Evaluation of silicon-modified DLC coatings in a dry sliding contact against aluminum EN AW-5083

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Abstract

Diamond-like carbon (DLC) coatings are known to reduce friction and wear. In a dry contact with steel the tribological behavior can be further optimized by adding a certain concentration of silicon to the amorphous hydrogenated carbon coatings structure (a-C:H:Si). However, in a dry contact with aluminum the tribological performance of a-C:H:Si coatings and the optimal silicon concentration is still unknown. In order to define an optimum regarding the silicon concentration, a-C:H:Si coatings with different silicon contents were deposited on 1.2379 tool material by plasma assisted chemical vapor deposition (PACVD) technology. Tribometer tests were used to analyze the friction and adhesive wear behavior against EN AW 5083 aluminum. Additional, strip drawing were conducted tests to investigate the efficiency of the different silicon contents with a higher proximity to industrial processes. The tests showed a deteriorating friction behavior of the a-C:H:Si coatings with an increase of the silicon concentration. Superposing wear mechanisms between the silicon and the aluminum were determined by a subsequent investigation of the wear tracks as an explanation for the tribological behavior.

Keywords: Coatings, DLC, aluminum, wear, friction, dry forming

1 Introduction

Due to its weight advantage and excellent energy absorption capacity, aluminum is an excellent material for an exceptionally wide range of applications within the general lightweight trends. However, aluminum alloys have high demands towards the design of forming technologies. Especially in dry forming processes, the strong adhesion tendency of aluminum alloys leads to high adhesive wear on forming tools [1] and affects the surface quality of components, the process stability and the targeted tolerances. As a consequence of the adhesive wear, the production of components made of aluminum alloys is limited to lubricated forming processes.

In the last decades, numerous investigations proved the potential of diamond-like carbon coatings (DLC) to reduce friction and wear for many fields of application [2]. Horiuchi et al. [3] introduced DLC coatings as a solution to reduce friction in deep drawing of aluminum at room and elevated (200°C) temperature. Murakawa et al. [4] reported that the adhesion tendency of aluminum to forming tools with and without lubrication can be lowered by applying DLC on the forming tool. Ni et al. [5] compared the tribological behavior of a non-hydrogenated DLC coating with a hydrogenated DLC coating in pin-on-disc tests against aluminum. Both coatings showed no aluminum adhesion at room temperature. Furthermore, the friction value of the hydrogenated coating was lower than the value of the non-hydrogenated coating. Nevertheless, the performance of the current DLC coatings is not sufficient so as to utilize these coatings for dry forming of aluminum alloys in an industrial scale [6].

The properties of DLC coatings can be considerably modified by incorporating metallic (e.g. Ti, Ta, W, Cu and Au) or non-metallic (e.g. Si, O, F, B and N) elements. By comparing various test results [7] Donnet summarizes that the lowest friction values with DLC coatings were observed with silicon-modified amorphous hydrogenated carbon coatings (a-C:H:Si) sliding against steel in ambient condition in experiments conducted by Oguri et al. [8]. They reported a variation of the friction value depending on the silicon concentration. The lowest friction value of $\mu = 0.04$ was detected with a silicon concentration ranging between 15-25 at.-%. This is ascribed to the
silicon concentration depended microstructure and nano-
mechanical properties of the a-C:H:Si coatings [9, 10].

However, the potential of a-C:H:Si coatings to re-
duce the strong adhesive wear in dry forming of alu-
ninum alloys is still not clarified. Tribological tests with a-
C:H:Si coatings sliding against aluminum performed by
Weber [11] showed a high potential to reduce friction and
wear. On the other hand Murakawa et al. [12] reported an
increased adhesion tendency of a-C:H coatings after the
incorporation of silicon.

2 Experimental setup

2.1 Coating preparation and characterization

The deposition processes were carried out by plasma
assisted chemical vapor deposition (PACVD). Acetylene
(C2H2) and tetramethyilsilane TMS (Si(CH3)4) were
used as precursors to prepare the a-C:H:Si coatings.
Coatings with different Si-contents (8 at%, 16 at%, 25
at%, 34 at%) were deposited by adjusting the ratio of the
precursor mixture. To enhance the coating adhesion, the
substrates were sputtered clean for 20 min at the begin-
ing of the coating process. Furthermore, an interlayer
based on pure TMS with a thickness of 0.5 µm was ap-
plied to improve the adhesion of the coating.

