to obtain for the first time the wear coefficients of seal rings, revealing a value of $k = 6.3 \times 10^{-10}$ $mm^3/N.m$ for biodiesel lubrication that is two orders of magnitude lower than for reciprocating sliding ball-on-flat experiments. The advantage of the multilayer strategy was demonstrated in the sealing of pressurized water, where full sealing conditions were achieved within the $P \times V$ range of 0.75–5.5 MPa ms^{-1}. For these coatings, full delamination is prevented due to structural discontinuity at the MCD/NCD interfaces whereas for a MCD monolayer coating, premature failure takes place and sealing is not reached.

1. Introduction

Mechanical face seals are critical parts of equipment and machinery across all human activities related to transport of fluids, wherever leakage control and safety issues regarding facilities, environment and people are at stake. The materials used for the seal itself, the moving surfaces where the sealing is performed, span a large range both in their nature and their working principles. There are basically two types of seal combinations: i) one of the materials is sacrificial and will wear over time, with scheduled replacements every few months (or years) of operation; ii) both counteracting materials are hard, can be made of the same or different materials, and they will have expectedly longer running lives. Common to both types of interfaces is the need of very low friction coefficients, minimization of wear rates and a maximum reliability. Additional criteria that will affect the choice of materials are e.g. enhanced corrosion resistance, low weight, ability to operate at higher temperatures or the need to endure dry cycles. From the standpoint of the materials, this means a mixture of thermo-mechanical and surface or chemical properties that seldom are satisfied by a single material. Diamond is an exception, being extremely hard and corrosion resistant, possessing the highest thermal conductivity and a very high thermal shock resistance [1,2].

In the present work, the hot filament chemical vapour deposition (HFCVD) technique was used to produce multilayer diamond coatings, alternating microcrystalline diamond (MCD) and nanocrystalline diamond (NCD) to a total of ten layers. Silicon nitride ceramic seal rings coated with this multilayer system and with a monolayer MCD coating were tested under biodiesel lubrication and water sealing in self-mated hard contacts. Biodiesel has become an important ecological alternative to petrol. Pumping is extensively used in the biodiesel industry for raw materials, processing and fluid transfer, where the pump rotors and seals are frequently subjected to hot and aggressive conditions and the transfer pumps may become clogged with wax and fatty deposits. The testing with water, the most widely pumped fluid, serves to validate this tribosystem.

The strategy of the multilayer coating is to deposit a first MCD layer, due to its superior adhesion to the Si_3N_4 ceramic [3] and finish with a top NCD layer. This smooth NCD layer [4–7] prevents the high starting friction coefficients that would happen if the rougher MCD was the top surface, thus decreasing the probability of coating failure. Also, the successive intercalation of NCD layers between the MCD ones has twofold advantages: i) it interrupts the columnar growth mode of MCD [8]; ii) it delays the propagation of cracks that would otherwise easily propagate in the growth direction of MCD, thus improving the fracture toughness of the coating [9,10].

The extremely low wear rates of diamond coated rings prevented so far the evaluation of the wear coefficient. In this work, a

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relevant and innovative approach of the research is the use of 3D optical profilometry to measure the volume loss after sliding, a feat unreachable by conventional methods such as weighing or AFM analysis.

2. Materials and methods

Seal rings were prepared in house by ball milling a mixture of 89.3 wt.% Si_3N_4 (HC Starck M11), 3.7 wt. % Al_2O_3 (Alcoa 116SG) and 7.0 wt. % Y_2O_3 (HC Starck C), drying, pressing and sintering under N_2 atmosphere. The sintered silicon nitride rings were machined and flat lapped to the following final dimensions in mm: 43 × 33 × 8.5 and 46 × 31 × 7.5 (external diameter × internal diameter × thickness). All rings were plasma etched by CF_4 during 10 min and ultrasonically seeded with diamond in a nano-diamond slurry during one hour.

Diamond coating was accomplished in a home-made semi-industrial HFCVD (hot filament chemical vapour deposition) apparatus with a deposition area as large as 20 × 30 cm^2, using H_2/CH_4 gas mixtures. Two types of diamond coatings were grown on the Si_3N_4 seal faces: a tenfold multilayer with alternating layers of micro and nanocrystalline diamond (MCD/NCD) and, for comparison, a monolayer microcrystalline diamond (MCD), both with total thickness of about 10 µm. The deposition conditions of MCD/NCD multilayer coating are given in Table 1. The monolayer was obtained with the same parameters of MCD but for a total deposition time of 10 h.

A rotary tribometer (TE–92 Plint) was used for the tribological tests of the diamond coated Si_3N_4 mechanical seal faces. The tribological behaviour was investigated in ring–on–ring self–mated planar contact configuration according to ASTM D3702 standard [11]. Fig. 1 shows one pair of rings mounted on the holders to fix on the tribometer. The ring on the left hand side of the photograph is the rotational ring (narrower), while the larger stationary ring (larger width) is on the right hand side.

