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AMORPHOUS AND DIAMOND-LIKE CARBON COATINGS FOR SEY REDUCTION OF DIELECTRIC MATERIALS FOR ACCELERATING STRUCTURE APPLICATIONS

Alexej Grudiev[1], Holger Neupert[1], Wil Vollenberg[1],
Chunguang Jing[2], Shashi Poddar[2], Ben Freemire[2]

[1]CERN, Geneva, Switzerland
[2]Euclid Techlabs LLC, Bolingbrook, Illinois, USA

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Abstract

Multipactor discharges in dielectric accelerating structures are a major limitation on the performance of this otherwise very promising technology for future high energy physics machines and other applications. Multipactor occurs when the Secondary Emission Yield (SEY) of the dielectric material used in accelerating structures is significantly higher than 1. In this work, SEY coefficient versus energy of primary electrons is measured for different dielectric materials used for accelerating structure applications: MgTiO3 (D16) and MgTiO-based conductive ceramic. Furthermore, effect of SEY reduction by means of amorphous Carbon (a-C) and Diamond-Like Carbon (DLC) coatings is investigated experimentally. The results are presented and discussed.

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Amorphous and Diamond-Like Carbon coatings for SEY reduction of dielectric materials for accelerating structure applications

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Abstract

Multipactor discharges in dielectric accelerating structures are a major limitation on the performance of this otherwise very promising technology for future high energy physics machines and other applications. Multipactor occurs when the Secondary Emission Yield (SEY) of the dielectric material used in accelerating structures is significantly higher than 1. In this work, SEY coefficient versus energy of primary electrons is measured for different dielectric materials used for accelerating structure applications: MgTiO3 (D16) and MgTiO-based conductive ceramic. Furthermore, effect of SEY reduction by means of amorphous Carbon (a-C) and Diamond-Like Carbon (DLC) coatings is investigated experimentally. The results are presented and discussed.

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1. Introduction

Significant progress has been made in the development of high gradient dielectric accelerating (DLA) structures: GV/m gradients at THz and 100MV/m at 10 GHz were achieved in dielectric wakefield accelerating structures [1-4]. As a consequence, dielectric loaded accelerating structures are emerging as a possible, important future alternative to all metal based accelerating structures, facilitating the possibility of very high accelerating gradients and more compact and cost-effective accelerators. One principal effect limiting further advances in this technology is the problem of multipactor, particularly for externally powered DLA structures [4, 5]. Severe multipactor may absorb considerable fraction of the nominal rf input power, which would prevent the structure from producing sufficient accelerating field. In addition, severe multipactor might create many other problems, including degrading the vacuum, detuning the structure, and even damaging the surface of dielectric material. Different TiN coating techniques including Physical Vapor Deposition (PVD) and Atomic Layer Deposition (ALD) have been applied to reduce the multipactor. Up to 20% reduction of the multipactor loading has been demonstrated in previous experiments [4, 6]. However, there is still around 10% rf power lost inside the DLA structure, along with the accompanying fluorescent light emission which occurs at a much higher level compared to the multipactor effect in metal structures. Using geometric factors, particularly the introduction of surface grooves to reduce the multipactor in DLA structures, has also been suggested as well [7], but it significantly increase the complexity of the structure fabrication. Large solenoidal magnetic fields have been proved to effectively suppress multipactor [8], but the magnets are too bulky to be applied for a compact linac system.

Inspired by the work presented in Ref. [9], the DLC (diamond-like carbon) coating seems to be the answer we are looking for. In their work, by applying a DLC that lowers the SEY (secondary electron yield) without reducing the Q-value, they demonstrated 10 MV/m of accelerating fields (limited by the rf power source) in 6-μs-long pulses with multipactor completely suppressed.

The DLC has numbers of variation depending on ratio of diamond and graphite content. It has been learned that a-C (amorphous carbon) has the best performance in terms of multipactor suppression (lower SEY). Higher diamond content and hydrogen terminated surface are subject to higher SEY. Each category of DLC types uses different coating techniques.

CERN developed over the last 10 years a-C coatings with a low SEY [10]. a-C coatings were already successfully applied on 500 meters of beam-pipe for the SPS during LS2 [11]. Vacuum chambers are coated by hollow cathode sputtering [12] as shown in Fig. 1. A hollow graphite cathode is introduced in the vacuum chamber and translated in continuous mode back and forth. An Argon plasma at $1.12*10^{-1}$ mbar is created in the hollow cathode and Carbon atoms are sputtered away from the cathode and deposited uniformly on the inner wall of the vacuum chamber.

In this study, we try for the first time to conduct a comprehensive examination of DLC (including a-C) coating from different vendors on *Magnesium Titanate*-based ceramics and evaluate its performance for the future dielectric accelerator development.

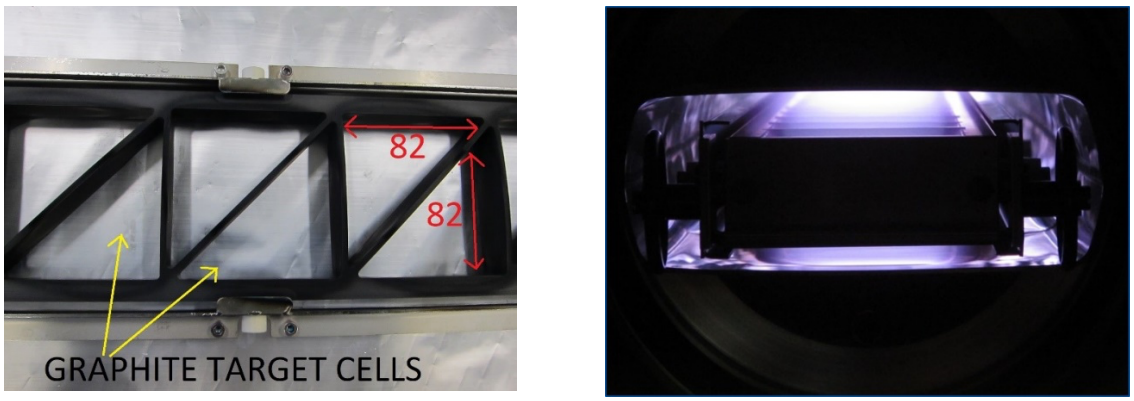


FIG. 1: Hollow cathode (left) and hollow cathode plasma (right)

2. Sample Preparation

There were a total of 11 samples that were supplied by Euclid Techlabs. Three $MgTiO_3$ samples were shipped to CERN for a-C coating. Six coupons were coated with DLC from two vendors: Acree Technologies Inc. based in USA and Nanotec Co. in Japan. Two bare coupons work as the control. Major information is summarized in Table 1.

#	Other name	material	Coating vendors	Note
1	CERN # 1	MgTiO3	a-C, CERN	
2	CERN # 2	MgTiO3	a-C, CERN	
3	CERN # 3	MgTiO3	a-C, CERN	
4		MgTi Oxide based Conductive Ceramic	NONE	
5	DISK-2	MgTi Oxide based Conductive Ceramic	US vendor	
6	DISK-7	MgTi Oxide based Conductive Ceramic	Japan vendor	
7	D16	MgTiO3	NONE	
8	D16-1	MgTiO3	US vendor	
9	D16-2	MgTiO3	Japan vendor	

2.1 a-C coating at CERN

$MgTiO_3$ discs were positioned on the inner wall of a spare vacuum chamber. Sample is at 15 mm from hollow cathode. After a night of pump-down followed by 12 hours of bake-out at 120 °C the coating process was started. In an Argon plasma at 0.12 mbar and a power of 120 W/meter the deposition rate achieved was 20 nm/hour. In two separate coating runs the hollow cathode sputtering (HCS) was used to make a-C coatings of 210 and 400 nm on the $MgTiO_3$ discs, respectively

A third $MgTiO_3$ disc was coated by DC planar magnetron sputtering (DCMS). The sample was loaded at 170 mm from graphite target. After a night of pump down followed by 12 hours of bake-out at 120 °C coating process was started. In an Argon plasma at $7*10^{-4}$ mbar and 200 W, an a-C thin film of 210 nm was deposited in 2 hours. The disc before and after coating is shown in Figure 2.

