Development of an Energy-Sensitive Detector for the Atom Probe Tomography

Christian Bacchi*, Gérald Da Costa, Emmanuel Cadel, Fabien Cuvilly, Jonathan Houard, Charly Vaudolon, Antoine Normand and François Vurpillot

Normandie Université, UNIROUEN, INSA Rouen, CNRS, Groupe de Physique des Matériaux, 76000 Rouen, France

Abstract
A position and energy-sensitive detector has been developed for atom probe tomography (APT) instruments in order to deal with some mass peak overlap issues encountered in APT experiments. Through this new type of detector, quantitative and qualitative improvements could be considered for critical materials with mass peak overlaps, such as nitrogen and silicon in TiSiN systems, or titanium and carbon in cemented carbide materials. This new detector is based on a thin carbon foil positioned on the front panel of a conventional MCP-DLD detector. According to several studies, it has been demonstrated that the impact of ions on thin carbon foils has the effect of generating a number of transmitted and reflected secondary electrons. The number generated mainly depends on both the kinetic energy and the mass of incident particles. Despite the fact that this phenomenon is well known and has been widely discussed for decades, no studies have been performed to date for using it as a means to discriminate particles energy. Therefore, this study introduces the first experiments on a potential new generation of APT detectors that would be able to resolve mass peak overlaps through the energy-sensitivity of thin carbon foils.

Key words: atom probe tomography, carbon foil, mass peak overlaps, position-energy-sensitive detector

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Introduction
Nanotechnology is one of the fastest growing areas of research in materials science. The reason for that originates from the growing need to create new materials that would simply significant technological breakthroughs over the next decades (Miodownik, 2015). Whether it is a question of increasing the efficiency of solar cells, developing carbon storage at a large scale or finding alternatives to rare earths, it becomes essential to understand the internal nanostructures and microstructures of materials for those future technologies. With the aim of correctly characterizing nanostructures, it is sometimes necessary to both determine their shape and their composition at the atomic scale. It turns out that the best analytical tool suited for those requirements is an atom probe tomography (APT) microscope. In contrast to electron microscopes, APT instruments are able to both localize and chemically identify atoms, not only from sample surfaces, but also from internal layers of analyzed materials.

The basic principle of this instrument relies on the field evaporation of atoms from the material surface. By applying a high electric potential $V_{DC}$ (≈1–20 kV) on a material sample shaped as a sharp tip, with a radius of curvature in the range 10–100 nm, it is possible to extract atoms from the sample surface. This phenomenon called “field ion emission” (Tsong, 1990; Vurpillot, 2016) can be controlled by electric (or laser) pulses $V_p$, allowing atoms from the tip surface to be ionized and repelled toward a position-sensitive detector. During their projection, ions reach a kinetic energy $E_K$ that tends toward the value of the potential energy $E_P$ generated on the tip apex.

$$E_K = \frac{mv^2}{2}, \quad (1)$$
$$E_P = ne(V_{DC} + V_p). \quad (2)$$

With $m$ the mass, $v$ the speed, $n$ the charge state of the ions, and $e$ the elementary charge of the electron. The theoretical conservation between those two energies [equations (1) and (2)] allows to determine a mass-to-charge ratio for each detected ion. To determine those mass-to-charge ratios, it is then necessary to perform time-of-flight (TOF) measurements between the start pulse, applied on the tip sample, and ions arrival on the detector. Therefore, this technique called “time-of-flight mass spectrometry” (TOF-MS) can provide information on the elemental nature of each detected ion.

Regarding the working process of the instrument, it might be thought that APT could be the best means for characterizing a materials’ nanostructure with high spatial and mass resolution. However, since its creation more than 30 years ago (Cerezo et al., 1988), the instrument is still not fully recognized as a reliable tool perfectly suited for material analyses. The first cause of this originates from the multiple difficulties in retrieving reliable
3D reconstructions, which sometimes do not really correspond to the reality (Miller, 1987; Vurpillot et al., 2000). The second main cause originates from biases brought by APT detectors.

One of the main biases brought by APT detectors relates to their inability to distinguish some elements from one another. Indeed, it can be noticed that the composition of some analyzed materials may involve the evaporation of elements having almost equal mass-to-charge ratios. That is the case, for instance, for nitrogen and silicon in TiSiN systems (Engberg et al., 2018) and in field-effect transistors (Martin et al., 2018), or titanium and carbon in cemented carbide materials (Thuvander et al., 2011). To overcome this limitation, it is necessary to couple the TOF-MS with a new spectroscopy technique that would be able to resolve those interferences, called mass peak overlaps (MPOs).

It should be noted that a significant part of MPOs concerns elements having both different charge states and different masses (Thuvander et al., 2011; Kirchhofer et al., 2014; La Fontaine et al., 2016; Engberg et al., 2018). Consequently, it can be deduced that a significant number of ions involved in MPOs could be indirectly resolved through the measurement of their kinetic energy [equations (1) and (2)]. This assumption has already been stated a few years ago (Kelly, 2011; Broderick et al., 2013) but has never been practically realized. This can be explained by the difficulty to measure additional physical quantities, while preserving the existing high performances of current APT instruments (Da Costa et al., 2012).

Recent developments of position-energy-sensitive detectors (PESDs) greatly compounds this difficulty (Funsten et al., 2004; Buhr et al., 2010; Ohkubo et al., 2014; Fuji et al., 2015), where it can be stated that existing arrays of energy-sensitive cells introduce a very slow response time, that does not go below hundreds of nanoseconds, which is a long way from the few nanoseconds required in APT experiments. This limitation should have the effect of restricting the detection rate and the multi-hit capability of APT instruments. In addition to this, the limited number of energy-sensitive cells of those PESDs should also have the effect of degrading the high spatial resolution of current APT detectors. When the number of energy-sensitive cells is increased, there is also the risk of increasing the complexity of the detection system through an extension of the parallel processing.