Tests were carried out in biodiesel and in pressurized water (2 bar) conditions, Table 2. The biodiesel is a commercial soybean oil (77%), palm olein (22%) and rapeseed oil (1%) derived biodiesel, with a kinematic viscosity of 4.3 mm^2/s at 40 °C. It has an ester content of 98.9 wt%, a cetane number of 51.4 and a maximum of 6 mg/kg of organic and inorganic contaminants.

A series of preliminary experiments of short duration of about 2.5 h (Screening in Table 2) was conducted for achieving the stable working conditions under biodiesel lubrication. After these, a single run test with 16 h duration was performed in stable conditions regarding the absence of vibrations or increase in temperature (Stable conditions in Table 2). The coefficient of friction coefficient (COF) values were taken from the instantaneous torque appraised from a load cell.

The sealing tests were done with pressurized water (P_{water}= 2 bar) and not with biodiesel due to safety issues related to excessive heating of the latter. Test conditions that guarantee full sealing are given in Table 2 (Minimum P × V condition and P × V limit condition).

Field emission scanning electron microscopy (FE–SEM) was carried out using a Hitachi SU–70 system for surface morphology characterization of the diamond coated Si_3N_4 ceramic components before and after the tribological testing. Surface roughness, topographic analysis and quantification of the wear volume was done using a 3D optical profilometer (Sensofar S-neox).

Table 1
HFCVD conditions for multilayer diamond deposition on Si_3N_4 ceramic seal rings (5 MCD+5 NCD alternating layers).

	MCD layers	NCD layers
Filament temperature (°C)	2260	2330
Substrate temperature (°C)	850	750
CH_4/H_2 flow ratio	0.02	0.04
Total gas flow (ml min^{-1})	1800	900
Total pressure (mbar)	75	25
Deposition time per layer (min)	60	84
Total deposition time (h)	5	7

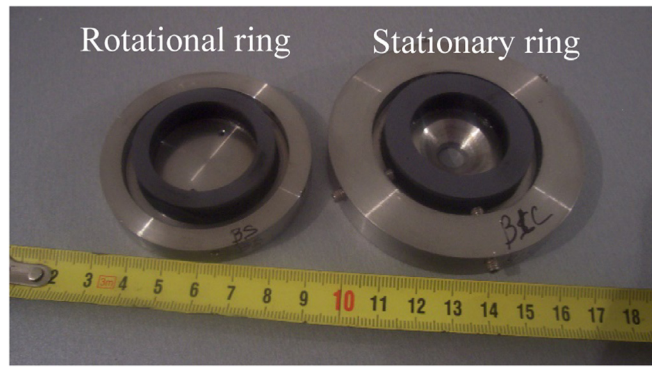

Fig. 1. Example of multilayer diamond coated seal rings mounted on the tribometer fixture.

Table 2
Test conditions for tribological testing in ring–on–ring self–mated planar contact configuration. F – normal applied load; V – linear velocity; P – nominal contact pressure; t – testing time; L – sliding distance.

Type of test	Test objective	V ($m\,s^{-1}$)	F (N)	P (MPa)	L (km)	t (h)
Biodiesel lubrication	Screening	0.5–1.2	150–900	0.25–1.50	up to 396	up to 220
	Stable conditions	0.5	350	0.59	28.8	16
Sealing of pressurized water	Minimum P × V condition	0.5	350–1000	0.59–1.68	2.8–9	1.5–5
	P × V limit condition	3.9	350–900	0.59–1.50	295	21

3. Results and discussion

3.1. Morphology and surface roughness of the diamond films

The typical morphology of the top surfaces of the diamond coated Si_3N_4 ceramic mechanical seal rings are illustrated for the monolayer MCD diamond in Fig. 2a and the multilayer MCD/NCD coatings in Fig. 2b. The MCD coating is coarse–grained, having pyramid–like shape diamond crystals, contrarily to the multilayer coated seal rings that have a top layer of nanocrystalline diamond. Details about the crystallinity and phase composition of the MCD and NCD types of diamond are given elsewhere [12].

As illustrated in the above SEM micrographs, the monolayer MCD coating has a much higher surface roughness than the multilayer coatings that have a top NCD layer. The bar chart in Fig. 3 compares the average values of the surface roughness (areal root mean square parameter, S_q) of these two types of coatings as deposited, and also contains the values of the worn surfaces as discussed below in Section 3.2. Since it is a relevant feature in the as-deposited surfaces, it should be mentioned that there is some lack of uniformity between the borders and the centre of the ring