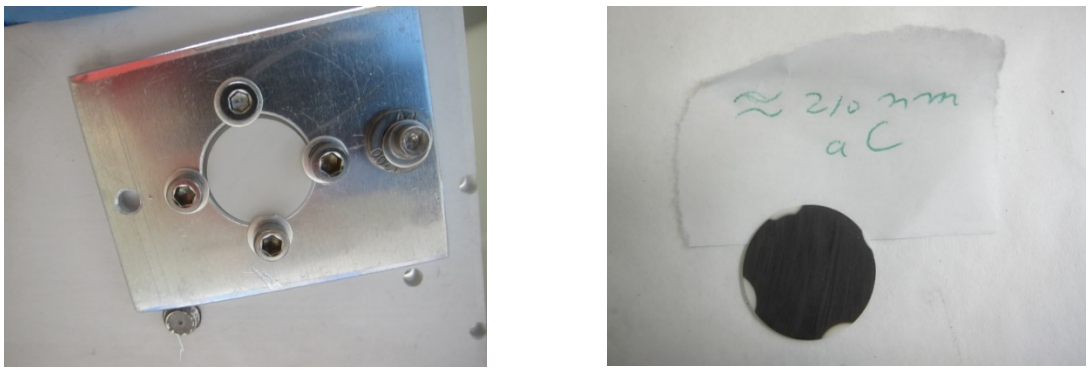


FIG. 2: $MgTiO_3$ disc prior to the coating (left) and after a-C coating (right)

2.2 DLC coating at two other vendors

Neither the US nor Japanese vendor provide details of their coating techniques. Limited information indicates the US vendor, Acree Technologies, uses the sputtering technique; and the Japanese vendor, Nanotec, uses plasma assisted CVD for the DLC coating. Both companies applied ~500nm of the coating thickness in our samples, but no accurate measurement from vendors.

3 SEY Measurements of MgTiO3 (D16) before and after a-C coating

3.1 SEY measurement setup for metals and insulators at CERN

Whereas SEY measurements of metal surfaces is straight forward and is usually done in continuous current mode in the setup schematically shown in Figure 3, measuring SEY of insulators has an additional complication due to surface charging. At CERN this problem is solved by doing the measurements in pulsed mode exposing the insulator surface to 30 µs long electron pulse and performing time-resolved measurements of primary and secondary electron currents. To avoid charging effects the surface was bombarded with low energy electrons (36 eV) between each measurement step. The maximum applied dose to measure one data point was about $2 * 10^{-12}$ C. The total dose to measure one spectrum was therefore about $3 * 10^{-11}$C.

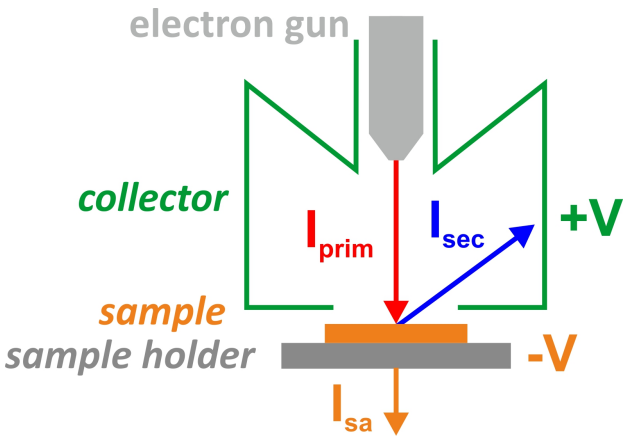


FIG.3: Schematic of the SEY spectra measurement setup.

3.2 Description of the samples

- Sample 1 (CERN # 1): sample MgTiO3 with 210nm amorphous Carbon (a-C) coating by DCMS in building 101
- Sample 2 (CERN # 2): sample MgTiO3 with 210nm a-C coating by HCS in building 867
- Sample 3 (CERN # 3): sample MgTiO3 with 400nm a-C coating by HCS in building 867
- Sample shape: diameter 15mm disks 0.2mm thick
- Sample storage after reception: Kept in plastic container, wrapped in silk paper as received,
- Put under vacuum in SEY instrument:
- Aim of analysis: Measurement of SEY on a-C coated ceramic, pulsed mode with neutralisation
- on raw substrate, DC mode measurement on a-C coating

3.3 Results

3.3.1 SEY spectra

The SEY for different primary electron energies was measured on the backside of a coated sample (Figure 4) on three different spots as received. Measurements on a-C coatings are done in DC mode because charging does not occur on conductive surfaces. Figures 5 and 6 show SEY on two coatings of 210 nm thickness on MgTiO3 done on different systems (Magnetron in bldg.101 and hollow cathode in building 867) respectively. Figures 6 and 7 show SEY on two coatings of 210 nm and 400 nm thickness on MgTiO3 done on hollow cathode system in building 867, respectively. Coating performance of building 867 setup has been checked with coating stainless steel samples. Results are presented in Figures 6 and 7 as well for comparison.

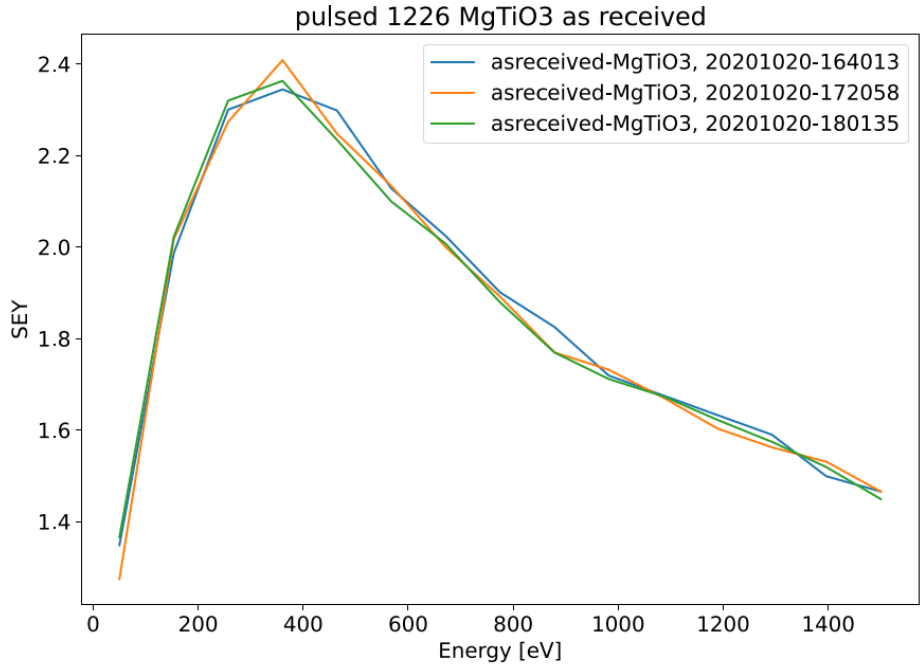


FIG.4: SEY spectra on three different spots on MgTiO3 sample as received.

