Supporting Information

Topography Measurements Using High Mass Resolution Time-of-Flight Secondary Ion Mass Spectrometry: Application to Banknotes

Alice Bejjani^{‡*}, Manale Noun[‡], Serge Della-Negra[†], Raymond Tannous^{††}, Georges Chalhoub^{††}, Mazen Hamdan^{††}, Bilal Nsouli[‡].

‡ Research and Development Department, Lebanese Atomic Energy Commission-CNRS, P.O. Box: 11-8281, Beirut, Lebanon, †Institut de Physique Nucléaire, CNRS-IN2P3, 91406 Orsay, France, ††Cash operations department, Banque du Liban, Masraf Lubnan Street, P.O. Box 11-5544, Beirut, Lebanon.

*abejjani@cnrs.edu.lb

Additional text:

Nine figures and one table are presented to detail some part of the work and to describe some of the choices we made while using our suggested methodology for measuring a topography using TOF-SIMS technique.

Table of Content:

- 1- Figure S-1: Schematic illustration of the used analyzer.
- 2- Table S-1: List of the analyzer different components with the additional information about the TOF-SIMS description.
- 3- Figure S-2: Graphical illustration of the Post-it® sample, its scan and profiler study.
- 4- Figure S-3: Real samples used in this study taken from a trademark printed document (specimen) and circulated banknotes.
- 5- Figure S-4: Orientation of Post-it® sample relative to the primary ion beam and electrons beam azimuth direction.
- 6- Figure S-5: TOF values of Na peaks relative to the surface potential when AnaTGT is done.
- 7- Figure S-6: Variation of the step height between the two layers of the post-it® sample when a scan mode is conducted.
- 8- Figure S-7: TOF-SIMS analysis of a sample created from three post-it® sheets.
- 9- Figure S-8: TOF-SIMS analysis of a braille dot taken from a brand new banknote.
- 10- Figure S-9: topographical measurements of banknotes using an optical profiler.

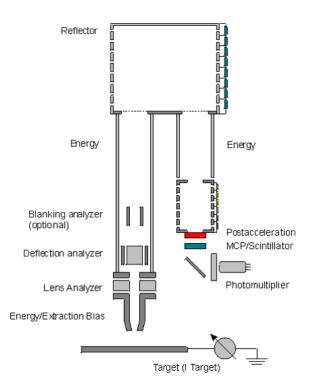


Figure S-1: Schematic illustration of the TOF-SIMS 5 instrument's analyzer, from ION-TOF GmbH, showing its different components

	Surface potential		
Analyzer components	-100 V	0 V	+40 V
Energy voltage (V)	2100	2000	1960
Extractor (V)	2100	2000	1960
Lens (V)	4985	4885	4845
Sensitivity $X (4.5 \rightarrow 6.5\%)$	4.90%	5.10%	5.20%
Sensitivity Y (4.5→6.5%)	5.50%	5.80%	5.90%
Deflection X (-5→+5%)	1%	1.10%	1.10%
Deflection Y (+12 → +24%)	21.80%	22.90%	23.40%
Reflector (V)	-80	+20	+60
Post-Acceleration (V)	9600	9500	9460

Table S-1: The modifications in the analyzer components values when the charge compensation is used and the "virtual" surface potential value of the sample is changing by the user.

TOF-SIMS description:

The ion images which present the localization of a selected peak are obtained using the spectroscopy mode and a digital primary ion raster in "random" mode to minimize sample charging effects. The x axis of the spectra is presented in ns (TOF of secondary ions). Square areas of 500 μ m to 2 mm side sizes were investigated in positive ionization mode with a pixel size of ~ 4 μ m². Correspondingly, the time of analysis has varied between 100 and 300 seconds. The color scales are presented as [0, MC]; where MC corresponds to the maxi-mum counts of the selected ion. The ion beam is incident from the right with its azimuth parallel to the x-axis of the images. The data were acquired and processed with SurfaceLab software using two different versions

(6.6 and 6.7). Using the last version, when the surface potential box is checked the value of the reflector is +20 V relative to the sample potential for positive ions.

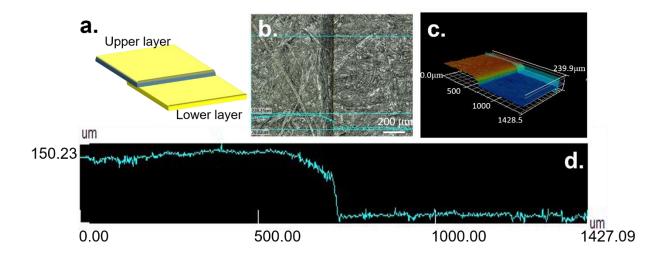


Figure S-2. (a) Graphical representation of the Post-it® model with the position of the two layers. (b) Scan of the interface area of the Post-it® using KEYENCE 3D laser scanning confocal microscope VK-X 200. (c) and (d) Profile study of the analyzed area.

