

PVD-/PECVD-DLC Thin Coatings as a Potential Solution for Tailored Friction Conditions for Dry Sheet Metal Forming Tools

R. Zhao^{1,a}, S. Tremmel^{1,b}

1. Engineering Design, Friedrich-Alexander-Universität Erlangen-Nürnberg,

Martensstraße 9, 91058 Erlangen, Germany

^azhao@mfk.fau.de, ^btremmel@mfk.fau.de

ABSTRACT

Diamond-like carbon (DLC) is a series of amorphous carbon material. Due to its excellent tribological characteristics it is applied as coatings on forming tools for lubricant-free processes. In this research, three types of DLC coatings (a-C:H, a-C:H:W and ta-C) were deposited on polished tool steel substrate and then characterized regarding their coating thickness, roughness and mechanical properties. Tribological tests under lubricant-free conditions were performed using a ring-on-disc tribometer. As a reference, tribological tests with uncoated tool steel under the same conditions were conducted. In addition, scanning electron microscope (SEM) analysis on worn surfaces was performed to identify the dominant friction and wear mechanisms. Furthermore, energy dispersive X-ray spectroscopy (EDX) measurements of metallic adhesions on worn surfaces were carried out to determine their elemental contents. Based on acquired results the application potentials of these three kinds of DLC coatings for real forming processes, especially for metals with high adhesion tendency, like aluminium alloys, were evaluated.

KEYWORDS: DLC Coatings, Dry sheet forming, adhesive wear

1 INTRODUCTION

The enhanced environmental awareness and limited global oil resources motivate to raise ecological efficiency of forming processes with minimal lubricant usage /1/. The lubricant-free production processes have a number of ecological and economic benefits. Besides the abandonment of some lubricants with environmentally hazardous substance, expensive costs to remove the remained lubricant rest from the formed workpiece are saved. However, to achieve a lubricant-free forming process for mass production many challenges have to be faced. Due to direct contact between workpiece and tool surface high friction and adhesive wear on the forming tool surfaces are the biggest problems. For applications of mass production tool service life for dry processes has to be prolonged. One of the concepts to realize such dry forming process is to generate tailored friction conditions on the tool surface. Thus, the material flow can be accurately controlled with minimal wear remaining on the tool surface and die life is prolonged.

Diamond-like carbon (DLC) coatings are one of the potential solutions to reduce friction and wear under lubricant-free conditions. It is possible to alloy diverse metallic or non-metallic elements in their carbon network. In comparison to pure carbon coatings, coating properties and multifunctionality of these doped carbon coatings are improved /2/. According to the references /3/ and /4/, the tungsten doped amorphous carbon coating variants (a-C:H:W) showed their potentiality for sheet bulk metal forming processes, even under highly loaded conditions. It was reported in /5/ that the a-C:H:W coating on the forming tool surface can reduce the adhesive wear against steel sheet. In /6/ the tribological behavior of a-C:H coatings applied to sheet-forming tools under process relevant conditions are shown. The tetrahedral hydrogen free amorphous carbon coating system (ta-C) used on deep drawing dies showed good anti-wear property /7/. A lower coefficient of friction /5/ against aluminum sheet during sliding processes without lubricant was reported. This is due to its high hardness and lower adhesion tendency to aluminum- and magnesium-based light metals in comparison to other carbon coating types /8/.

The friction coefficients in the majority of studies on amorphous carbon coatings are measured by a ball-on-disc setup. It involves a circular point contact. However, tribological behaviour in surface-contact has been reported rarely. In this paper, three representative amorphous carbon coatings were explored. Their tribological behaviors against industrial common used steel and aluminum sheets were investigated by a ring-on-disc-tribometer in surface contacts.

2 EXPERIMENTAL DETAILS

2.1 Experimental setup

The friction and wear tests under lubricant-free conditions were conducted using a ring-on-disc-tribometer (Wazau, TRM 1000) under ambient atmosphere with $RH = 43 \% \pm 2.4 \%$ relative humidity at $T = 23.2 \pm 0.4 \text{ }^\circ\text{C}$. The tribological system consists of a unidirectional rotating ring and a fixed sheet material disc. A schematic diagram of the test configuration is shown in Figure 1. A tool steel ring (X155CrVMo12, 1.2379) coated with different amorphous carbon coatings with inner and outer diameter of 10 mm and 20 mm was loaded against a flat squared-shaped metal sheet. Prior to tribological testing the specimens were carefully cleaned with isopropanol. The experiment was repeated three times. The detailed test conditions are summarized in Table 1.