For a comparison with an established industrial coat-
ing system, an unmodified a-C:H coating (Si0%) was de-
posited by PACVD combined with physical vapour dep-
osition (PVD). Acetylene were used as precursor for the
PACVD process in addition to a sputter process of pure
carbon (PVD) to prepare the a-C:H coating. Analog to
the a-C:H:Si coatings, sputter cleaning was performed to
enhance the adhesion of the coating. In comparison to the
a-C:H:Si coating, the interlayer was made of titanium
with a thickness of 0.4 µm. The deposition process of the
interlayer was carried out by a PVD process.

Additional to the substrates for the tribological tests, pol-
ished flat samples made of bearing steel (1.3505) and
cold work steel (1.2379) and silicon wafers were coated
and allowed a subsequent analysis of the coatings.
Coating hardness were determined with a commercial in-
strument (Fischerscope H 100) recording load versus
depth curves up to 30 mN. Roughness data were derived
using a profilometer (Talysurf/ Taylor-Hubson) for the
steel balls and by confocal white light microscopy in case of
the strip drawing tools wherefore the two data sets are
only limited comparable. Abrasive wear rates were mea-
sured with the ball cratering test [13] operating with an
alumina (Al2O3) suspension (mean alumina grain size 1
µm).

Table 1 summarizes the silicon concentrations and
coating properties of the tested a-C:H:Si coatings. The
stated composition of the coatings was determined by
electron probe microanalysis (EPMA).

2.2 Tribological tests

The coating systems were initially tested by the strip
drawing test in order to investigate the general tendency
of adhesion which can be expected in an industrial appli-
cation. The strip drawing test emulates the typical tribo-
logical load spectrum of deep drawing and stretch form-
ing processes [1]. In this test, a pressure is applied to a
strip of sheet material by an upper and lower tool while
the strip is drawn with the defined sliding speed and slid-
ing distance summarized in Table 2. Fig. 1 shows the test
principle schematically. In order to apply a characteristic
load spectrum for sheet metal forming the typical linear
load and corresponding high contact pressure at the die
radius of a deep drawing tool is reproduced through the
cylinder-plane geometry in the strip drawing. The upper
tool has a cylindrical surface with a radius r = 258 mm
while the lower tool is flat. The basic measurements of
both tools are equal to 40 mm x 40 mm. Tool material
(1.2379) tested coatings are identical in both tribometer
tests and it is equally tested against EN AW 5083 alumi-
num. The strip material was cleaned before starting the
experiment in order to establish dry forming conditions.
For an initial application orientated qualification one test
were conducted for each coating.

![Fig. 1: Schematic of the strip drawing test](image1)

![Fig. 2: Schematic of the oscillation ball-on-disc tribometer test](image2)

The 3D-structure of the adhesions on the tool surface
is scanned and digitalized with confocal white light mi-
croscopy. The maximum height of the adhesions is taken
as a quantitative indicator of the magnitude of the adhe-
sive wear on the tool surfaces. For all configurations the
contact area on the cylindrical tool was analyzed.

Oscillating ball-on-disc tribometer tests (Fig. 2) were used to determine the friction and wear behavior of
the coatings in contact with aluminum. Coated and un-
coated steel balls were slid against sheets made of EN
AW 5083 aluminum. The balls with a diameter of 10 mm
were made of cold work steel (1.2379). According to
DIN 5401, the surface quality of these balls equals G100.
The 5083 aluminum sheets were in H111 condition and the
surface measured an average roughness Ra = 0.308 µm. The coated balls and aluminum sheets were cleaned to ensure a technical pure contact during the
tribometer tests. The tests were conducted three
The results in Fig. 3 show the macroscopic adhesions on the tool surface after the first stroke. For all tool surfaces, a section of 34 mm by 5 mm (x by y) is digitized by confocal white light microscopy. All profiles are extracted for each measurement and their envelope represents the maximum height of adhesions in this area. The test results do not allow a prediction about differing adhesiveness of the different coating configurations. In all cases the adhesions after the first stroke are already macroscopic and do not show any systematics regarding Si-contents. Due to the instant formation of adhesions and the early strip failure it is not possible to investigate the underlying mechanisms for which reason the following ball-on-disc tests were performed.