Other devices, that are to date not known or even used as PESDs, could both overcome the lack of energy-sensitivity in APT instruments and resolve those last limitations from existing PESDs. An example here is the secondary electron emission (SEE) induced by ion impacts on thin foils. It has been known for many decades that the secondary electron yield (SEY) generated from positive ion bombardment on thin foils is a function of both mass and kinetic energy of ion projectiles (Jackson, 1927; Oliphant, 1930; Hill et al., 1939). Until now, the main practical purpose of thin foils was their use as electron multipliers for getting the start and stop signals for TOF spectrometers by being coupled with microchannel plates (Gloeckler & Hsieh, 1979; Saro et al., 1996; Shapiro et al., 2000; Montagnoli et al., 2005), with no consideration for previous studies on the mass and energy sensitivity of thin foils. Since most of those experiments were conducted with ion beams rather than through atom-by-atom analyses, it was not clear whether this phenomenon could be useful for APT experiments. Moreover, very few publications have reported measurements of the SEY induced by ions having the kinetic energy range encountered in APT experiments (around 5–40 keV). Therefore, the following study is both aimed at conducting experiments that could confirm the mass and energy-sensitivity of thin foils in the energy range of APT experiments, and at developing the first prototype of PESD for APT instruments, that would be able to discretely resolve MPOs.

Materials and Methods

The Ion-Induced Secondary Electron Emission Phenomenon

The secondary electron emission (SEE) induced by ion–matter interactions, also known as ion-induced electron emission, is generally considered as a three-step process (Baragiola, 1993; Rothard et al., 1995; Fig. 1). First, an ion projectile transfers its kinetic energy to electrons from a target material; next, in case of thin foils, a fraction of these secondary electrons (SEs) moves from the internal structure of the target material both toward the entrance and exit surfaces; finally, a fraction of those SE will be ejected from the target, provided that their kinetic energy is sufficient to overcome the surface barrier between the target and the vacuum medium. It has been reported that most of these SE have an initial energy in the range 30–90 eV (Baragiola, 1993; Töglhofer et al., 1993), then reduced to less than 5 eV for SE ejected to the vacuum medium (Palmberg, 1967; Dennison et al., 2006; Tomita et al., 2006; Rymzhnov et al., 2015; Allegrini et al., 2016). Moreover, studies have shown that those ranges of energies from SE do not depend on the nature of ion projectiles, nor on their kinetic energy, but mainly depend on the target material (Hasselkamp, 1992; Baragiola, 1993).

It must be specified that there is some variability in the outcomes from ion–matter interactions. The SEE is part of a set of events that may be observed when ions are projected toward a thin foil. On the one side, the high stopping power, from heavy and low energetic ions on solid matter, may lead to the high probability of ions to lose their whole energy at low depth (Fig. 2). Having the effect of only ionizing target atoms at low depth, and then leading to the decrease of transmitted SE at the rear surface of the foil (Rothard et al., 1998). On the other side, due to their weaker interactions with solids, light and energetic ions may cross through the whole thickness of the foil, by ionizing a very low number of target atoms, and thus releasing very low number of SE (Ziegler & Biersack, 1985). Therefore, for exclusively generating SE, it is necessary to meet two main conditions:

1. The mean free path of SE must be higher than the distance between their start position, where target atoms have been ionized, and exit surfaces of the foil.
2. The stopping power from ion–matter interactions must be sufficient for avoiding the transmission of primary ions through the foil, which may lead nonexclusive SE detections.

As previously mentioned, it has been known for many decades that SEY generated from positive ion bombardment on thin foils is a function of both mass and kinetic energy of the ions (Jackson, 1927; Oliphant, 1930; Hill et al., 1939). More precisely, it has been stated that the average number of secondary electrons emitted per primary ion is proportional to the electronic stopping power (Rothard et al., 1990). Given that the value of the electronic stopping power depends on the ion’s elemental nature, the ion’s kinetic energy, and the composition of target material, it becomes complex and difficult to accurately predict the evolution of the SEY. One must also consider that the transmitted SEY may also vary with the thickness of target materials (Rothard et al.,
1995). It is for those reasons that there is no unique theoretical basis for this phenomenon, but only semi-empirical models adapted to specific conditions (Sternglass, 1957; McCracken, 1975; Rothard et al., 1990, 1995). Therefore, as a first step, it is necessary to investigate the potential energy sensitivity of thin foils under APT experimental conditions.

**Collection of Secondary Electrons from Carbon Foils**

By recalling that SE can be generated both from entrance and exit surfaces of thin foils (Fig. 1), one can note that those SE can be collected at the proximity of those two surfaces. For that reason, different collecting techniques were developed for decades as a function of the different SEE applications (Šaro et al., 1996). Most of those detection techniques use amorphous thin carbon foils (CFs) in combination with a microchannel plate (MCP)

assembly for both getting timing pulses (start and stop time measurements) triggered by the SE, and for determining the original position of ion impacts (Shapira et al., 2000).

Regarding the different studies of SEE from thin foils, there is evidence to suggest that CFs would be the best choice for generating high amounts of SE. Indeed in 1955, Murdock et al. reported that CFs have one of the highest SEY compared with most other materials (Murdock & Miller, 1995). Although there are exceptions such as cesium iodide (CsI) thin foils, which produce much higher SEYs (Chianelli et al., 1988), but there are difficulties to manufacture robust and regular thicknesses of CsI under 100 μm (Weathers & Tsang, 1996; Chen et al., 2015). In fact, Maier-Komor (1993) reported that CFs can be manufactured one order of magnitude thinner than foils made with other elements, with thicknesses below 5 nm. This can explain why most of the current applications using the SEE phenomenon use CFs
(Allegrini et al., 2016). With the aim of correctly selecting the best CF thickness, specifically intended for APT analyses with ion’s energy range around 5–40 keV, one can refer to the synthetic study from Allegrini et al. (2003), who compared CF thicknesses from 0.5 to 10 μg/cm² (2.5 to 50 nm thick). From this study, it appears that CF thicknesses between 2 and 4 μg/cm² (approximately around 10 to 20 nm thick) are the most appropriate to yield the highest SEY in the energy range of APT experiments. This range of thicknesses can also be deduced by considering both the mean distance that primary ions travel before generating SE, and the mean distance that SE travel inside the CF (Bacchi, 2020).

