Dry Metal Forming Open Access Journal

Fast Manuscript Track

Edited by Frank Vollertsen
Available online at elib.suub.uni-bremen.de

www.drymetalforming.de

Dry Met. Forming OAJ FMT 2 (2016) 011–017
Received 12 April 2016; published 09 May 2016

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

Tim Abraham*[1], Günter Bräuer[1], Felix Kretz[2], Peter Groche[2]

[1]Fraunhofer Institute for Surface Engineering and Thin Films, Bienroder Weg 54 E, 38108 Braunschweig, Germany
[2]Institute for Production Engineering and Forming Machines, Otto-Berndt-Straße 2, 64287 Darmstadt, Germany

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

*E-mail address of corresponding author: tim.abraham@ist.fraunhofer.de

silicon concentration depended microstructure and nanomechanical properties of the a-C:H:Si coatings [9, 10].

However, the potential of a-C:H:Si coatings to reduce the strong adhesive wear in dry forming of aluminum 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 tetramethylsilane 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 beginning of the coating process. Furthermore, an interlayer based on pure TMS with a thickness of 0,5 µm was applied to improve the adhesion of the coating.

For a comparison with an established industrial coating system, an unmodified a-C:H coating (Si0%) was deposited by PACVD combined with physical vapour deposition (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, polished flat samples made of bearing steel (1.3505) and cold work steel (1.2379) and silicon wafers were coated and allowed a subsequent analyzation of the coatings. Coating hardness were determined with a commercial instrument (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 measured 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 application. The strip drawing test emulates the typical tribological load spectrum of deep drawing and stretch forming 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 sliding 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 aluminum. 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.

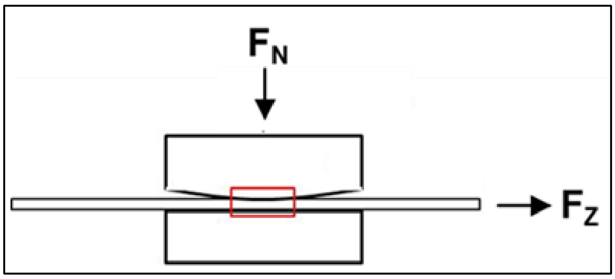

Fig. 1: Schematic of the strip drawing test

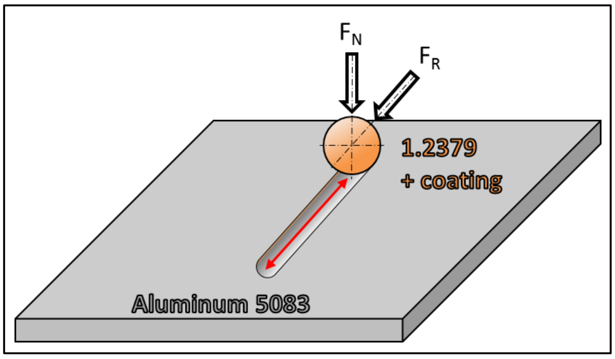

Fig. 2: Schematic of the oscillation ball-on-disc tribometer test

The 3D-structure of the adhesions on the tool surface is scanned and digitalized with confocal white light microscopy. The maximum height of the adhesions is taken as a quantitative indicator of the magnitude of the adhesive 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 uncoated 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

Tab. 1: Si-contents and properties of the tested a-C:H coatings

Coating	Si-Content [Atm.-%]	Thickness [µm]	Hardness [GPa]	Ra ball-on-disc [µm]	Ra strip drawing [µm]	Abrasive wear $[10^{-15} m^3/Nm]$
Si 0%	0	2.2	34.1	0.055	0.050	1.0
Si 8%	8	2.7	20.2	0.088	0.034	1.4
Si 16%	16	2.3	17.0	0.070	0.049	6.1
Si 25%	25.3	2.8	18.6	0.079	0.050	10.1
Si 34%	33.8	2.7	19.6	0.053	0.078	16.42

Tab. 2: Test parameters of the ball-on-disc tribometer and strip drawing test

Test	Load [N]	Initial contact stress [MPa]	Sliding speed $[mms^{-1}]$	Sliding distance [m]	Humidity [%]	Temperature [°C]
Ball-on-disc Tribometer	1	300	50	180	40-60	24.5
Strip-drawing	9000	~ 75	100	0,1	-	26.0