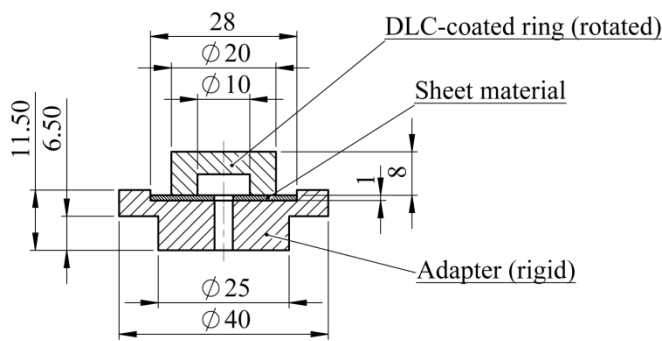


Figure 1: Cross section of the ring-on-disc-tribometer

Table 1 Parameters of the tribological tests

Parameter	Value
Base body	Square-shaped sheet materials with electrical discharge textured: hot-dip galvanized DC04 (1.0338), AA6014 (EN AW-6014), AA5182 (EN AW-5182)
Counter body	Ring ($\text{\O}10/20 \text{ mm}$): ta-C, a-C:H, a-C:H:W
Movement mode	rotatory, continuously
Normal load	500 N ($\triangleq 2.1 \text{ MPa}$)
Rotate velocity	120 min^{-1}
Distance	10 m

2.2 Coating deposition

The coating specimens were deposited using PVD and PECVD techniques. The substrates were ring-shaped from tool steel and hardened to $60 \pm 1 \text{ HRC}$. The surfaces of the uncoated substrates were lapped and polished to a roughness of $R_z = 0.9 \pm 0.1 \text{ }\mu\text{m}$ and $R_{pk} = 0.1 \pm 0.01 \text{ }\mu\text{m}$, which represents the surface qualities of conventional deep drawing tools. The substrates for the ta-C coating were exceptionally coating-suitable polished to a roughness of $R_z = 0.16 \pm 0.01 \text{ }\mu\text{m}$ and $R_{pk} = 0.03 \pm 0.01 \text{ }\mu\text{m}$.

The tetrahedral hydrogenfree amorphous carbon coating system (ta-C) consists of an adhesive layer of Cr and the ta-C functional layer. For a smooth coating surface the Cr adhesive layer of about 600 nm was generated by sputtering. The ta-C coating of about 700 nm was manufactured by laser arc process. The arc process was initiated by a laser pulse on the graphite target /9/. In order to ensure a smooth coating, a

magnetic field was applied to remove the macro particles from the arc process. After the deposition the coated specimens were mechanically treated by polishing and brushing with diamond paste with a grain size of 3 μm . The hydrogenated amorphous carbon coating (a-C:H) was deposited by using a hybrid PVD/PECVD coating machine (H-O-T, TT 300) and a twofold rotating charging rack. For the Cr adhesive layer and the WC interlayer arc evaporation and unbalanced magnetron sputtering were used as coating technologies, respectively. The a-C:H coating functional layer was deposited by plasma enhanced chemical vapour deposition using acetylene as precursor gas. The substrate bias voltage U_{bias} and the argon flow $\phi(\text{C}_2\text{H}_2)$ in the deposition chamber were 450 V and 220 sccm. The tungsten doped hydrogenated amorphous carbon coatings (a-C:H:W) functional layer was deposited by reactive unbalanced magnetron sputtering of a binder-free WC target using MF power supply (40 kHz, pulse width 5 μs) in an argon-acetylene atmosphere. The substrate bias voltage U_{bias} and the argon flow $\phi(\text{Ar})$ in the deposition chamber were 130 V and 180 sccm. Prior to tribological tests both of the coating surfaces were polished using 3 μm and 1 μm diamond suspension.