CFs have already been applied in several detection systems requiring ultra-high vacuum conditions in the same order of magnitude as APT instruments (<10⁻⁹ Torr) (Drexler & DuBois, 1996; Kuznetsov et al., 2000). Moreover, reports from different space programs allow calculations that show that CFs have a sufficient lifetime for performing more than one billion APT analyses without exhibiting any major difference in the SEE properties (Allegrini et al., 2016). From data originating from Allegrini et al. (2016), it can be calculated that a CF surface of 40 cm² (∼7 cm of diameter) would lose less than 1 nm in collecting ∼8 × 10¹⁶ ions. For APT analyses comprising 100 million of ions each, this thickness loss would be reached after 800 million analyses.

In the framework of our study, particular interest has been paid to a configuration using the transitory secondary electrons (forward SE), with the aim of keeping a standard APT geometry, having a detection surface orthogonal to the probed sample (Fig. 3).

**Measurement of the SEY Induced by Ion Beams on Carbon Foils**

Regarding the lack of references on SEY measurements induced by low energy ions, as in APT experiments (around 5–40 keV), it was decided to perform preliminary studies to safely confirm the hypothetical mass and energy sensitivity of CFs. These preliminary studies were aimed at observing the potential differences in SEY between different elements having very close mass-to-charge ratios. To do so, the fine control of specific elements, with specific energies, was performed through ion beam sources, also known as focused ion beams (FIBs; Ward et al., 1985; Wortmann et al., 2013).

SEY measurements were performed with two different beam sources from the GPM lab, a Ga⁺ beam from a ZEISS—NVision 40 (www.zeiss.fr), and an Xe⁺ beam from a Helios G4 PFIB UXe DualBeam (www.thermofisher.com). Given that the respective masses, from most abundant isotopes, of gallium and xenon are different by approximately a factor two (132 Da for xenon and 69 Da for gallium), it is possible to reproduce a case of detection similar to an MPO from APT experiments, through the analysis of Xe⁺ ions having twice the kinetic energy of Ga⁺ ions. The reason for this experiment is questionable, since the difference of ∼3 atomic mass units (amu) between most abundant isotopes of Xe²⁺ and Ga⁺ ions are sufficient for easily distinguishing both elements in classical APT experiments. However, it can be assumed that SEY measurements from those two different elements would not be significantly different from hypothetical SEY induced by two other elements with closer mass-to-charge ratios. This last assumption is supported by the study of Allegrini et al. (2003) where the SEY induced by oxygen, argon, and iron ions on thin CFs are almost similar with less than 2 SEY differences, with equal ion kinetic energies, and within the energy range of 5–40 keV.

**Figure 4** introduces the device specially designed for those measurements. SEY measurements have been performed by collecting forward SE emerging from commercially available CF mounted on TEM grid from EMS (www.emsdiasum.com). The forward SEY has been determined by comparing the ion beam current Ibeam, originating from the FIB source, to the current induced by forward SE ISE measured at the output of the foil. Ibeam and ISE currents have been separately measured through the same Faraday cup and by means of a pico-ammeter (Fig. 4). In a first step, one of the cells of the TEM grid that support the CF has been cleared to get a probe hole for the measurement of Ibeam. In a second step, ISE has been measured through another cell of the TEM grid, this time covered by CF. In order to minimize the sputtering of surface carbon atoms, and preserving SEE properties from the initial CF thickness throughout data acquisitions, it has been decided to apply the following experimental conditions:

1. Input ion currents have been set to less than 130 pA, with an ion beam spot on the foil surface of less than 60 μm.
2. SEY values were determined by averaging tens of current measurements from the Faraday cup, at different positions on the foil, for each ion energy.

![Fig. 3. Illustration of a material analysis performed with an APT instrument equipped with a CF detector: (a) schematic of the CF-MCP-DLD setup with the detection of ²⁸Si²⁺ and ¹⁴N⁺, that conventionally leads to an MPO issue; (b) hypothetical mass spectra that would be obtained after the energy discrimination performed by the CF detector.](image-url)
Those experimental conditions made it possible to get output current variations around 0.5 pA. Meaning that the reduction of CF thickness, through carbon sputtering, and ion implantation, were limited during those experiments.

According to the literature and the previous assessments, it must be recalled that ions may pass through the foil depending on their energy and their mass (Allegrini et al., 2003). It means that the current $I_{\alpha}$ measured on the pico-ammeter is the combination of a fraction $\alpha$ of incoming ions coming from the FIB source and forward SEs induced by ion impacts. Therefore, in order to get a reliable estimation of the SEY, special attention has been taken in the subtraction of the positive current induced by ions transmitted through the foil. In the framework of this setup, currents of transmitted ions have only been statistically determined through the use of SRIM simulation tool (Ziegler & Biersack, 1985; Ziegler et al., 2010). The SEY can be then calculated through the following equation:

$$\text{SEY} = \frac{I_{\alpha} - I_{\text{beam}} \times \alpha}{I_{\text{beam}}},$$

where $\alpha$ is the transmission coefficient of the foil calculated on SRIM.

It is also necessary to specify that, in order to avoid the electrostatic charging of the foil surface, its front end has been discharged through a tungsten micro-manipulator connected to an external ground.

CFs with a thickness of $2 \mu g/cm^2$ (10 nm thick) has been chosen for those experiments.

The results from Figure 5a clearly indicate the energy sensitivity of the CF through the increase of the SEY as a function of ions kinetic energy. Then, in order to estimate the ability of the CF to discriminate close mass-to-charge ratios, such as for Xe$^{2+}$ and Ga$^+$ ions in this study (3 amu difference), one can rely on the theoretical relation between mass-to-charge ratios and ion speeds [equation (3)]. In other words, equal mass-to-charge ratios in APT experiments also represent equal ion speeds. Considering that Xe$^+$ ions have a kinetic energy nearly twice higher than Ga$^+$ ions, it is possible to reproduce a case of MPO by projecting them at the same speed toward the CF. Consequently, the two different evolutions of SEY in Figure 5b, respectively, from Ga$^+$ and Xe$^+$ beams, as a function of ion speed, provides an indirect evidence that different elements with close mass-to-charge ratios may be distinguished through a significant difference between their respective SEY.