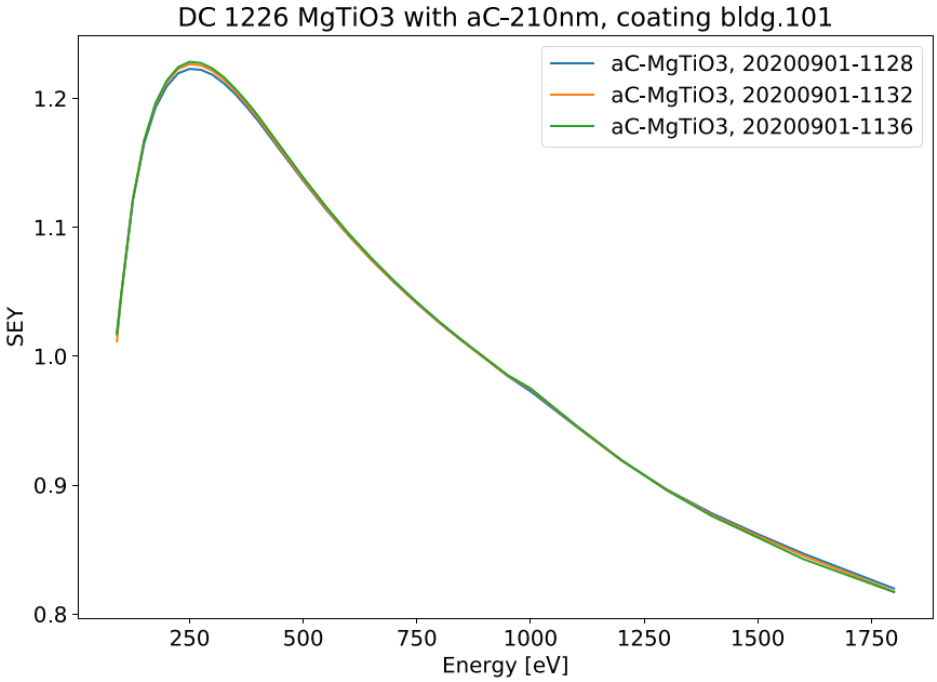


FIG. 5 : SEY spectra on three different spots on MgTiO3 with 210 nm a-C coating done in building 101 (sample 1).

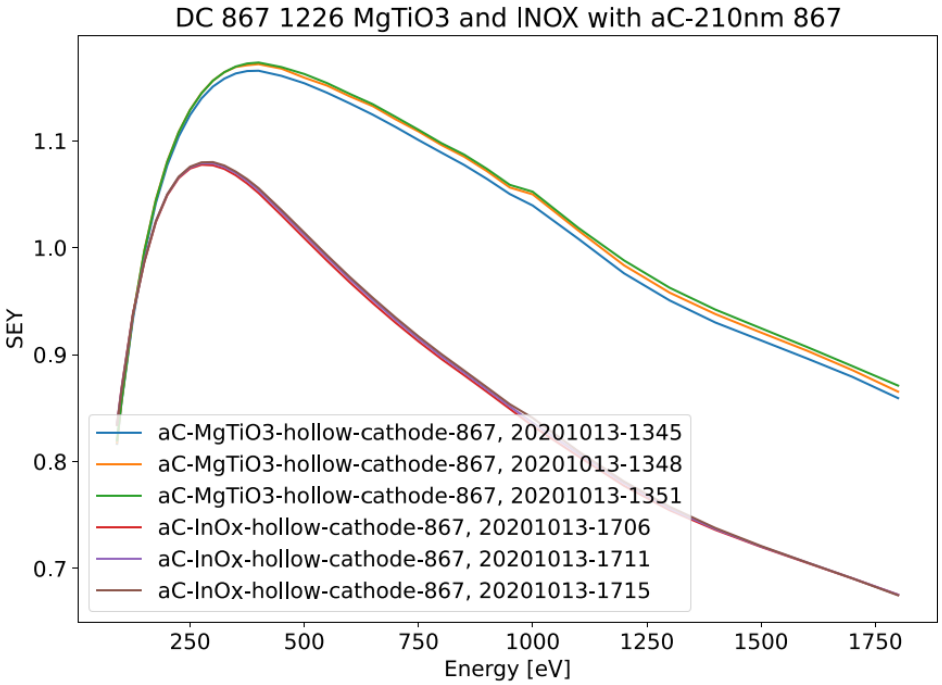


FIG. 6. SEY spectra on three different spots on MgTiO3 with 210 nm a-C coating done in building 867 (sample 2), including witness stainless steel samples.

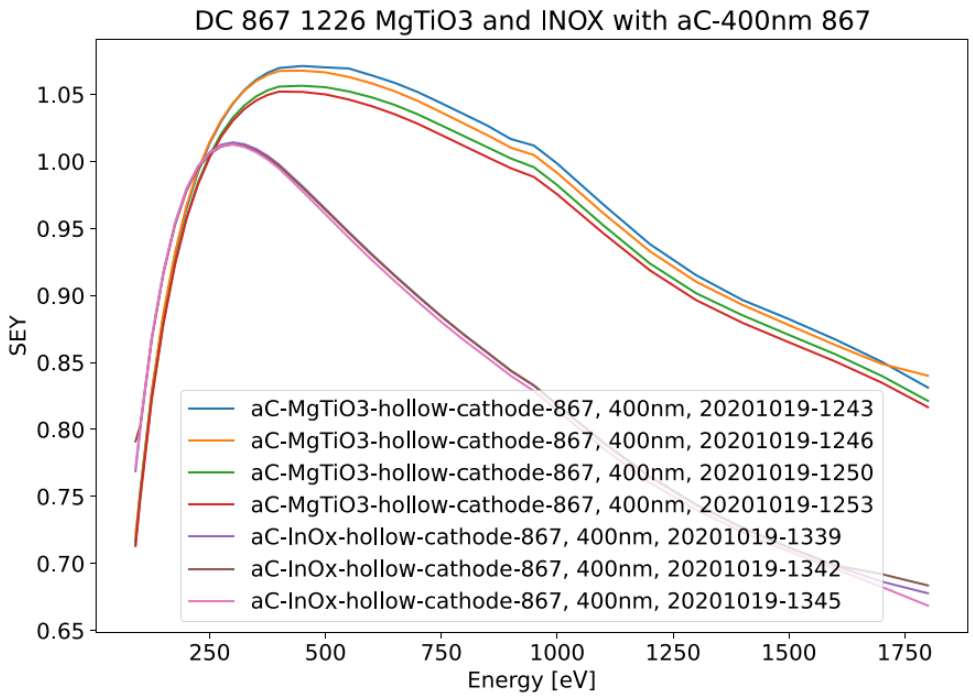


FIG. 7. SEY spectra on three different spots on MgTiO3 with a-C coating done in building 867, a-C thickness 400nm (sample 3), including witness stainless steel samples.

Table 2: Maximum SEY values for each scan and mean value

scan	1st	2nd	3rd	mean
sample as received	2.3	2.4	2.4	2.4
Sample 1: a-C coating bldg 101 210nm	1.2	1.3	1.3	1.3
Sample 2: a-C coating bldg 867 210nm	1.2	1.2	1.2	1.2
Sample 3: a-C coating bldg 867 400nm	1.1	1.1	1.1	1.1

3.3.2 Raman spectra, SEM image, and EDAX spectra.

Raman spectroscopy was also performed on the three samples as received by Euclid at the Center for Nanoscale Materials of Argonne National Laboratory using 785 nm laser source with 1200 lines/mm grating. The characteristic Raman peaks for crystalline diamond-like coatings exhibit a Raman active band at 1580 cm^{-1}, typically known as the G-band (E_{2g}) and a broad peak around 1345-1350 cm^{-1} from the amorphous carbon; and a further peak at 1332 cm^{-1}, close to the D-band, which is typical of crystalline diamond (F_{2g}). The G-band manifests itself mainly due to the trigonal sp^2-carbon present in graphitic layered structures, the D-band is also related to sp^2 bonds in carbon rings and the further peak is identifying diamond in as it is well explained in reference 14 [13] [14]. The presence of significant amount of amorphous carbon will broaden the crystalline characteristic peaks and shift them slightly. For sample 1 and sample 2 which both have amorphous-Carbon coating thickness of 210 nm, but deposited in two different chambers, the Raman signals are identical exhibiting slightly higher D-peak over G-peak, indicating a composition between nano-crystalline graphite and a-C. For thicker a-C coatings in sample 3, the the D component is slightly higher indicating more disorder.