2.3 Coating properties

The surface roughness of the coating specimens was measured after polishing process. The average distance between the highest peak and lowest valley R_z and the reduced peak height R_{pk} values were characterized by tactile stylus measurements according to DIN EN ISO 4287 /10/. The interfacial adhesion between the coating and the substrate was measured by scratch test according to DIN EN 1071-3 /11/. Using an optical microscope the normal force at which the first failure of the coating L_{c1} was evaluated. The crater grinder method was applied to measure the coating thickness according to DIN EN 1071-2 /11/. Micro hardness of 20 points on different places of the coatings was determined by Vickers indentation (Fischer, FISCHERSCOPE H100) with indentation force of 20 mN according to DIN EN ISO 14577-1 /12/. The coating designations und their essential properties are summarized in Table 2.

Table 2 Coating samples for the ring-on-disc tribological tests

Coating	Thickness	Adhesion	Hardness	Indentation modulus	Surface roughness	
	t in μm	L_{c1} in N	HV 0.002	E_{IT} in GPa	R_z in μm	R_{pk} in μm
ta-C	1.33 ± 0.03	12.0	$5\ 017.9 \pm 1\ 164.0$	330.6 ± 52.4	0.26 ± 0.01	0.045 ± 0.005
a-C:H	2.40 ± 0.10	26.5	$1\ 287.2 \pm 187.3$	126.9 ± 15.1	0.59 ± 0.07	0.05 ± 0.009
a-C:H:W	2.35 ± 0.05	29.3	690.7 ± 155.7	86.1 ± 7.5	0.63 ± 0.08	0.05 ± 0.007
Substrate	–	–	642.8 ± 88.2	164.6 ± 16.7	0.90 ± 0.10	0.10 ± 0.01

In order to identify different wear mechanism under lubricant free conditions in surface-to-surface contact, the tested surfaces were investigated by scanning electron microscope SEM (Zeiss, Cross Beam 1540), especially the areas with adhesive wear. Energy dispersive X-ray (EDX) analyses of metallic adhesions were conducted to identify the chemical compositions.

3 RESULTS

3.1 Friction coefficients

Figure 2 demonstrates the mean friction coefficient μ_{rod} from a distance of 2.5 to 10 m against three sheet materials. As reference test the uncoated steel substrate was applied, which was polished to the same R_{pk} like the ta-C coated surface. In general all of the three DLC coatings show lower friction coefficient than the uncoated tool steel and thus, exhibit strong potential for reduction of friction.

The results of the reference test show high friction coefficients under lubricant-free conditions. The friction coefficients are 0.71 for steel/DC04, 0.53 for steel/AA6014 and 0.39 for steel/AA5182. The light metals, like aluminium and magnesium in AA5182, formed a continuous, dry and adherent

friction-reducing lubricant layer on the tool steel's surface, thus the friction coefficient is relatively low, even without additional lubrication. The application of the ta-C coating leads to a reduction of friction, especially during the sliding against DC04 with over 50 % reduction of the friction coefficient. On other hand, tests of ta-C coatings against both aluminum alloys show only slightly reduction of friction in comparison with results against steel sheet. Considering the a-C:H coating as second potential candidate, it is noticeable in Figure 2 that it shows significantly better friction behaviour against AA6014 than against AA5182. The friction coefficient of a-C:H against AA6014 is 0.26, which is about 54 % lower than that of the reference test. The friction coefficients of a-C:H against AA5182 and a-C:H against DC04 are 0.37 and 0.44, respectively. Whereas the standard deviation of μ_{rod} for a-C:H/DC04 is exceptional high despite of additionally repeats of the test. The reason may be Fe adhesions stuck to the a-C:H surface. However, adhesions are irregularly distributed on the ring surface, which led to strongly scattering results. The tests of a-C:H:W coatings against diverse sheet materials show always similar friction coefficients with 0.44 for a-C:H:W/DC04, 0.47 for a-C:H:W/AA6014 and 0.44 for a-C:H:W/AA5182.