Specifying that, in the case of those experiments, it is necessary to reach ion speeds above 155,000 m/s to show no signal loss from all ions (SEY $\geq 1$). Transposing this ion speed threshold to APT experiments should mean that no selective losses would be seen above an effective evaporation potential of 8.8 kV.

Those results only yield a statistical point of view of the phenomenon, so it would be necessary to check if this method can be applied for single particle detection, such as for APT experiments.

**Single Particle Sensitivity of the Carbon Foil Detector**

According to the literature, it has been observed that the angular distribution of forward SE exhibits a cosine-dependence (Hachenberg & Brauer, 1959; Klein, 1965; Schou, 1980; Hasselkamp, 1992), suggesting that the distribution of the emitted SE is isotropic within the target. In that way, it would be highly probable that single ion hits would be transformed into unwanted SE multi-hits spread on the detection surface (Fig. 6).

As a consequence, those potential SE multi-hits have to be spatially focused on a small area in order to get a single position and a single TOF for each single ion impact on the CF. To achieve that it can be recalled that each impact treated with an MCP-DLD detector is surrounded by a dead-zone (DZ), where no other successive impact can be spatially resolved (Jagutzki et al., 2002; Miller & Forbes, 2014; Meisenkothen et al., 2015; Peng et al., 2018; Bacchi et al., 2019). Therefore, SE coming from a single ion impact on the CF must be focused on a small area that does not exceed this specific DZ. According to the study of Bacchi et al. (2019), it is known that no successive impacts can be resolved within the following conditions:

$$\Delta \text{TOF} + DT \geq \frac{2|\Delta X|}{v_X} \geq \Delta \text{TOF} - DT,$$

$$\Delta \text{TOF} + DT \geq \frac{2|\Delta Y|}{v_Y} \geq \Delta \text{TOF} - DT,$$

where $\Delta \text{TOF}$ is the TOF difference between ion pairs; $v_X$ and $v_Y$ are the transversal propagation speeds of electric signals,
respectively, on X and Y delay line; ΔX and ΔY are, respectively, the relative distances between multi-hit impacts on X and Y delay line axes; and DT is the instrument dead-time. Given that ΔTOF is nearly equal to zero for SE coming from a single ion impact on the CF, it can be deduced that the DZ where they must be confined together is reduced to the following conditions:

\[ |ΔX| \leq DT \frac{v_X}{2}, \]

\[ |ΔY| \leq DT \frac{v_Y}{2}. \]

The graphical representation of this model in Figure 7b shows that the intensity of \( F_{\text{CF-MCP}} \) must be higher than 310 V/cm for ensuring the focusing of SE, having a maximum energy of 5 eV, in a same DZ where the maximum inter-impact distance required is 2.14 mm on this setup.

This model relies on three initial parameters:

- Due to mechanical constraints in our setup, the distance \( D \) between the foil and the MCP front-end could not be set under 1 cm. Thus, to maximize the focus of the SE to very small areas, it has been decided to keep this distance to 1 cm.
- By considering the cosine dependence of the SE angular distribution (Hachenberg & Brauer, 1959; Klein, 1965; Schou, 1980; Hasselkamp, 1992), a mean angle of incidence ±60° related to the surface normal of the CF exit surface has been set for covering half of the maximum probabilities of the angular distribution.
- The initial kinetic energy of SE has been set between 1 and 5 eV, in accordance with studies that showed that this order of magnitude does not depend on the kinetic energy or the mass of incoming ions (Tomita et al., 2006; Allegrini et al., 2016).

The graphical representation of this model in Figure 7b shows that the intensity of \( F_{\text{CF-MCP}} \) must be higher than 310 V/cm for ensuring the focusing of SE, having a maximum energy of 5 eV, in a same DZ where the maximum inter-impact distance required is 2.14 mm on this setup.

Theoretically, it could be assumed that applying a very high value of \( F_{\text{CF-MCP}} \) would be favorable for the single sensitivity of the CF detector, but two main limitations of this setup may induce significant losses. First, it should be noted that the saturation regime of the MCP assembly not only implies a limit on the output charge from a single microchannel, but also from the microchannels that are very close to original points of impact (Ladislas Wiza, 1979; Barnstedt, 2016). This means that the very high proximity of SE can trigger the saturation of the MCP around adjacent microchannels. The second main limitation concerns the restricted electron detection efficiency of MCPs. It has been reported that the highest detection efficiency for electron projectiles can only be reached with energies varying between 200 and 400 eV (Goruganthu & Wilson, 1984; Müller et al., 1986). However, since the only references linked to the electron detection efficiency of MCPs are several decades old, it can be assumed that properties of latest versions of MCP assembly would be different than older versions (microchannel bias angle, channel length to diameter ratio, additional material on MCP substrate, etc.). Therefore, one should not exclude maximal electron detection efficiencies with electron energies above 400 eV. Given that the
The initial kinetic energy of SEs outcoming from CFs is limited to a few eVs, so it can be deduced that, for a distance D between the foil and the MCP front-end of 1 cm, \( F_{\text{CF-MCP}} \) should be set above 200 V/cm.

**Experimental Setup for the Carbon Foil Detector**

**CF-MCP-DLD Assembly**

As previously mentioned, with the aim of fully exploiting the benefits of CFs mass and energy-sensitivity, it has been decided to combine a conventional MCP-DLD detector with a carbon foil mounting placed on its front-end (Fig. 8). CFs of 2 \( \mu \)g/cm\(^2\) (\( \sim \)10 nm) thick were provided by Arizona Carbon Foil Company (ACF) and the mounting was designed at the GPM lab. In addition to providing information from the CF detector, a direct comparison with the conventional MCP-DLD detector has been performed by splitting the detection area into two distinct parts (Figs. 8, 9). The MCP assembly is composed of two MCP stacks with an effective diameter of 77 mm, and an open area ratio (OAR) of \( \sim 60\% \) (nearly equal to the detection efficiency of the MCP assembly). A bias voltage of \( \sim 1,800 \) V has been used.