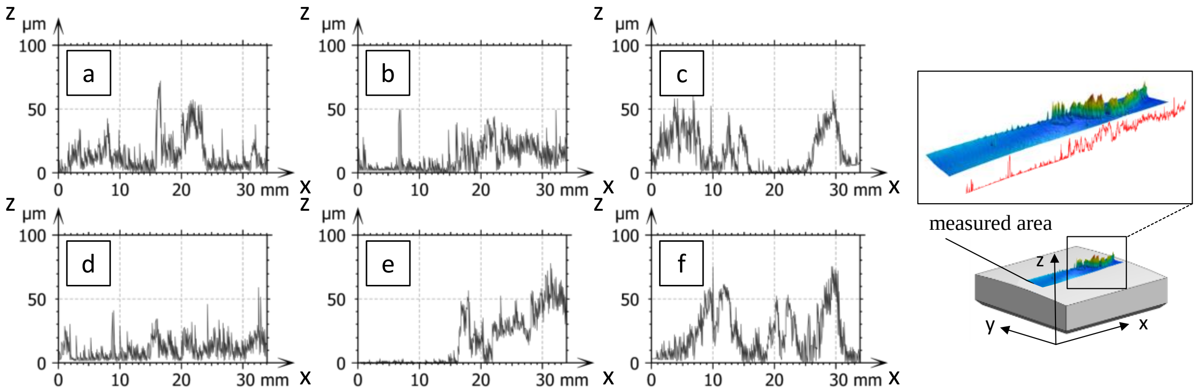

Fig. 3: Adhesions characterized by envelopes of a profile series for each tool configuration

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 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 spectroscope (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.

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 digitalized 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.

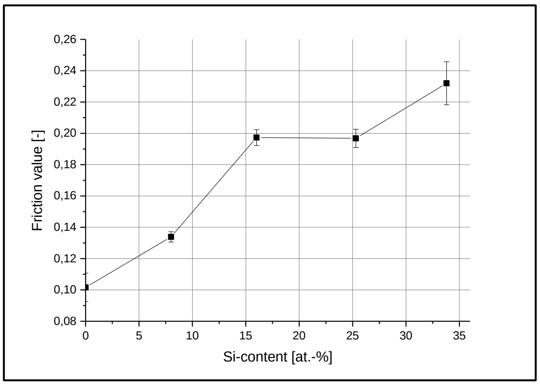

Fig. 4: Average friction value of the tested coatings after the run-in period

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 μ = 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 tri = 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 μ = 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.

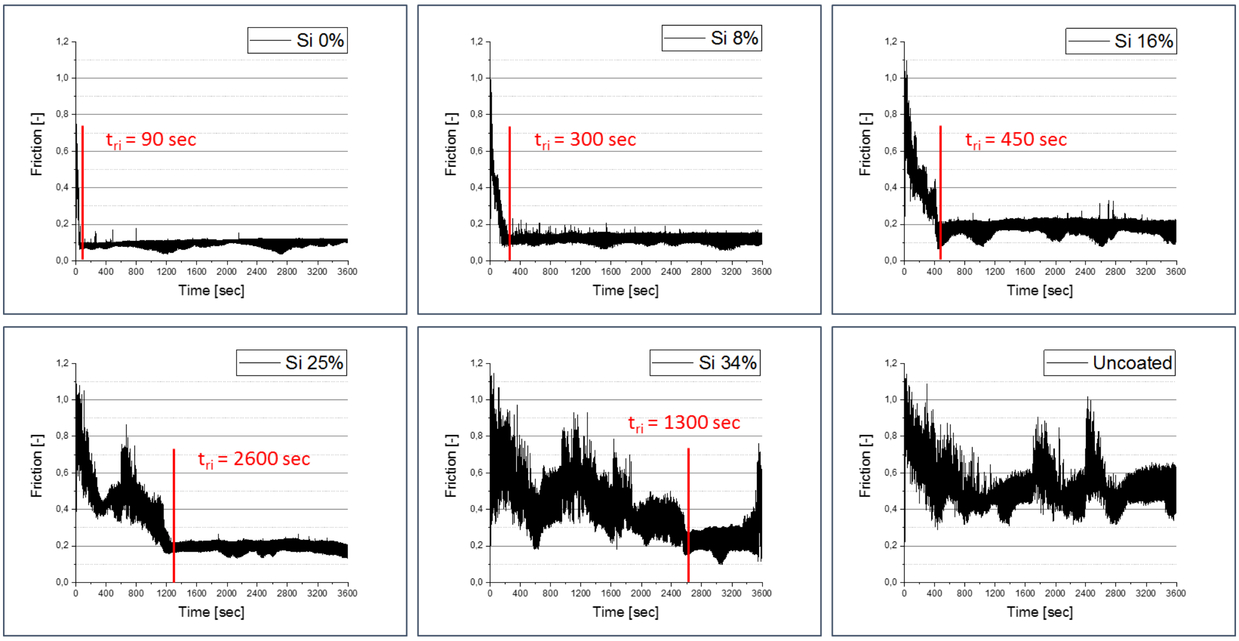

Fig. 5: Average friction development of the coated and uncoated steel balls sliding against aluminum EN AW 5083