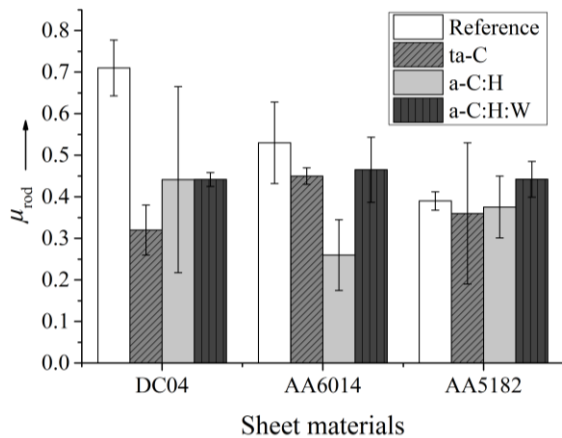


Figure 2: Mean friction coefficient μ_{rod} of DLC coating samples ($n = 3$) against diverse sheet materials for sliding distance of 2.5 to 10 m

3.2 Characterization of wear surfaces

Figure 3 summarizes microscopic images of wear surfaces against diverse sheet materials. As shown in Figure 3, spot like steel adhesions transferred from the DC04 sheet were observed on the ring surfaces (No. 01 to No. 03) due to adhesion and galling of the steel/steel material pairing. As seen on samples No. 04 to No. 09, the adhesions from aluminium sheets are ring-like and distributed over the inner and outer areas of the ring surfaces.

All the carbon layers were not delaminated and not completely worn out under all test conditions. For all the wear surfaces coated with ta-C high adhesion resistance behaviour was observed. In the tests against DC04 no visible adhesion was detected on rings of No. 11 to No. 13. For a-C:H/DC04 thin ring-shaped wear tracks in terms of steel adhesion was observed in the outer ring areas, especially on the rings No. 21 and No. 23. However, in the inner ring area no wear track or any adhesive wear was observed. It is noticeable, that on the a-C:H:W surface, which was also tested against DC04, no obvious adhesive wear was overserved. But the tested ring surfaces can be divided into an inner and an outer ring area, e.g. the sample No. 32. Due to the higher velocity compared to that of the inner ring area, the outer area was smoothed and the inner area remained in its original state. Despite of different wear appearances and wear mechanism, these two kinds of amorphous carbon coatings exhibits the same mean friction coefficient against DC04 sheet. The extreme high standard deviation for the material pairing a-C:H/DC04 implied, that the friction was relative dependent on the formed adhesion in the outer ring area. The more adherent wear on the ring, the higher are the friction coefficients caused by cold-welded Zn-Fe interfaces. Considering this fact that the a-C:H:W exhibits a stable friction coefficient and little adhesive wear, its application potential is higher if the tool is applied for deep drawing of DC04 sheet.





































Specimens →	Uncoated Steel	ta-C	a-C:H	a-C:H:W
DC04	 01	 11	 21	 31
	 02	 12	 22	 32
	 03	 13	 23	 33
AA6014	 04	 14	 24	 34
	 05	 15	 25	 35
	 06	 16	 26	 36
AA5182	 07	 17	 27	 37
	 08	 18	 28	 38
	 09	 19	 29	 39

Figure 3: Wear tracks on tested ring surfaces against diverse sheet materials

The coated surfaces were also tested against two aluminium alloys under the same conditions as shown in Figure 3. The tests of ta-C coatings against aluminium alloys showed significantly less adherent areas on rings No. 14 to No. 19 than that of the uncoated ones. In contrast to the other coatings, the a-C:H coating behave differently when tested against the two aluminium alloys: against AA6014 (No. 24 to No. 26) no visible adhesive wear was observed after a sliding distance of 10 m; but against AA5182 material was to some extent transferred to the ring surface. The reason may be the higher magnesium content in AA5182 than that in AA6014. The AA5182 sheet contains 4.0 to 5.0 wt.% Mg, whereas in AA6014 only 0.4 to 0.8 wt.%. The a-C:H:W coating shows in general no anti-adhesion effect against both aluminium alloys due to its high tungsten content in the amorphous carbon net of 7.2 g/m², which was measured by X-ray fluorescence spectroscopy analysis (XRF).

3.3 SEM investigations

Figure 4 (a) shows the ta-C coated surface after brushing. The surface reveals a typical polished structure with a few valleys and peaks. Due to its high fraction of diamond like carbon bonds (sp^3 bonds) the ta-C coating exhibit extremely high hardness and abrasive wear resistance [13]. In Figure 4 (b) only little adhesive wear was detected, which implies a low tendency of adhesion of ta-C against aluminium compared to tool steel. As showed in Figure 3, on the surface of sample No. 16 a bright adhesive ring was formed during tribometer testing, which results from these spot like adhesions in Figure 4 (b). They stick to more and more aluminium swarf departed from aluminium sheet during sliding. In this way the adhesions propagate until the surrounding area is all covered with aluminium material.