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![Figure 7](image1.png)

**Figure 7.** Electrostatic model developed for focusing SE into a restricted DZ on the MCP front-end: (a) Two-dimensional representation of the electrostatic model used for computing the required electric field for focusing SEs on a restricted area on the MCP; (b) graphical representation of the electrostatic model used for computing the required electric field for focusing SE on the restricted DZ. The red area defines the required electric fields for ensuring the focusing of SE in a same DZ.

![Figure 8](image2.png)

**Figure 8.** Design of the carbon foil (CF) detector especially designed for this study. (a) Top view of the CF assembly introducing a detection surface half covered by a CF mounted on a 70 lpi Nickel mesh, with an open area ratio of 90%. (b) Photograph of the CF-MCP-DLD assembly mounted on a 200-CF flange.

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![Figure 9](image3.png)

**Figure 9.** Setup of the dedicated CF detector workbench.
set to the MCP assembly for saturating the generation of secondary electrons at each microchannel output. Consequently, every SE collected at the entrance of the MCP assembly will generate approximately the same output pulse, which will have the effect of reducing selective losses caused by the energy spread of SE outcoming from the CF.

The DLD used in this setup has been specifically designed at the GPM lab (Bacchi, 2020).

Acquisition System of the Carbon Foil Detector
In order to estimate the SEY induced by ion impacts on the CF, it was decided to use an APT detection system that was able to record amplitudes of output signals, known as the advanced delay line detector (Da Costa et al., 2005, 2012), or aDLD detection system. The aDLD detection system is based on fast digitizers, sampling MCP output signals at 4 Gs/s and DLD output signals at 1 Gs/s.

Fig. 10. (a) Pulse height distributions (PHDs) extracted from the LaWATAP instrument for the analysis of a pure aluminum sample. (b) Box plots generated from data in (a). Central red marks from box plots indicate median values, and the bottom and top edges of boxes indicate the 25th and 75th percentiles, respectively. Whiskers from box plots go from the end of the interquartile range to the furthest observation within the whisker length (1.5 times interquartile range). PHDs originating from double and triple ion hits on the MCP assembly have been selected for events introducing TOF differences that do not exceed an arbitrary value of 100 ps. Considering that ions kinetic energy follows the evolution of the evaporation potential [equations (1) and (2)], MCP amplitudes have been normalized by each corresponding DC potential applied on the analyzed material from each detection event.

Fig. 11. 2D distribution of MCP amplitudes on the detector surface. The upper part of the detection surface is the area without foil and the lower part is the area covered by a CF. This figure clearly reveals the amplification of MCP signals through the CF detector that is induced by the secondary electron emission for each ion impact on the foil.
Given that all SE collected by the MCP assembly are converted into approximately equal output pulses, it can be assumed that recorded amplitude levels will be approximately proportional to the SEY. This assumption can be confirmed through the study of pulse height distributions (PHDs) originating from spatially resolved and simultaneous ion multi-hits from a conventional MCP-DLD detector, where the number of collected particles can be compared with resulting MCP amplitudes. Figure 10 clearly confirms this last assumption, since the increase in the number of simultaneous ion impacts has the effect of increasing in line with MCP amplitudes. As a consequence, the study of PHDs from MCP output signals can be used as SEY indicators for the CF detector. Given that the counting of collected ions is constrained by the spatial resolution of the detector, it must be specified that PHDs from Figure 10a do not strictly correspond to MCP amplitude spreads from a discrete number of collected ions. This means that a part of the PHDs, identified as single hits or multi-hits, may introduce wider dispersion caused by spatially unresolved ion hits. Which can explain the increase of MCP amplitude spreads from single to multiple events (Fig. 10b).

**Results and Discussion**

**Analysis of an FeAl Alloy**

With the aim of evaluating the CF detector, a first analysis was performed with an FeAl (B2) alloy, containing 40 at% Al, for...
highlighting the MPO located at 27 Da between the $^{27}$Al$^{+}$ peak and the $^{54}$Fe$^{2+}$ peak (Seol et al., 2013; Marceau et al., 2015). For this analysis, a DC voltage in the range 8.8–9.3 kV was applied to the sample. In addition, with the aim of determining the effect of the CF detector on composition measurements of the analyzed material, it has been decided to vary the potential difference

![MCP pulse height distributions](image)

**Fig. 13.** MCP pulse height distributions respectively from the detection of $^{56}$Fe$^{+}$ ions (a) and ions originating from the MPO between $^{27}$Al$^{+}$ and $^{54}$Fe$^{2+}$ (b) with the CF detector. Original PHDs (solid lines) have been decomposed into individual distributions (dotted lines) with the help of a reference PHD taken from single ion hits.

Table 1. (a) Estimation of the number of detected SE per ion hit through PHD decompositions (Fig. 13). Estimation of relative SEY in (b) represent mean numbers of detected SE per ion hit from (a).

| Number of detected SE | $^{56}$Fe$^{+}$ | $^{56}$Fe$^{2+}$ | $^{27}$Al$^{2+}$ | $^{27}$Al$^{+}$/ $^{54}$Fe$^{2+}$ |
|-----------------------|-----------------|-----------------|-----------------|------------------|
| 1                     | 74.45 %         | 39.51 %         | 37.85 %         | 55.96 %          |
| 2                     | 16.47 %         | 23.09 %         | 22.47 %         | 23.98 %          |
| 3                     | 5.60 %          | 20.28 %         | 18.00 %         | 13.01 %          |
| 4                     | 2.53 %          | 8.58 %          | 11.29 %         | 3.82 %           |
| 5                     | 0.96 %          | 4.29 %          | 4.27 %          | 2.18 %           |
| 6                     | 0.00 %          | 2.81 %          | 3.60 %          | 0.81 %           |
| 7                     | 0.00 %          | 0.72 %          | 1.63 %          | 0.20 %           |
| 8                     | 0.00 %          | 0.37 %          | 0.90 %          | 0.03 %           |
| 9                     | 0.00 %          | 0.25 %          | 0.00 %          | 0.00 %           |
| 10                    | 0.00 %          | 0.10 %          | 0.00 %          | 0.00 %           |

| Relative SEY | $^{56}$Fe$^{+}$ | $^{56}$Fe$^{2+}$ | $^{27}$Al$^{2+}$ | $^{27}$Al$^{+}$/ $^{54}$Fe$^{2+}$ |
|--------------|-----------------|-----------------|-----------------|------------------|
| 1.39         | 2.30            | 2.43            | 1.76            |
ΔV_{CF-MCP} between 100 and 800 V. The choice of this voltage range is justified by the interest of highlighting both the lack of SE focusing, which may bias composition measurements, and the high SE focusing, which may transform unwanted SE multi-hits into single particle detections (Fig. 7b).