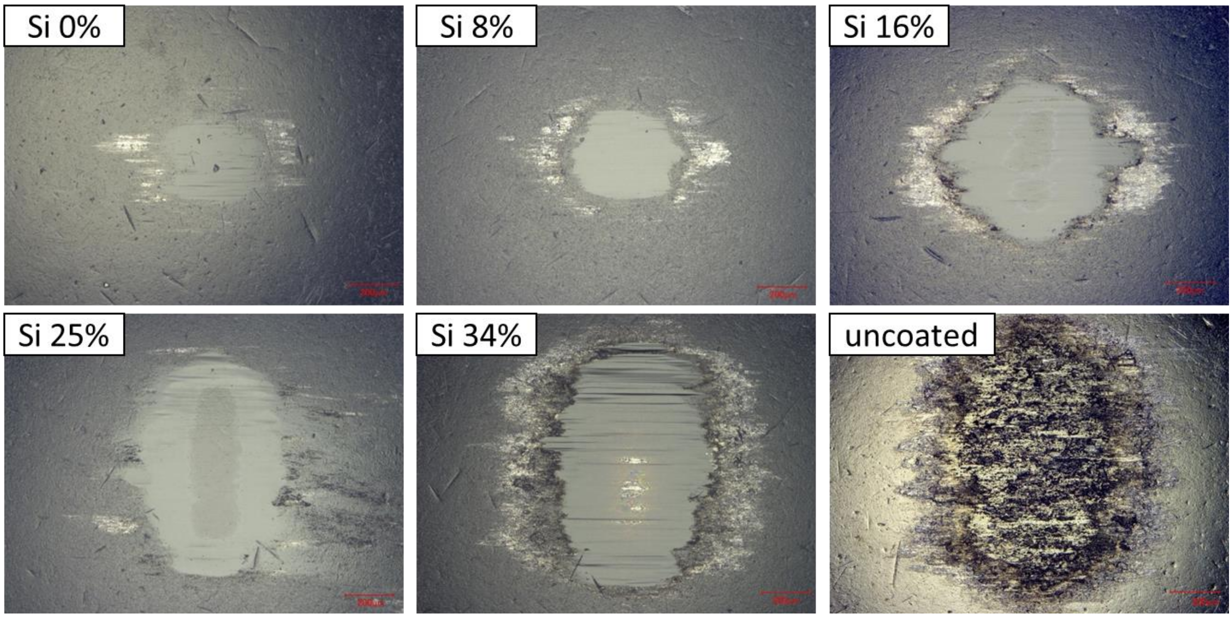

Fig. 6: Wear tracks of the coated and uncoated steel balls after sliding against aluminum EN AW 5083

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.

Fig. 7: SEM-image and EDX-mapping of aluminum adhesions on the exposed steel after a partial removal of the Si 34% coating

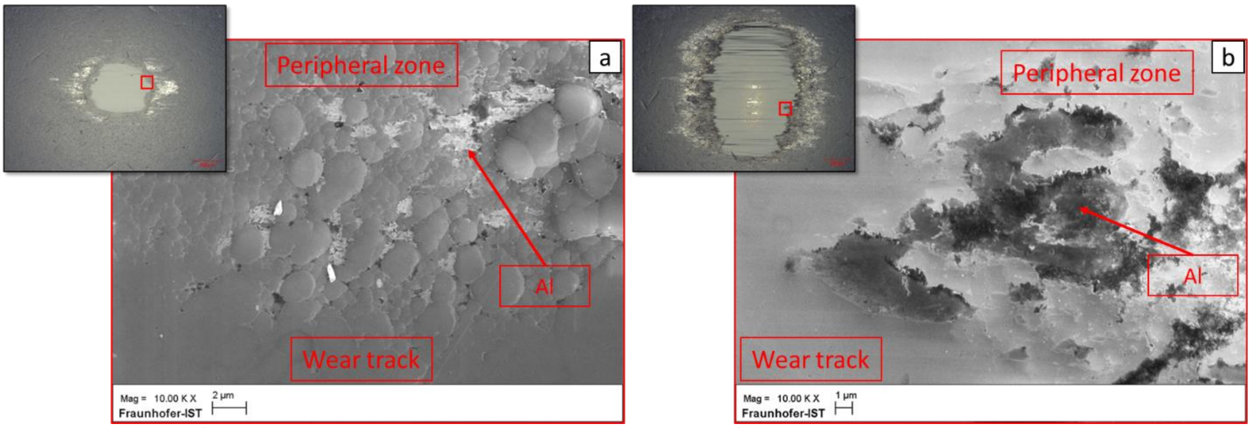

Fig. 8: SEM-analysis of the intersection between the wear track and peripheral zone of Si 8% (a) and Si 34% (b

