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# Amorphous hydrogenated carbon thin films deposited on stainless steel using high energy plasma focus device



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### article info abstract

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Amorphous hydrogenated carbon (a-C:H) films have been deposited on stainless steel — AISI 304 substrates using a high energy plasma focus device in acetylene and acetylene/hydrogen discharge. The influence of hydrogen content on the microstructural and mechanical properties of hydrogenated amorphous carbon (a-C:H) films has been studied in detail. The films were deposited with the same numbers of focus shot. They were placed at the same distance from anode tip but at different angular positions with respect to the anode axis. The results of Raman spectroscopy show successful deposition of (a-C:H) films. Also, it confirms the influence of hydrogen content on the diamond character. The peak intensity ratio of the D-band to G-band  $(I_D/I_C)$ , the G-peak position and the full width at half maximum of the G-peak, FWHM (G), are used to characterize (a-C:H) films. The total hydrogen and copper atomic contents were obtained from elastic recoil detection analysis (ERDA) experiments and Rutherford backscattering spectrometry (RBS) analysis, respectively. Their results confirm the dependency of hydrogen and copper atomic contents on the angular positions. Field emission scanning electron micrographs show the granular surface morphology of the deposited films. X-ray diffraction (XRD) analysis confirms the amorphous structure of the films. The hardness measurement performed on the samples shows that the sample hardness is significantly improved.

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### 1. Introduction

Carbon is one of the most interesting elements because of its three different chemical states, including  $sp^3$ ,  $sp^2$  and  $sp^1$  chemical coordinations [\[1\]](#page--1-0). It can also exist in both crystalline and amorphous structures. Graphite and diamond have crystalline forms which exhibit different bonding structures and different physical properties. Graphite has  $sp^2$  bonding structure, and so it has three covalent bonds in a layered planar structure. In each layer, the carbon atoms are arranged in a honeycomb lattice. The fourth electron can move freely in the plane, paving the way for anisotropy physical properties. Diamond has  $sp<sup>3</sup>$ bonding structure by four covalent bonds transferring interesting properties such as extreme hardness, thermal conductivity and wide band gap [\[2](#page--1-0)–4]. These properties make it one of the most desirable materials for industrial applications.

Carbon also exists in amorphous structure, without any crystal form. Diamond-like carbon (DLC) has an amorphous structure with significant fraction of  $sp^3$  C–C bonds [\[2\]](#page--1-0). DLC films have a wide range of applications in industry because of their exceptional properties such as high mechanical hardness, high corrosion resistance, low friction and wear coefficients [5–[8\]](#page--1-0), optical transparency [\[8,9\]](#page--1-0) and wide band gap as a semiconductor [\[8,9\]](#page--1-0). So far, DLC films have been synthesized by a variety of techniques such as vacuum arc deposition [\[10\],](#page--1-0) pulsed laser ablation [\[11,12\]](#page--1-0), chemical vapor deposition [\[13\]](#page--1-0) and deposition by plasma focus [\[1,12,14](#page--1-0)–17].

Since hydrocarbon precursors are usually applied in synthesis procedure of amorphous carbon, synthesized materials consist of not only carbon but also hydrogen atoms. This type of material is known as hydrogenated amorphous carbon (a-C:H) which exhibits lower friction and wear coefficient in comparison with hydrogen free DLCs [\[3\].](#page--1-0) These properties and their relatively high hardness make them suitable to be used in tribology fields. Physical, mechanical, optical and electrical properties of a-C:H films strongly depend on their hydrogen contents. Therefore, some literatures have studied the effect of the hydrogen content on the properties of DLC films [\[18](#page--1-0)–20].

Plasma focus device, is a discharge system which can compress plasma to a high density (~10<sup>25</sup>–10<sup>26</sup> m<sup>-3</sup>) and high temperature  $(-1-2 \text{ keV})$  for a short period of time  $(-10^{-7} \text{ s})$  using self-generated magnetic field [\[21\]](#page--1-0). Historically, this device was developed as a fast neutron source. Later, some applications in other areas such as an appropriate source for energetic electrons and ions and pulsed X-ray. Previous experiments show that the plasma focus devices emit highly energetic (hundreds of keV to tens of Mev) ions [\[22\].](#page--1-0) Also, the energetic ions of this system have been used for deposition of thin films [\[1,12,14](#page--1-0)–17,22]. Although, there are various methods for deposition of various thin

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films, deposition by plasma focus has attracted particular attention, because of its attractive features such as high deposition rate, energetic deposition and deposition with reactive working gases [\[22\]](#page--1-0).

The diamond-like carbon thin films have previously been deposited by some researchers, utilizing the plasma focus devices. Shaista Zeb et al. [\[16,17\],](#page--1-0) Z.P. Wang et al. [\[12\]](#page--1-0) and E. Ghareshabani et al. [\[14\]](#page--1-0) deposited diamond-like carbon films on Si substrate using graphite target as a carbon source with different working gases. They reported successful growth of diamond-like carbon films and investigated the films' characteristics.