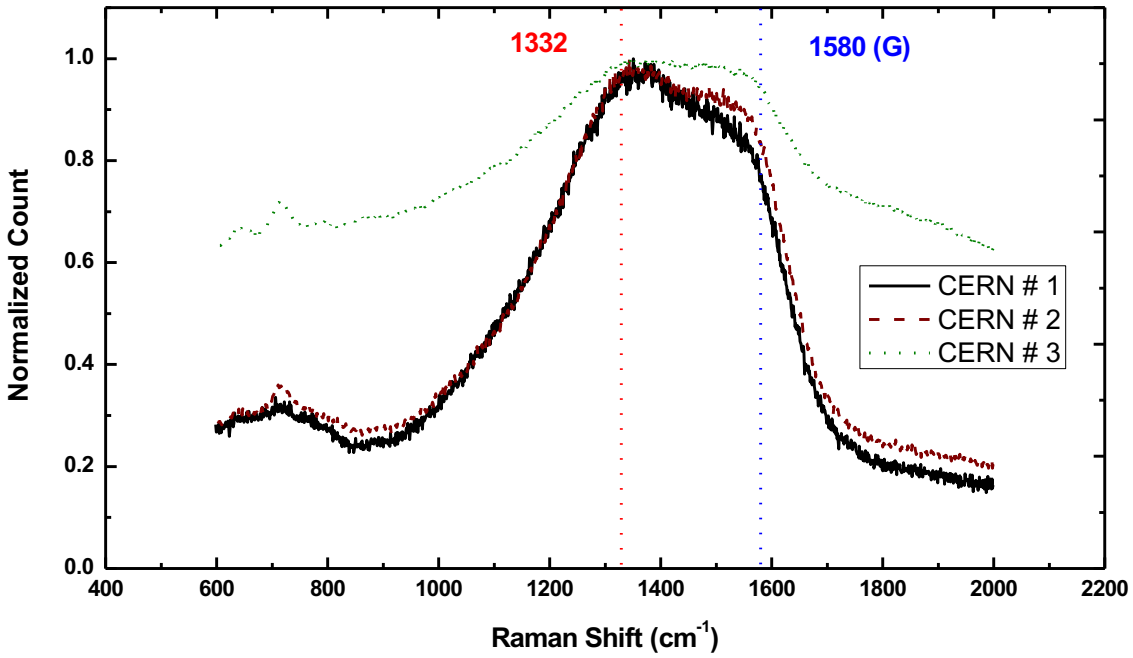


FIG.8 : Raman signal on three $MgTiO_3$ with a-C coating fabricated at CERN.

The three individual $MgTiO_3$ ceramic disc with a-C coatings were also characterized with scanning electron microscope (SEM) integrated with EDAX detector for elemental and grain size analysis. The grain sizes of the a-C coatings have a broad distribution as can be seen from SEM micrographs of Figure 9.

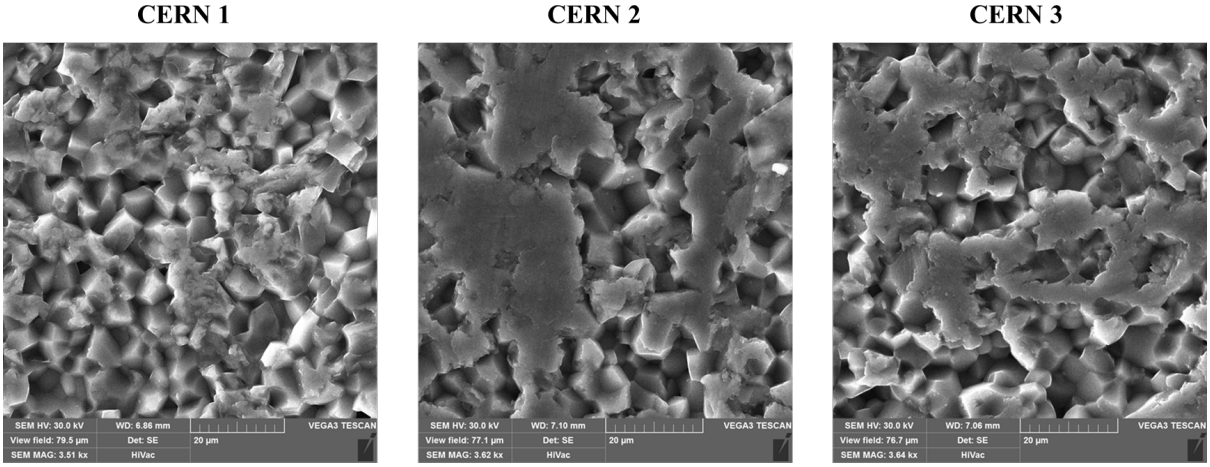


FIG. 9: SEM micrographs of the a-C coating on $MgTiO_3$ ceramic discs.

The elemental mapping for thinner films clearly shows a carbon peak in the EDAX spectra besides the usually expected Mg, Ti and O peaks (Figure 10). As the films get thicker (CERN 3), the background also increases, and the carbon peak is buried in the background. The Al peaks sometimes shows up since the sample holder puck is made of Al.