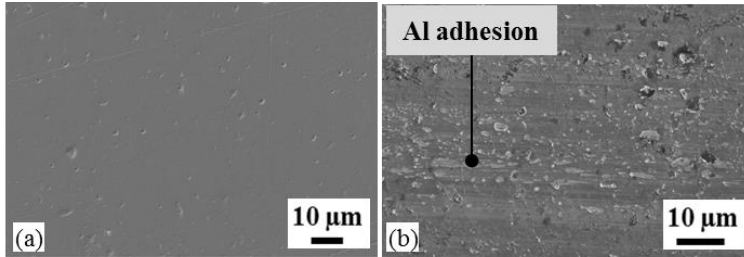


Figure 4: (a) ta-C coated surface before testing and aluminium adhesions on the worn ta-C coated surface taken from sample No. 16

Similar to the ta-C coating, the a-C:H coating also shows anti-adhesion behaviour to metallic sheets, as shown exemplarily in Figure 5, where only little Zn adhesion could be observed. Observing the adhesions closer, the interfaces as a result of the cold welding between tool and workpiece, can be seen. x

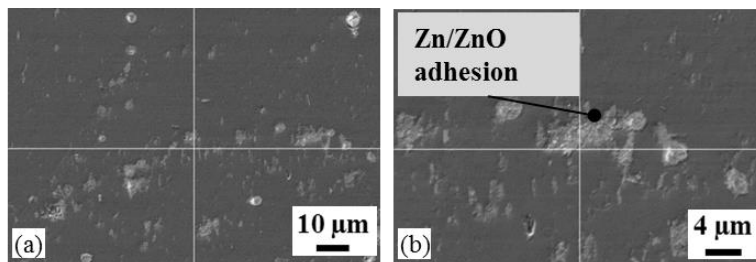


Figure 5: (a) Zn/ZnO adhesions from DC04 transferred on the a-C:H coated surface taken from specimen No. 23 and (b) interfaces as a result of cold welding

EDX analysis results of a worn a-C:H surface against DC04 is shown in Figure 6 (a). The red region mainly consisted of O and green of Zn. Blue region of Fe was expressed at low to undetectable level. It is observed in Figure 6 (b) against AA5182 that the adhesion mainly consisted of O in red, Mg in green and Al in blue.

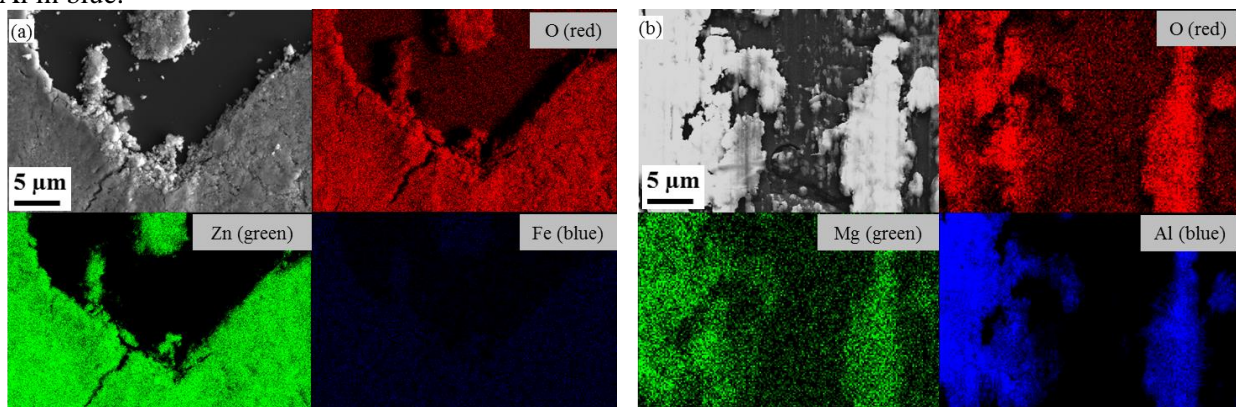


Figure 6: (a) EDX analysis of a a-C:H coated surface against DC04 taken from the specimen No. 23 and (b) against AA5182 taken from the specimen No. 36

Before investigating the worn a-C:H:W coated surfaces, it is necessary to clarify the original characteristic surface texture of the a-C:H:W coating. Figure 7 (a) and (b) shows SEM images of a representative a-C:H:W coated surface after deposition before and after mechanical post treatment. Figure 7 (a) depicts that surface asperities due to small particles are distributed on the surface, which result from a large number of macro-particles (droplets) entrapped in the Cr adhesive layer. After polishing the surface asperities in the form of particles grains were flattened.