In a global aspect, it can be primarily observed in Figure 11 that the CF properly plays its role of signal amplifier. As discussed earlier, it can be observed that the increase of the electric field between the CF and the MCP has the effect of focusing SEs and creating overlapped electric signals for each ion impact. On the lower part of 2D maps (Fig. 11), representing the CF area, it can be observed that MCP amplitudes are nearly twice higher than the upper part, representing the MCP-DLD area (without CF). It has to be specified that the narrow circular band of lower MCP amplitudes, following the edge of the CF area, is caused by the focusing of peripheral SE toward the center of the area through the applied electric field between the CF and the MCP front-end.

Now for the potential MPO at 27 Da, a comparison between the two areas has been performed through their respective MCP PHD (Fig. 12). Since there is a theoretical relation between the kinetic energy \( E_K \) of ions and the electric potential \( V \) applied on the sample [equations (1) and (2)], MCP amplitudes have been normalized by each corresponding DC potential from each detection event. MCP PHDs from the CF area (Figs. 12b, 12d) clearly show that detected ions with multiple charge state introduce higher amplitudes than ions with single charge state. Noting that this charge state dependence can also be observed on PHDs from conventional MCP-DLD detector (Figs. 12a, 12c), but with fewer differences between MCP amplitudes. Moreover, contrary to the meaningful separation between PHDs from the separate well-defined number of collected ions (Fig. 10), Figure 12b may suggest that all PHDs extracted from the CF area are composed of MCP amplitudes originating from different numbers of collected SE. Regarding the potential variability of the SEE phenomenon (see "The ion-induced secondary electron emission phenomenon" section), coupled with the lack of transparency of the MCP assembly, this observation seems to be consistent. This makes the estimation of the SEY very difficult.

However, by referring to the previous assumption on the proportionality of MCP amplitudes with the number of collected particles (Fig. 10), it can be assumed that the number of collected SE per ion impact on the CF can be determined through the decomposition of PHDs into individual distributions, representing discrete values of SE multi-hits (Fig. 13 and Table 1a). This decomposition has been performed through a shape recognition by considering PHD

| \( \Delta V_{CF-MCP} = 100V \) | \( 54\text{Fe}^+ \) | No Foil (%) | Foil (%) | Gain/Loss (%) | \( \text{Fe/Al Fraction} \) (%)(No Foil) | \( \text{Fe/Al Fraction} \) (%)(Foil) | Gain/Loss (%) |
|---|---|---|---|---|---|---|---|
| \( 54\text{Fe}^+ \) | 1.55 | 0.56 | -0.99 |
| \( 56\text{Fe}^{+3}\text{Fe}^+ \) | 24.75 | 6.50 | -18.25 | 63.75 | 62.09 | -1.66 |
| \( 56\text{Fe}^{+3}\text{Fe}^+ \) | 37.45 | 55.03 | 17.58 |
| \( 27\text{Al}^{+54}\text{Fe}^{+2} \) | 34.62 | 35.26 | 0.64 | 36.25 | 37.91 | 1.66 |
| \( 27\text{Al}^+ \) | 1.63 | 2.66 | 1.02 |

| \( \Delta V_{CF-MCP} = 500V \) | \( 54\text{Fe}^+ \) | No Foil (%) | Foil (%) | Gain/Loss (%) | \( \text{Fe/Al Fraction} \) (%)(No Foil) | \( \text{Fe/Al Fraction} \) (%)(Foil) | Gain/Loss (%) |
|---|---|---|---|---|---|---|---|
| \( 54\text{Fe}^+ \) | 1.26 | 0.85 | -0.41 |
| \( 56\text{Fe}^{+3}\text{Fe}^+ \) | 18.77 | 8.98 | -9.78 | 63.29 | 62.47 | -0.82 |
| \( 56\text{Fe}^{+3}\text{Fe}^+ \) | 43.26 | 52.64 | 9.37 |
| \( 27\text{Al}^{+54}\text{Fe}^{+2} \) | 33.05 | 33.40 | 0.35 | 36.71 | 37.53 | 0.82 |
| \( 27\text{Al}^+ \) | 3.66 | 4.14 | 0.48 |

| \( \Delta V_{CF-MCP} = 800V \) | \( 54\text{Fe}^+ \) | No Foil (%) | Foil (%) | Gain/Loss (%) | \( \text{Fe/Al Fraction} \) (%)(No Foil) | \( \text{Fe/Al Fraction} \) (%)(Foil) | Gain/Loss (%) |
|---|---|---|---|---|---|---|---|
| \( 54\text{Fe}^+ \) | 1.00 | 0.93 | -0.06 |
| \( 56\text{Fe}^{+3}\text{Fe}^+ \) | 16.97 | 9.80 | -7.16 | 63.27 | 62.32 | -0.96 |
| \( 56\text{Fe}^{+3}\text{Fe}^+ \) | 45.31 | 51.58 | 6.27 |
| \( 27\text{Al}^{+54}\text{Fe}^{+2} \) | 32.14 | 32.73 | 0.59 | 36.73 | 37.68 | 0.96 |
| \( 27\text{Al}^+ \) | 4.58 | 4.95 | 0.37 |

Results from the different tables have been obtained from the same sample and at different bias voltages between the CF and the MCP front-end.