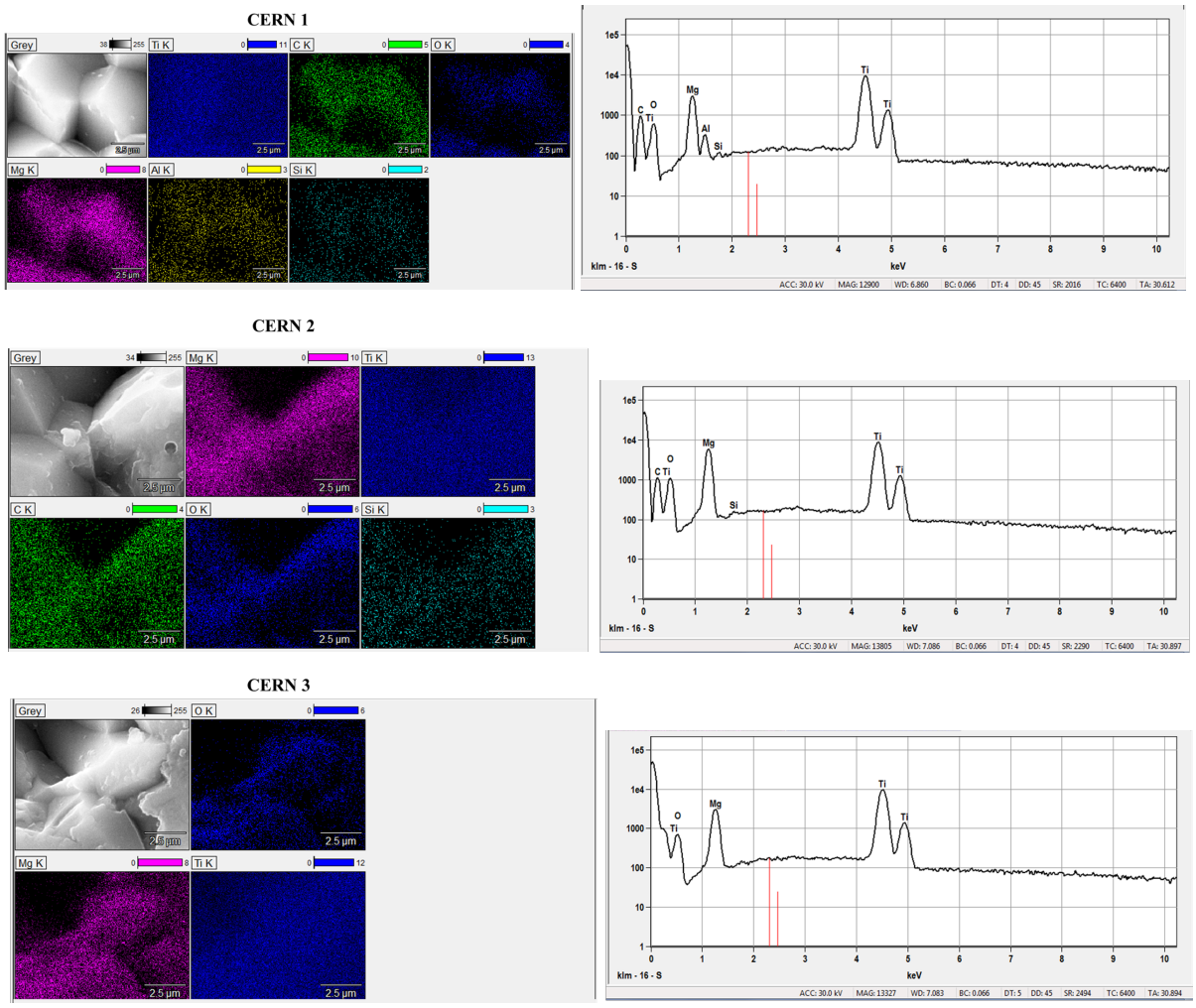


FIG. 10: SEM micrographs of the a-C coating on $MgTiO_3$ ceramic discs.

3.3.3 Resistivity measurements of the a-C coating

The square sheet resistance of the coated $MgTiO_3$ discs were measured with a standard four-point probe of Jandel [15] and calculated with the Valdez method [16, situation no 7]. Results are presented in Table 2.

Table 3: Sheet resistance of the coatings R in Ω per □

samples	R [Ω per □]
Sample 1: a-C coating, DCMS coating lab, bldg. 101, 210nm	3.6E+03
Sample 2: a-C coating, hollow cathode system2, bldg 867, 210nm	3.7E+05
Sample 3: a-C coating, hollow cathode system2, bldg 867, 400nm	7.9E+04

3.4 Summary on the a-C coatings

- The maximum SEY value of MgTiO3 as received is 2.4.
- Coating MgTiO3 with a-C of 210 nm thickness decreases SEY to 1.3 or 1.2 for DCMS (building 101) or HCS (building 867), respectively.
- Coating MgTiO3 with a-C of 400 nm thickness for HCS (building 867) decreases SEY even lower to 1.1.

4. SEY Measurements of MgTiO3 (D16) and MgTiO-based conductive ceramic before and after DLC coating

4.1 Description of the samples

- Sample 4: MgTiO: MgTi Oxide based Conductive Ceramic (Euclid's special ceramic material), uncoated
- Sample 5: Disk-2: The Conductive Ceramic with one side DLC coated; the DLC was done by a US vendor.
- Sample 6: Disk-7: The Conductive Ceramic with one side DLC coated; the DLC was done by a Japan vendor.
- Sample 7: D16 : MgTiO3, uncoated.
- Sample 8: D16-1 : D16 with one side DLC coated; the DLC was done by a US vendor.
- Sample 9: D16-2 : D16 with one side DLC coated; the DLC was done by a Japan vendor.
- Sample shape: diameter 15mm disks 0.2mm thick, partially broken.
- Sample storage after reception: Kept in plastic container, no protection directly touching the plastic. Put under vacuum in SEY instrument.
- Aim of analysis: Measurement of SEY on DLC coated ceramic, pulsed mode with neutralisation on D16.

4.2 Results

4.2.1 SEY spectra

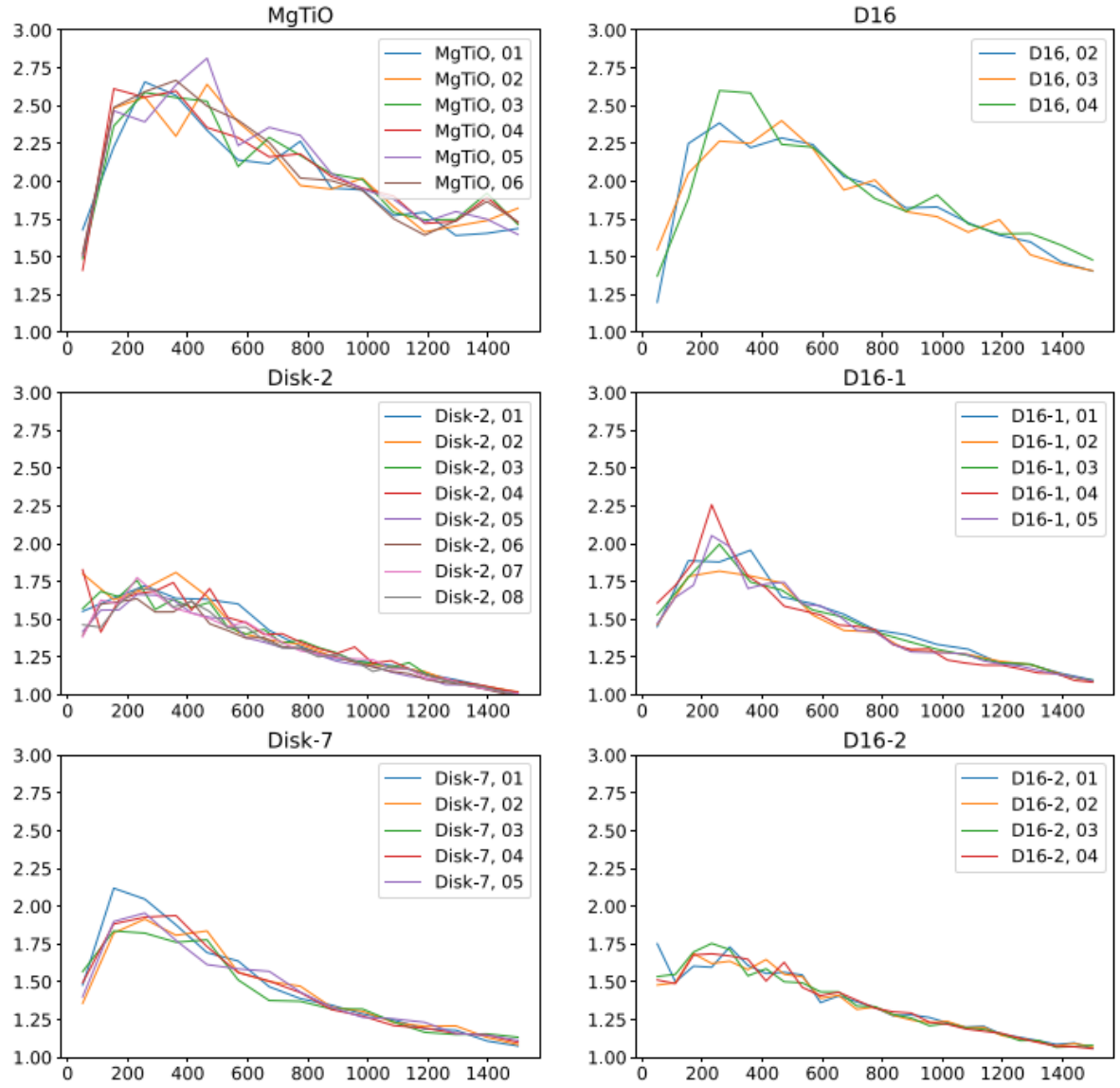


FIG. 11: SEY spectra on at least three different spots on MgTiO-based conductive ceramic samples on the left and MgTiO3 (D16) samples on the right uncoated top and DLC coated, middle (US vendor) and bottom (Japan vendor), respectively.