Recently, M. Hassan et al. [\[1\]](#page--1-0) reported the successful diamond-like carbon coating on stainless steel by the plasma focus device using methane as a working gas. In this research, the hydrocarbon gas was used as a carbon source. Also the characteristics of DLC films were studied.

In this work, we shall report the deposition of a-C:H thin films with different hydrogen contents, at room temperature on stainless steel — AISI 304 substrates, using high energy plasma focus device. Also, we shall investigate the structural, mechanical properties and surface morphology of the deposited thin films.

To the best of our knowledge, this is the first time that a-C:H films are derived from acetylene and the mixture of acetylene/hydrogen gas using high energy plasma focus device. Also, the effect of hydrogen content on the structural and mechanical properties of a-C:H thin films, prepared by plasma focus device has not been reported.

### 2. Experimental setup

Fig. 1 shows schema of our experimental setup used for deposition of DLC thin films. This device consists of 24 capacitors (6 μF) which can be charged up to 40 kV. Its vacuum chamber includes a cylindrical brass made anode with 220 mm length and 125 mm diameter. The anode is surrounded by 12 equidistance cylindrical copper cathodes in a squirrel cage shape. Each cathode has a length of 220 mm with 10 mm diameter. The central anode has been separated from the surrounding cathodes by a Pyrex glass insulator with 125 mm inner diameter and 3 mm thickness covering 50 mm of anode length. The maximum energy and current are about 115 kJ and 1.2 MA, respectively. The capacitors are connected to the chamber through the spark gap system. The breaking voltage can be variable using a trigger system.

Electrical energy will transfer to the anode and it will result in electrical discharge on the surface of the insulator. This phase is known as breakdown phase, which produces plasma sheath on the surface of the insulator. The axial acceleration phase starts when the plasma sheath is detached from the insulator and accelerates toward the open



Fig. 1. Schema of plasma focus system.

end of the anode because of Lorentz force caused by self-generated magnetic field and ends by reaching the plasma sheath at the end of the anode. The plasma sheath reaches the top of the anode and produces a hot and dense plasma column there, during the radial phase. At this stage, the working gas species are ionized because of high temperature and produce ions, atoms and molecules of working gas. Then sausage instability  $(m = 0)$  locally enhances the induced electric field, which is coupled with the magnetic field. Therefore, the focused plasma column is collapsed and produces high energetic working gas ions which are accelerated toward the top of the chamber and relativistic electrons which move rapidly toward the anode. The material of anode tip can be ablated by impacting the energetic electrons to it. High energetic working gas ions and anode tip ablated materials can be deposited on various substrates [\[22\]](#page--1-0).

In this experiment for deposition of a-C:H thin films on stainless steel — AISI 304 substrates, the traditional brass anode is replaced by an oxygen-free high thermal conductivity (OFHC)copper made hollow anode for reducing the impurity atoms in deposited thin films. In addition, electrical conductivity of OFHC is higher than that of brass and this reduces the current loss during the axial acceleration phase.

Highly polished  $5 \times 5 \times 1$  mm<sup>3</sup> stainless steel 304 substrates were cleaned ultrasonically using alcohol and acetone for 10 min respectively. The substrates were mounted at distances of 120 mm from the anode tip using a substrate holder. A removable shutter was used between the substrate holder and the anode in order to avoid deposition during the initial shots for reducing impurities of thin films.

In order to investigate the influence of different hydrogen contents in the a-C:H thin film structure, two sets of experiments were done using two types of gas as working gas. The first working gas was pure analytical grade acetylene, and the second working gas was the mixture of acetylene/hydrogen in 9/1 ratio. In both cases, the samples were deposited at the same distance from the anode tip but at different angular positions (0°, 30° and 60°) with respect to the anode axis. In the first set, the gas pressure was kept at  $5.2 \times 10^{-1}$  mbar, while in the second set; the pressure was kept at  $4.6 \times 10^{-1}$  mbar, for reaching strong focus. Moreover, the charging voltage was kept at 12 kV for both sets. The strong focusing action was indicated by steep current dip in the current coil signal recorded by a four channel TDS 2014 B (100 MHz) Tektronix digital oscilloscope. The chamber was vacuumed by a rotary pump up to  $10^{-3}$  mbar and then it was filled with working gases. The shutter was removed after some strong focusing actions.

The structure of the a-C:H films was investigated using a Philips PW 1840 X-ray diffractometer (XRD) with Cu K $\alpha$  ( $\lambda = 1.5406$ ) radiation.



Fig. 2. Hydrogen contents of samples which are deposited in acetylene and acetylene/ hydrogen discharges, as function of angular position with respect to anode axis.

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