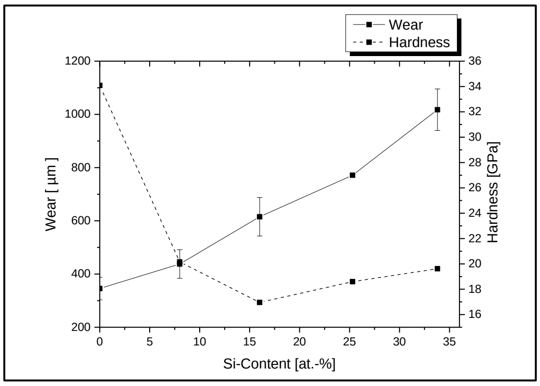

Fig. 9: Average wear and hardness of the tested a-C:H and a-C:H:Si coatings

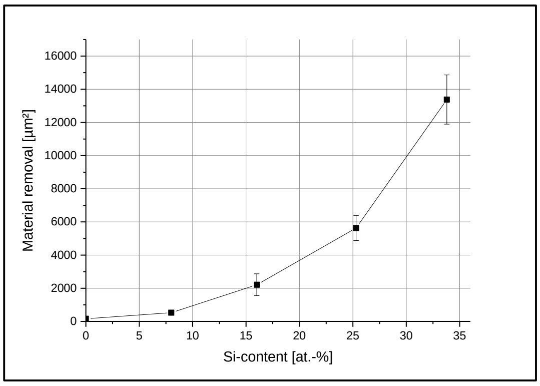

Fig. 10: Average wear of the aluminum EN AW 5083 sheet as a function of the silicon content in the a-C:H:Si coating

4 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.

Acknowledgements

The authors would like to thank the German Research Foundation (DFG) for funding of the project “Dry forming of aluminum alloys: from fundamental material and surface characterization to new tribological systems” received within the priority program SPP 1676 “Dry metal forming - sustainable production through dry processing in metal forming”.

Literatur

[1] P. Groche, F. Resch, 2015, Dry forming of aluminum alloys – Wear mechanisms and influencing factors, Materials Science and Engineering Technology 46

[2] K. Bewilogua, 2014, History of diamond-like carbon films – From first experiments to worldwide applications, Surface and Coatings Technology 242

[3] T. Horiuchi, S. Yoshihara, Y. Iriyama, 2012, Dry deep drawability of A5052 aluminum alloys sheet with DLC-coating, Wear 286-287

[4] M. Murakawa, N. Koga, S. Takeuchi, 1999, Diamondlike carbon-coating dies for deep drawing of aluminum sheets, Journal of Manufacturing Science and Engineering 121

[5] W. Ni, Y.-T. Cheng, A. M. Weiner, T. A. Perry, 2006, Tribological behavior of diamond-like-carbon (DLC) coatings against aluminum alloys at elevated temperatures, Surface and Coatings Technology 201

[6] K. Taube, B. Straehler, 1997, Beschichtung zur Verminderung der Umweltbelastung bei Umformprozessen, VDI Kolloquium „Schmiermittelarme Fertigung und Anwendungen“

[7] C. Donnet, 1998, Recent Progress on the tribology of doped diamond-like and carbon alloy coatings: a review, Surface and Coatings Technology 100-101

[8] K. Oguri, T. Arai, 1991, Tribological properties and characterization of diamond-like carbon coatings with silicon prepared by plasma-assisted chemical vapour deposition, Surface and Coatings Technology 47

[9] J.F. Zhao, P. Lemoine, Z.H. Liu, J.P. Quinn, P. Maguire, 2001, A study of microstructure and nanomechanical properties of silicon incorporated DLC films deposited on silicon substrates, Diamond And Related Materials 10

[10] J. Meneve, R. Jacobs, L. Eersels, J. Smeets, E. Dekempeneer, 1993, Friction and wear behaviour of amorphous hydrogenated $Si_{1-x}C_x$ films, Surface and Coatings Technology 62

[11] M. Weber, 2009, Tool coatings for the cold and hot forming of aluminium and magnesium alloys, 10. Umformtechnisches Kolloquium Darmstadt

[12] M. Murakawa, S. Takeuchi, 2003, Evaluation of tribological properties of DLC films used in sheet forming of aluminum sheet, Surface and Coating Technology 163-164

[13] T. Michler, C. Siebert, 2003, Abrasive wear testing of DLC coatings deposited on plane and cylindrical parts, Surface and Coatings Technology 163-164

[14] W.-J. Wu. T.-M. Pai, M.-H. Hon, 1998, Wear behavior of silicon-containing diamond-like carbon coatings, Diamond and Related Materials 7

[15] K. Oguri, T. Arai, 1992, Two different low friction mechanisms of diamond-like carbon with silicon coatings formed by plasma assisted chemical vapour deposition, Journal of Material Research 7