4.2.2 Raman spectra, SEM image, and EDAX specra.

The D16 batch of samples are insulating ceramics that consist of uncoated disk of $MgTiO_3$ (D16) and two disks coated with diamond-like-carbon (DLC) - one from a US vendor (D16_1) and the other from a Japanese vendor (D16_2). The Raman spectroscopy for the D16-series of samples is plotted in Figure 12. The reference ceramic sample (D16) shows a broad peak over the whole scan range most of which is background. The D16_1 and D16_2 shows an increased peak signal indicating major contribution from amorphous growth of carbon and small contribution from phase mixing of both sp^2/sp^3 carbon species. The peak position and the peak width change a lot on:

1. Presence of amorphous carbon.

2. Growth conditions

DLC films will always have sp^2 trigonal C-atoms. The quality of DLC films is given by the peak intensity ratio: I(1332):I_G. A high ratio (~ 4-5) indicates a high sp^3 diamond-like coating.

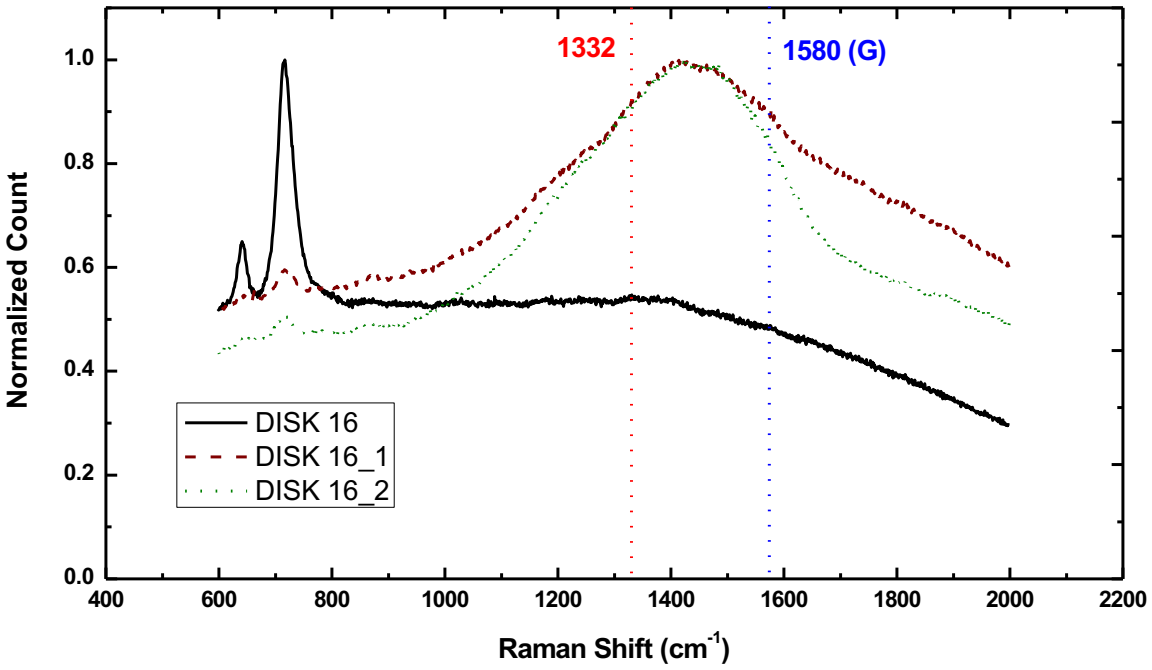


FIG. 12: Raman signal on $MgTiO_3$ insulating ceramic disk with DLC coating.

The SEM micrographs of the D16 series is shown in Figure 13. The uncoated disk charges up slightly during SEM inspection as is expected since these are insulating in nature. The grain topology changes when the ceramic is coated with DLC.

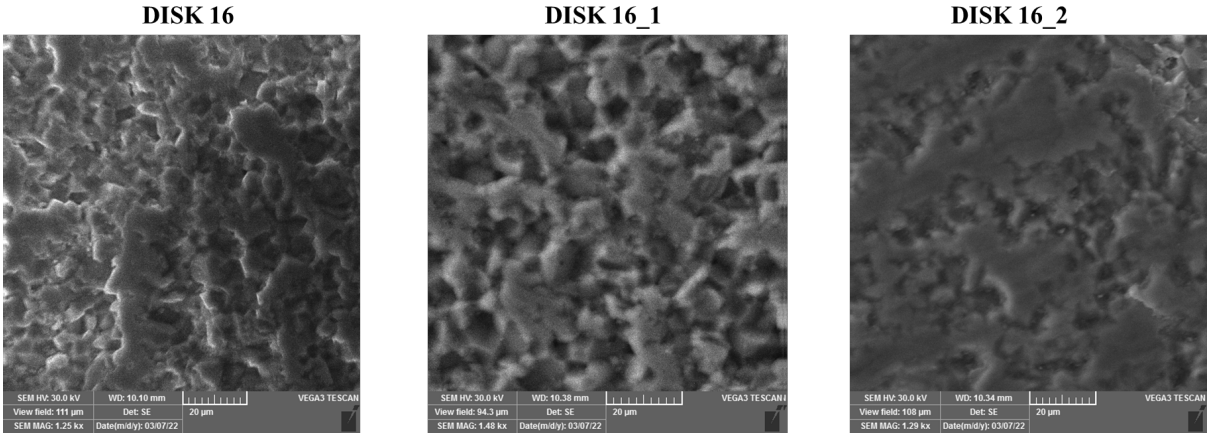
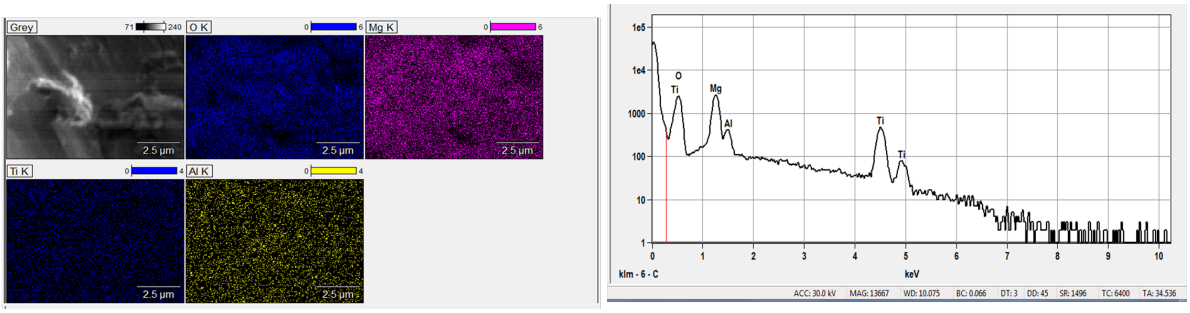


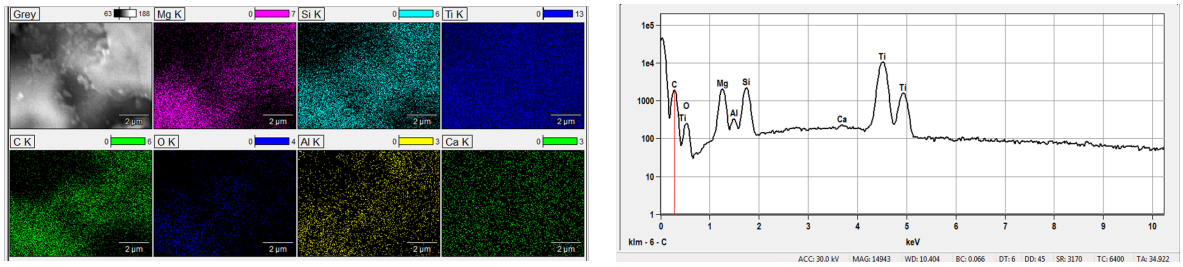
FIG. 13. SEM micrographs of the DLC coating on $MgTiO_3$ ceramic discs.