3.2 Ball-on-disc test

Fig. 4 shows the average friction values of the tested coatings after the run-in period. The friction values increase up to 16% and between 25% and 34% linear with an increasing content of silicon. Between 16% and 25% the value remains constant. This anomaly may be a result of superposing wear mechanisms and will be discussed later in this study. The increasing friction values validate the friction behavior of a-C:H:Si coatings sliding against aluminum A1100 reported by Murakawa [12] for the sliding contact against aluminum EN AW 5083.

The tested balls were analyzed with a light microscope to determine the wear amount indicated by the diameter of the wear track. Additional analyses with a raster electron microscope (REM) and an energy dispersive x-ray spectrooscope (EDX) allowed an identification of aluminum adhesions on the wear track.

3 Results and discussion

3.1 Strip drawing test

In general, all tested coatings show severe adhesive wear on the tool surfaces after the first 100 mm stroke under the applied load of approximately 75 MPa contact normal pressure.

times to improve the statistical quality of the friction and wear values. The test parameters were deduced from an industrial forming process of EN AW 5083 aluminum and are summarized in table 2. Higher contact stresses in the ball-on-disc tribometer tests were used in order to accelerate wear, whereas the strip drawing test is more oriented towards a realistic load spectrum in sheet metal forming.

After performing the tribometer tests, the wear amount on the aluminum sheets was determined by a tactile measurement method (DektakXT - Bruker). Therefore, a nano indenter measured the topography across the wear track. In the topography, the area above the zero line denotes material adhesion and the area under the zero line denotes material removal. The tactile measurements were performed at five different points along the wear track to gain an average wear value.

The results in Fig. 3 show the macroscopic adhesions on the tool surface after the first stroke. For all tool surfaces, a section of 34 mm by 5 mm (x by y) is digitized by confocal white light microscopy. All profiles are extracted for each measurement and their envelope represents the maximum height of adhesions in this area. The test results do not allow a prediction about differing adhesiveness of the different coating configurations. In all cases the adhesions after the first stroke are already macroscopic and do not show any systematics regarding Si-contents. Due to the instant formation of adhesions and the early strip failure it is not possible to investigate the underlying mechanisms for which reason the following ball-on-disc tests were performed.

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The results in Fig. 3 show the macroscopic adhesions on the tool surface after the first stroke. For all tool surfaces, a section of 34 mm by 5 mm (x by y) is digitized by confocal white light microscopy. All profiles are extracted for each measurement and their envelope represents the maximum height of adhesions in this area. The test results do not allow a prediction about differing adhesiveness of the different coating configurations. In all cases the adhesions after the first stroke are already macroscopic and do not show any systematics regarding Si-contents. Due to the instant formation of adhesions and the early strip failure it is not possible to investigate the underlying mechanisms for which reason the following ball-on-disc tests were performed.

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It is noticeable that every coating (Fig. 5a-e) showed an individual tribological behavior. At the beginning of the tribometer tests, the friction coefficients assume high values, ranging between $\mu = 0.78$ (Si0%) and 1.16 (Si34%) dropping to a lower level after a certain period of time and remaining steady. The high friction level is known as run-in period and varied depending on the tested coatings. With an increasing content of silicon in the a-C:H matrix, the duration extended from $t_{ri} = 90$ s for Si0% to 2600 s. for Si34%.

The tests with uncoated steel balls (Fig. 5f) show a different tribological behavior. The run-in period lasted 1000 s and the friction value remained at a high level with an average value of $\mu = 0.51$. This high friction level is typical for a dry sliding contact against aluminum and was caused by a constant formation and disintegration of aluminum adhesions on the ball surface [1], see fig. 6f.

The friction level during the run-in period of the Si25% and Si34% coatings equals the friction level of the uncoated balls. Thus a formation of aluminum adhesion on the coated surface is a possible explanation for the higher friction value. But in comparison to the uncoated ball, no aluminum adhesions were formed directly on the wear track of the coated balls, see fig. 6a-e. Due to the abrasive wear, one of the tested Si34% coatings was completely removed at the end of the tribometer test, see fig. 7. In consequence, aluminum adhesions were formed on the exposed steel surface and the friction value increased at the end of the test (fig. 5e). Both the non-adhesive wear tracks in fig. 6a-e and the formation of aluminum adhesions after a complete removal of the coating prove the low adhesion tendency of the tested coatings.