\( \Delta V_{CF-MCP} \)
from single ion hits as a reference for identifying successive shifted distributions. From those PHD decompositions, it can be calculated that double charge ions generate on average 1.7 times more SE than single charge ions (Table 1b). Which can confirm again the energy-sensitivity of CFs.

In addition to the energy-sensitivity, it is also possible to determine the energy-resolving power (ERP) of the CF detector through the computation of the uncertainty level of MCP amplitudes [equation (8)].

\[ ERP = \frac{A}{\Delta A}. \] (8)

By considering interquartile ranges, from Figure 12d, as amplitude spreads at the full width at half maximum (\(\Delta A\)), and median values from PHDs of double charge ions (\(A\)), the resulting ERP varies between 1.18 and 1.29. According to studies from Kelly (2011) and Bacchi (2020), ERP of ~10 would be sufficient for resolving most of all MPO involving ions with different charge states. At this point, it can be stated that the ERP obtained with this setup is not sufficient to totally resolve the MPO between \(^{27}\text{Al}^+\) and the \(^{54}\text{Fe}^{2+}\) mass peaks. However, despite this lack of statistical significance, it can be stated that nonoverlapped MCP amplitudes between PHDs, respectively, assigned to \(^{27}\text{Al}^+\) and \(^{54}\text{Fe}^{2+}\) ions, do imply statistical significance. This means that some of the double charge ions, having MCP amplitude levels outside the amplitude range of single charge ions, can be resolved. Selecting those nonoverlapped MCP amplitudes would allow the spatial identification of elements that was not distinguishable in conventional APT instruments.

Unfortunately, the homogeneous nature of the analyzed FeAl alloy did not allow the clear visualization of the spatial separation between \(^{27}\text{Al}^+\) and \(^{54}\text{Fe}^{2+}\) ions. Therefore, other materials introducing MPOs in correlation with heterostructures must be analyzed for spatially resolving the energy sensitivity of the CF detector (see “Analysis of a ZnO/Mg\(x\)Zn\(1-x\)O multi-quantum well system” section).

On the quantitative aspect of this study, one can ask whether the composition of the analyzed material has been altered by the ion-to-electron conversion performed by the CF. Since the design of the detector used in this study both includes a conventional

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Table 3. Relative detection efficiencies of Al and Fe ions in the analyzed FeAl alloy, determined through the comparison between the amount of ions detected with the CF part, related to ions detected on the conventional MCP-DLD part (Figs. 8, 9).

| Ions | \(\Delta V_{CF-MCP} = 100\text{V}\) | \(\Delta V_{CF-MCP} = 500\text{V}\) | \(\Delta V_{CF-MCP} = 800\text{V}\) |
|------|--------------------------------|--------------------------------|--------------------------------|
|      | Count (No Foli) | Count (Foli) | Relative Efficiency | Count (No Foli) | Count (Foli) | Relative Efficiency | Count (No Foli) | Count (Foli) | Relative Efficiency |
| \(^{54}\text{Fe}^+\) | 16919 | 3040 | 17.97% | 5947 | 3651 | 61.39% | 3979 | 3929 | 98.74% |
| \(^{56}\text{Fe}^+\text{Fe}^{2+}\) | 269586 | 35185 | 13.05% | 88539 | 38633 | 43.63% | 67673 | 41277 | 60.99% |
| \(^{56}\text{Fe}^{2+}\text{Fe}^{2+}\) | 407905 | 298044 | 73.07% | 204093 | 226377 | 110.92% | 180702 | 217178 | 120.19% |
| \(^{27}\text{Al}^{2+}\text{Fe}^{2+}\) | 377101 | 190968 | 50.64% | 155916 | 143627 | 92.12% | 128198 | 137808 | 107.50% |
| \(^{27}\text{Al}^{2+}\) | 17780 | 14383 | 80.89% | 471760 | 430073 | 91.16% | 18270 | 20858 | 114.17% |
| Total | 1089291 | 541620 | 49.72% | 471760 | 430073 | 91.16% | 398822 | 421050 | 105.57% |

Results from the different tables have been obtained from the same sample and at different bias voltages between the CF and the MCP front-end.

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![Fig. 14.](image-url)  
(a) Scheme of a tip sample extracted from a ZnO/Mg\(x\)Zn\(1-x\)O system; (b) partial mass spectrum around the potential MPO between \(^{64}\text{Zn}^{2+}\) and \(\text{O}^{2+}\) peaks.
MCP-DLD assembly and a CF-MCP-DLD assembly (Figs. 8, 9), it is possible to get an estimation of compositional biases brought by the CF detector, through a compositional reference brought by the conventional MCP-DLD assembly. Through this direct comparison (Table 2), it can be observed that low compositional biases are obtained above the minimum required bias voltages $\Delta V_{\text{CF-MCP}}$ for focusing simultaneous SE hits (>310 V). This improvement in composition measurements from the CF detector seems to be associated with the increase of the detection efficiency of single charge ions related to double charge ions (Table 3). This readjustment appears to be difficult to explain, since this increase of detection efficiencies is coupled with the increase of electron energies to 800 eV, whereas the maximum electron detection efficiency of MCPs theoretically lies between 200 and 400 eV (Goruganthu & Wilson, 1984; Müller et al., 1986). However, assuming that the maximum detection efficiency of the MCP lies in higher electron energies than values from the literature, it is possible that undetected ions, associated with very low SEY, can be now detected with higher SE energies, whereas the higher amount of SE generated from double charge ions may

![Fig. 15. MCP pulse height distributions originating from the analysis of the ZnO/Mg$_{x}$Zn$_{1-x}$O system: (a) and (c) from “no-foil” area, (b) and (d) from CF area (at $\Delta V_{\text{CF-MCP}} =$ 600 V). Box plots from the CF area show that detected ions with multiple charge state introduce higher amplitudes than ions with single charge state. Central red marks from box plots (c and d) indicate median values, and the bottom and top edges of boxes indicate the 25th and 75th percentiles, respectively. Whiskers from box plots go from the end of the interquartile range to the furthest observation within the whisker length (1.5 times interquartile range).](image)
have already high probabilities to trigger MCP signals. This may also lead to get higher probabilities to detect ions with the CF detector related to the conventional MCP-DLD detector.