The EDAX spectral mapping of the elements in the DISK 16 series of sample is shown in Figure 14. The surface of the uncoated insulating disk suffers significant charging for longer dwell times of the spectral mapping (set at 5 mins). The Al peaks are due to the background from the Al sample holder. The carbon count goes up for the coated disks.

DISK 16



DISK 16_1



DISK 16_2

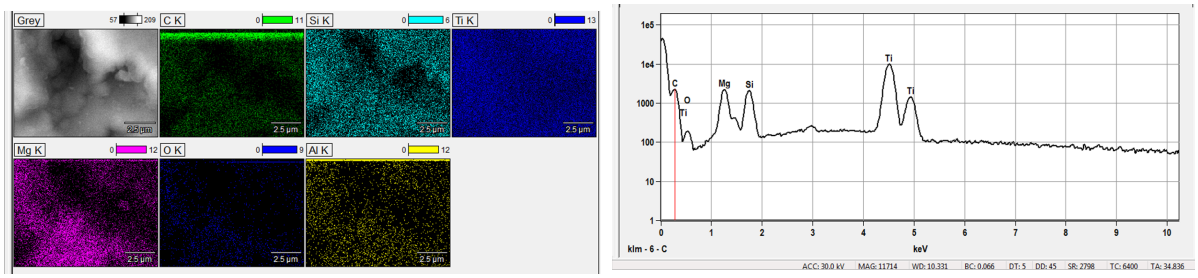


FIG. 14. SEM micrographs and EDAX maps of the DLC coating on $MgTiO_3$ insulating ceramics.

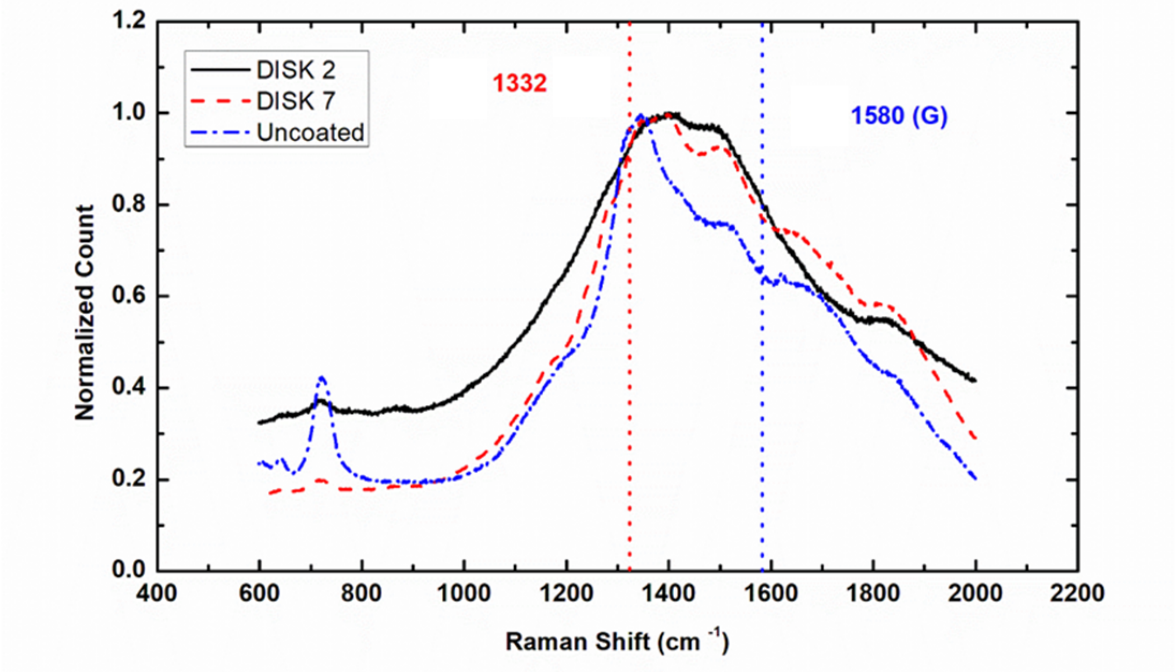


FIG. 15. Raman signal on MgO-based conductive ceramic disk with DLC coating.

Magnesium oxide-based conductive ceramic coupons were also coated with DLC from both the US and Japanese vendors labelled as DISK 2 and DISK 7 in this report, respectively. Raman spectroscopy revealed presence of mixed phases of sp^2 and sp^3-carbon with significant contribution from amorphous carbon as indicated by broad and shifted peaks (Figure 15). The peak around 1332 cm^{-1} for the uncoated disk is not symmetric as such carbon is not contributing to that peak and can be attributed to some artifact from the uncoated disk.

The surface topography as revealed by SEM micrographs for DISK 2 and DISK 7 look very similar with the EDAX identifying Mg, Ti, Si, C and low count O peaks as expected (see Figure 16). The uncoated conducting ceramic disk also has carbon peaks identified which may come from cross contaminations during sample handling.