In comparison to the tribometer tests against steel reported by Oguri [8], the incorporation of silicon leads to a linear increasing friction value in contact with aluminum EN AW 5083 after the run in period, see fig. 4. By incorporating silicon in the a-C:H matrix the abrasive wear of the coating leads to an additional formation of silicon containing nanoparticles [15]. In contact with steel the nanoparticles are able to decrease the friction value by interacting with the counter surface or the environment [8, 14]. Thus, the high friction and the extended run-in period are possibly caused by interdependencies between the silicon in the a-C:H matrix, the formation of silicon containing nanoparticles and the aluminum oxide or pure aluminum. The interaction between these factors increases with a higher silicon content.
On all coatings aluminum adhesions were formed around the wear tracks. Scanning electron microscope (SEM) images of the peripheral zone are shown in fig. 8a and 8b for Si8% and Si34%. Regarding the intersection between the wear track and the peripheral zone, a smoothening of the surface is noticeable for both coatings. On the Si8% coating aluminum adhesions were only formed between the asperities in the peripheral zone whereas on the Si34% coating aluminum adhered to the rough surface of the peripheral zone and the smoothed surface in the intersection.

The hardness of a coating measures the resistance against penetration and therefore the resistance against abrasive wear. In contrast to this, there is no correlation between the hardness and the wear of the tested coatings above 16 at.-% silicon, see fig. 9. Furthermore, the removal of the aluminum increases exponential with an increasing silicon content, see fig. 10. Hence, the abrasive wear of the coatings and the aluminum EN AW 5083 was superposed by another wear mechanism based on the incorporation of silicon. This fortifies the thesis of an interaction between the silicon in the a-C:H matrix, the formation of silicon containing nanoparticles and the contacting aluminum.

Due to wear, the contact area between the coated ball and the aluminum sheet increases and subsequent the contact pressure decreases during the tribometer tests. According to the wear values (fig. 9), the contact pressure differs at the end of the run-in period depending on the silicon content. Thus, the contact pressure is another possible factor influencing the tribological behavior of silicon modified a-C:H coatings.
Conclusions

In this paper silicon modified amorphous hydrogenated carbon coatings (a-C:H:Si) were deposited to determine the adhesion tendency against aluminum EN AW 5083 as a function of the silicon content (Si = 0 at.-%, 8 at.-%, 16 at.-%, 25 at.-% and 34 at.-%). Therefore, ball-on-disc tribometer tests and strip drawing tests were conducted, which allow a reproduction of tribological loads with a high proximity to industrial dry forming processes. The following conclusions were made based on the tests results:

1. The adhesion tendency of a-C:H and a-C:H:Si (Si = 8%, 16%, 25% and 34%) coatings against aluminum EN AW 5083 is significantly lower than the adhesion tendency of an uncoated steel surface.
2. The average friction value of the a-C:H:Si coatings after the run-in period rises linear with an increasing silicon content.
3. The run-in duration of a-C:H:Si coatings extends with an increasing silicon content. Differing smoothing processes are a possible explanation for this tribological behavior. The duration and high friction value of the run-in period should be considered for a dry forming process of aluminum EN AW 5083 and is maybe a reason for the distinct formation of aluminum adhesions in the strip drawing tests.
4. The wear amount of the a-C:H:Si coatings and the aluminum EN AW 5083 sheets rises with an increasing silicon content. A non-correlation between the hardness of the coatings and the wear amounts indicates superposing wear mechanisms.
5. An incorporation of silicon influences the tribological behavior of a-C:H coatings sliding against aluminum EN AW 5083. The tribometer tests indicated interdependencies between the silicon and the aluminum which were aggravated by an increase of the silicon content.

Further tests are needed to investigate the interaction between the incorporated silicon in the a-C:H coating and the aluminum EN AW 5083. As a part of this investigation, it is to verify whether the tribological behavior of the tested a-C:H:Si coatings changes in a sliding contact with other aluminum alloys. Clarifying the interactions will lead to a better understanding of the tribological functionality of silicon modified a-C:H coatings and of the adhesion mechanisms in a sliding contact against aluminum EN AW 5083.

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Fig. 8: SEM-analysis of the intersection between the wear track and peripheral zone of Si 8% (a) and Si 34% (b).

Fig. 9: Average wear and hardness of the tested a-C:H and a-C:H:Si coatings.

Fig. 10: Average wear of the aluminum EN AW 5083 sheet as a function of the silicon content in the a-C:H:Si coating.
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