Analysis of a ZnO/Mg\textsubscript{x}Zn\textsubscript{1-x}O Multi-Quantum Well System

For this next study, a ZnO/Mg\textsubscript{x}Zn\textsubscript{1-x}O quantum well (QW) system was analyzed, for specifically resolving the potential MPO located at 32 Da between 64Zn\textsuperscript{2+} and O\textsubscript{2}\textsuperscript{+} peaks (Dawahre et al., 2011; Park et al., 2013; Amirifar et al., 2015; Fig. 14b). The interest of using the ZnO/Mg\textsubscript{x}Zn\textsubscript{1-x}O system is to get a visual evidence of the energy sensitivity of the CF detector.

In the same manner as the analysis of FeAl alloy, a comparison between the two detection areas was performed through their respective MCP PHD, normalized with applied DC voltages in the range 8–8.4 kV. Figures 15b and 15d show the energy sensitivity of the CF detector through the different MCP PHDs, showing higher MCP amplitudes for double charge ions against single charge ions.

From the sample analyzed in this study, it can also be observed that zinc isotopes, that are not confused with other elements, introduce a higher density at the ZnO QW location, while oxygen ions are homogeneously distributed along the sample (Fig. 16b). In the case where the MPO at 32 Da can be partially resolved into 64Zn\textsuperscript{2+} and 32O\textsuperscript{2+} ions, it is possible to recover those same depth profiles through MCP amplitudes filtering. If highest MCP amplitudes correspond to highest ions charge state, it can be deduced that highest MCP amplitudes extracted from the MPO at 32 Da correspond to 64Zn\textsuperscript{2+} ions. However, the opposite cannot be totally true for 32O\textsuperscript{2+} ions due to the lack of statistical significance at low MCP amplitudes for both elements (Figs. 15b, 15d).

The spatial discrimination between 64Zn\textsuperscript{2+} and 32O\textsuperscript{2+} ions has been performed by considering the statistical significance between nonoverlapped MCP amplitudes from single charge ions and double charge ions (Figs. 15b, 15d). In other words, MCP amplitudes exceeding uncertainties from single charge ions have been attributed to double charge ions. Results from Figure 16a show that contrast in the 3D image brought by MCP amplitudes, from the MPO at 32 Da, is not sufficient for directly observing the spatial discrimination between 64Zn\textsuperscript{2+} and 32O\textsuperscript{2+} ions. However, by computing depth profiles corresponding to low and high MCP amplitudes, it is possible to recover depth profiles from both elements (Fig. 16c), which provides the first evidence of spatial discrimination between elements involved in MPO.

Conclusion

A first proof of concept of position-energy-sensitive detector has been developed for APT instruments in order to deal with some MPO issues encountered in APT experiments. Through this new type of detector, quantitative and qualitative improvements can be considered for critical materials introducing MPOs.

This new detector, based on a thin carbon foil positioned on the front panel of a conventional MCP-DLD detector, can generate a number of transmitted SE that mainly depends on both the kinetic energy and the mass of incident particles. Therefore, this study introduces the first experiments on a potential new generation of APT detectors that would be able to resolve MPOs through the energy-sensitivity of thin carbon foils.

In addition to the mass and energy sensitivity of the carbon foil, it has been demonstrated that this type of detector could also be used as a means for improving the APT detection efficiency. However, it has also been observed that significant losses may occur for massive and low energy ions, caused by their low probability to induce high SEY.

The noted lack of energy-sensitivity of the CF detector requires future in-depth studies to improve its performances. One of those improvements relies on the use of existing 90% OAR MCP assemblies, instead of conventional ~60% OAR. This can be explained by the reduction of uncertainties on SEY measurements by increasing the detectability of SE. Indeed, when SE multi-hits occur, a Bernoulli distribution must be considered. This kind of distribution is always consider the “coin toss” situation for

Fig. 16. (a) 3D reconstruction of the ZnO/Mg\textsubscript{x}Zn\textsubscript{1-x}O sample, introducing the MCP amplitude distribution from the mass peak at 32 Da (64Zn\textsuperscript{2+} and 32O\textsuperscript{2+}). (b) and (c) Introduce a comparison of depth profiles between the conventional MCP-DLD detector and the CF detector. The spatial discrimination between 64Zn\textsuperscript{2+} and 32O\textsuperscript{2+} ions in (c) has been performed through an MCP amplitude threshold of 0.017 (normalized value), which corresponds to the upper amplitude limit of single charge ions (Fig. 14d). The theoretical outburst of 64Zn\textsuperscript{2+} ions at the QW position, that could not be observed on the conventional MCP-DLD detector, can now be observed through the energy sensitivity of the CF detector.
each particle arriving at the MCP front-end, but this time with multiple trials for each event. Assuming that all ions generate two SE each, it can be calculated that an MCP OAR of 60% will lead to 36% of chance to detect two SE, 48% for one SE, and 16% for none, out of two generated SE. Whereas an MCP OAR of 90% theoretically lead to 81% of chance to detect two SE, 18% for one SE, and 1% for none, out of two generated SE. Consequently, it can be deduced that increasing the OAR of the MCP has the effect of reducing the uncertainties on SEY measurements, and thus increasing the energy resolution of the CF detector.

The other main improvement could be found in the fine control of the CF thickness, considering that SEs could be generated at different depths, depending on the mass and the energy ranges of analyzed ions. Since the mean depth, where SEs are generated inside the foil, is a function of mass and kinetic energy of ion projectiles (Ziegler & Biersack, 1985), it becomes crucial to correctly adapt the foil thickness for releasing the maximum of SE outside exit surfaces. Thus, the fine control of the CF thickness will potentially allow lower variability in the number of released SE from the foil, leading to better energy resolution from the CF detector. Other future in-depth studies on the ion-induced electron emission phenomenon would then also be necessary to achieve a fine control of this parameter.

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