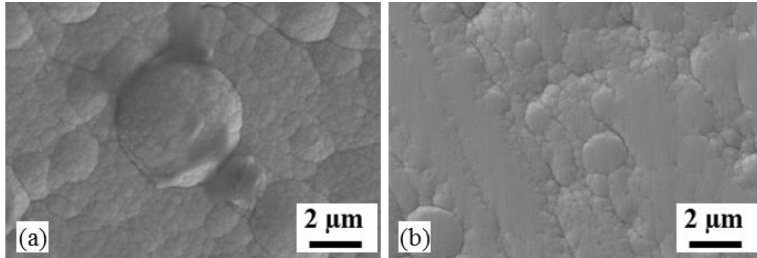


Figure 7: (a) Original and (b) polished a-C:H:W coated surface

Figure 8 (a) shows the aluminium adhesions distributed on the a-C:H:W coated surface after tribometer testing. Observing the adhesions closer in Figure 8 (b), the initial aluminium adhesion was interlocked by a sharp particle edge, which results from the polishing process.

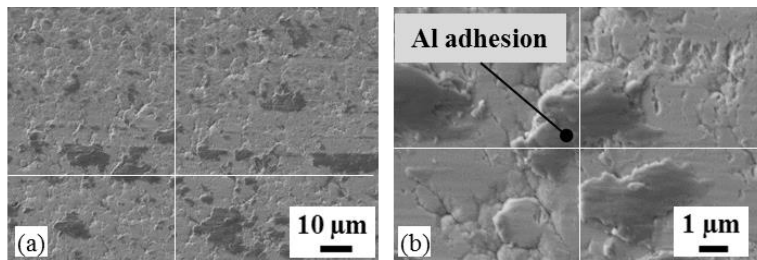


Figure 8: (a) Aluminium adhesions on the worn a-C:H:W coated surface and (b) adhesions caught by sharp edges of the flattened a-C:H:W grains taken from sample No. 36

Figure 9 (a) showed the Fe adhesions distributed on an a-C:H:W coated surface. Similar to the worn surface against AA6014 in Figure 8, the adhesions were trapped between the valleys around particle grains. Due to the characteristic coating surface texture as has been shown in Figure 7, which consists of particle-like grains, the effective contact surface is much larger than on a-C:H and ta-C coatings. The interlocked adhesions stores in the valleys between the particle grains during sliding. The remained adhesions may lead to subsequently growth of furthermore metallic adhesions. For this reason, the coating texture is undesired for the forming tool surfaces, since these textures increasing the growth of adhesive wear on the tool surface.

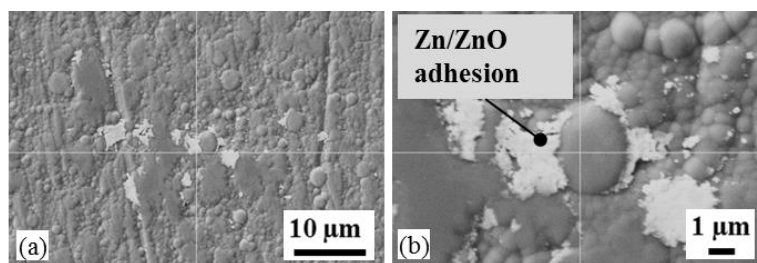


Figure 9: (a) Fe adhesions on the worn a-C:H:W surface and (b) adhesions trapped between valleys around polished particle grains taken from sample No. 32

4 CONCLUSION AND OUTLOOK

In general the amorphous carbon coatings have great potential in solving tribological problems under lubricant-free conditions, such as increasing friction and metallic adhesive wear resulting from a direct

surface-to-surface contact. Due to its high hardness and low adhesion tendency to metals, the anti-adhesion properties of ta-C and a-C:H coatings prevent the material transfer from metallic sheets to the tool surface and as a result the friction for dry forming is reduced. The a-C:H:W coating shows a stable friction-reducing behaviour against steel sheet DC04. However, its characteristic surface texture, even after polishing, offers place to storage metallic adhesions in valleys between particle grains and flattened surface asperities. This may be problematic, as the coating is used for tools against metals with high adhesion tendency, such as aluminium and magnesium. Because the adhesions tend to remain on the tool surface and thus lead to quickly growth and propagation of furthermore adhesions.