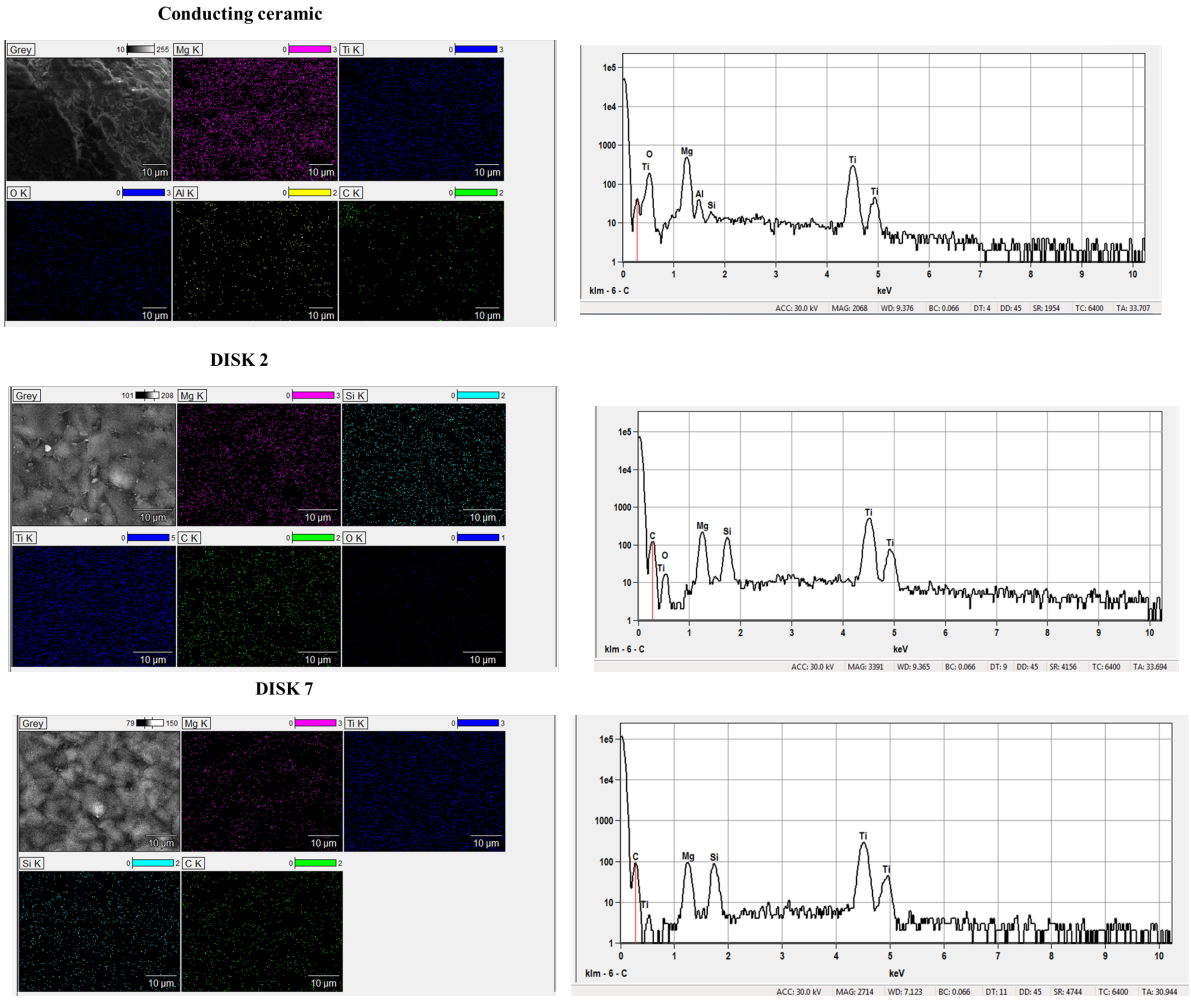


FIG. 16. SEM micrographs and EDAX maps of the DLC coating on MgO-based conducting ceramics.

4.2.3 Resistivity measurements of DLC coatings

Surface resistance of all DLC coatings is above 1MΩ per □ and could not be measured with the using a Cylindrical four-point probe head from Jandel.

4.2.4 Extra information: Surface roughness

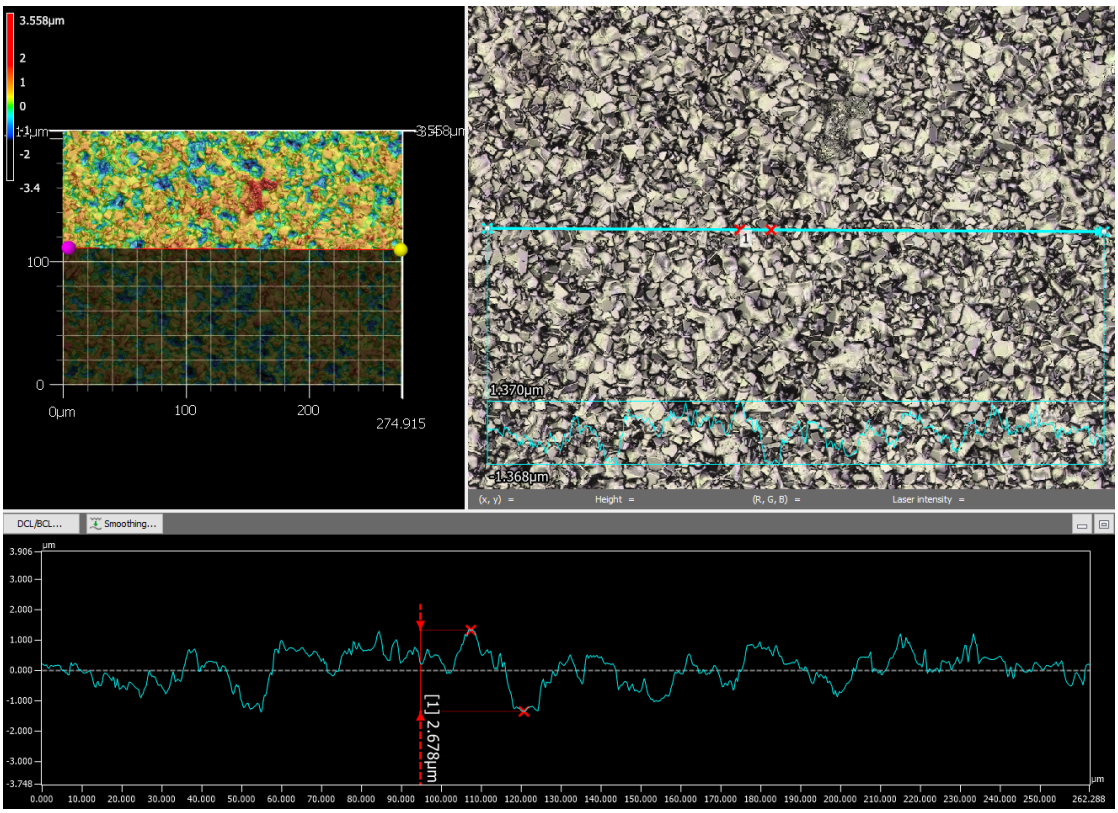


FIG. 17: 50x Magnification disk2. Average Roughness = .435um

4.3 Summary on the DLC coatings

- The maximum SEY value of MgTiO-based conductive ceramic as received is about 2.7
- Coating MgTiO-based conductive ceramic with DLC (Diamond Like Carbon) decreases SEY to 2 or 1.7 depending on the vendor.
- The maximum SEY value of D16 as received is about 2.5.
- Coating D16 with DLC (Diamond Like Carbon) decreases SEY to 2 or 1.7 depending on the vendor.

Acknowledgements

The authors would like to thank Mauro Taborelli for his valuable comments.

References

1. C. Jing, et al., Phys. Rev. Lett. 98, 144801 (2007)
2. M. C. Thompson, etc. Phy. Rev. Lett. 100, 214801 (2008).
3. M.E. Conde, et al, Proceedings of the 2008 European Particle Accelerator Conference, Genoa, Italy, June 2008, pp.2835-2837.
4. C. Jing, et al., IEEE Trans. Plasma Sci. 38, 6, June. 2010, pp1354- 1360.
5. J. G. Power, et al., Phys. Rev. Lett. 92, 164801 (2004).
6. C. Jing, et al., Proc. Advanced Accelerator Concepts,2008, Santa Cruz, CA, pp. 427-432.
7. Awarded DoE SBIR 2010 Phase I project under Contract #DE-SC0004323.
8. C. Jing et al., Appl. Phy. Lett. 108 (19), 101063 (2016).
9. Shingo Mori, Mitsuhiro Yoshida, and Daisuke Satoh, *Phys. Rev. Accel. Beams* 24, 022001, 2021.

10. M. Taborelli, P. Chiggiato, P. Costa Pinto, P. Cruikshank; “Nine years of carbon coating development for the SPS upgrade: achievements and heritage”; CERN-ACC-2016-0010
11. W. Vollenberg, P. Chiggiato, P. Costa Pinto, P. Cruikshank, H. Moreno, C. Pasquino, J. Perez Espinos, M. Taborelli; “Amorphous Carbon coating in SPS” IPAC2021; doi: 10.18429/JACoW-IPAC2021-WEPAB338
12. P. Costa Pinto et al; Implementation of carbon thin film coatings in the Super Proton Synchrotron (SPS) for electron cloud mitigation. Proc. IPAC’14, Dresden, Germany, June 2014, pp 2574-2576. Doi:10.18429/JACoW-IPAC2014-WEPRI043.
13. Thomas, E.L.H., et al., *A simple, space constrained NIRIM type reactor for chemical vapour deposition of diamond.* AIP Advances, 2018. **8**(3): p. 035325.
14. Dychalska, A., et al., *Study of CVD diamond layers with amorphous carbon admixture by Raman scattering spectroscopy.* Materials Science-Poland, 2015. **33**: p. 799 – 805
15. https://www.jandel.co.uk/product_categories/four-point-probes/
16. L.B. Valdes; Resistivity measurements on Germanium for transistors; Proc. Of the IRE; 42 (1954), 420 – 427.