For prospective investigations different non-metallic dopants (Si, O, N etc.) in amorphous carbon coatings will be tested. Furthermore an open tribological system will be used in place of the ring-on-disc model to prevent the influence of loose wear particles and metallic swarf resulting from damaged workpiece, which better simulates the real operation conditions in deep drawing processes. In addition, different operation surface pressures and operation velocities will also be investigated.

5 Acknowledgement

The authors thank the German Research Foundation (DFG) for supporting the presented investigations by funding the priority program SPP 1676 (Dry Metal Forming) project TR 1043/1-1 with the project title "Lubricant free forming with tailored tribological conditions". Furthermore the authors would like to gratefully thank lab technician Mrs. U. Wolf, Mr. M. Kolb from Institute of General Materials Properties for EDX images, as well as student assistants Mr. S. Frühwald and Mr. J. Riedel for performing the tribological experiments.

6 REFERENCES

- /1/ *F. Vollertsen, F. Schmidt*, 2014, Dry metal forming: Definition, chances and challenges, *International Journal of Precision Engineering and Manufacturing-Green Technology* 1
- /2/ *C. Donnet, A. Erdemir*, 2008, *Tribology of Diamond-Like Carbon Films. Fundamentals and Applications*. 1. Edition, New York: Springer
- /3/ *H. Hetzner, R. Zhao, S. Tremmel, S. Wartzack*, 2013, Tribological adjustment of tungsten-modified hydrogenated amorphous carbon coatings by adaption of the deposition parameters. *Proceedings for the 10th International Conference THE "A" Coatings*
- /4/ *H. Hetzner, C. Schmid, S. Tremmel, K. Durst, S. Wartzack*, 2014, Empirical-Statistical Study on the Relationship between Deposition Parameters, Process Variables, Deposition Rate and Mechanical Properties of a-C:H:W Coatings. *Coatings* 4
- /5/ *M. Merklein, M. Schmidt, S. Tremmel, S. Wartzack, K. Andreas, T. Häfner, R. Zhao, J. Steiner*, 2015, Investigation of Tribological Systems for Dry Deep Drawing by Tailored Surfaces, *Dry Metal Forming - OAJ FMT* 1
- /6/ *A. Ghiotti, S. Bruschi*, 2011, Tribological Behaviour of DLC Coatings for Sheet Metal Forming Tools. *Wear* 9-10
- /7/ *M. Murakawa, N. Koga, T. Kumagai*, 1995, Deep-drawing of aluminium sheets without lubricant by use of diamond-like carbon coated dies. *Surface and Coating Technology* 76-77
- /8/ *S. Bhowmick, A. Banerji, M.Z.U. Khan, M.J. Lukitsch, A.T. Alpas*, 2015, High temperature tribological behavior of tetrahedral amorphous carbon (ta-C) and fluorinated ta-C coatings against aluminum alloys. *Surface and Coating Technology*. In Press.
- /9/ *T. Stucky, U. Baier, C.-F. Meyer, H.-J. Scheibe, B. Schultrich*, 2003, Großflächenbeschichtung mit superhartem Kohlenstoff. *Vakuum in Forschung und Praxis* 6
- /10/ *German Institute for Standardization (DIN) DIN EN ISO 4287*. Berlin: Beuth, 2010
- /11/ *German Institute for Standardization (DIN) DIN EN 1071*. Berlin: Beuth, 2003
- /12/ *German Institute for Standardization (DIN) DIN EN ISO 14577*. Berlin: Beuth, 2003
- /13/ *Y. Mabuchi, T. Higuchi, V. Weinhacht*, 2013, Effect of sp²/sp³ bonding ratio and nitrogen content on friction properties of hydrogen-free DLC coatings. *Tribology International* 62