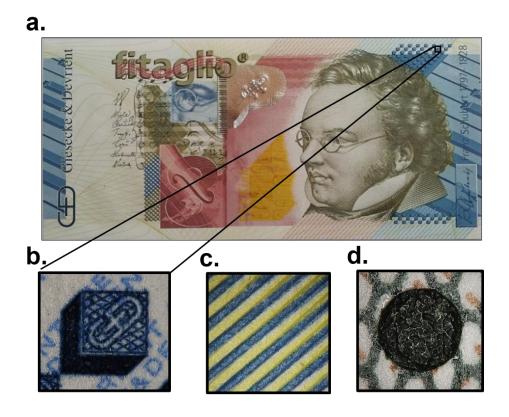


Figure S-3. (a) Photo of the Franz Schubert-Fitaglio® specimen (Courtesy of the G+D Company) with a zoom (b) to the studied area. (c) and (d) Photos of the studied areas taken from the 1000 Lebanese Lira banknote, arabic side.

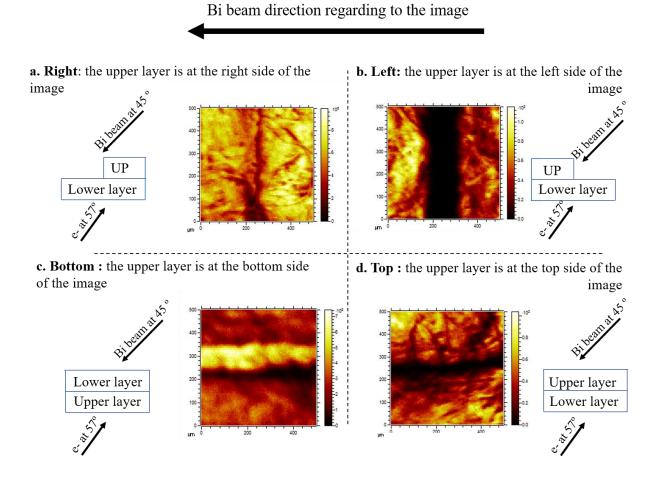


Figure S-4: Total ion images of the interface part of the post-it $^{\text{\tiny M}}$ sample with four different positions relative to the primary ion beam direction (Bi beam): (a) UP at the right side of the image, (b) UP at the left side of the image, (c) UP at the bottom side of the image and (d) UP at the top side of the image.

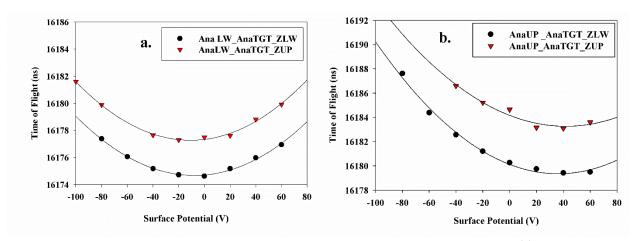


Figure S-5: The variation of the TOF values of the Na peaks maxima relative to the surface potential for (a) the LW taken form the analysis of the interface (Ana TGT) when Z is adjusted on the UP and the LW layers, and for (b) the UP taken from the analysis of the interface (Ana TGT) when Z is adjusted on the UP and the LW layers.

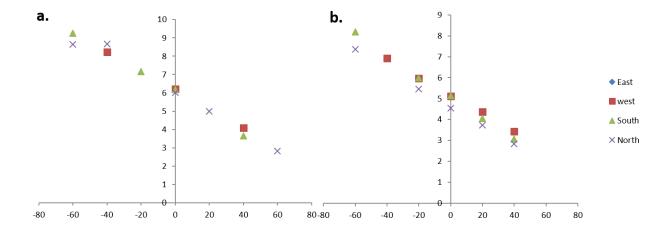


Figure S-6: The variation of the step height between the two layers of the post-itTM sample versus the surface potential value, the four orientation of the sample UP realtive to the primary ion beam azimuth direction and the Z adjustment ((a) ZLW or (b) ZUP) while using the scan mode of a 2*2 mm² area to collect the needed data from the interface region.

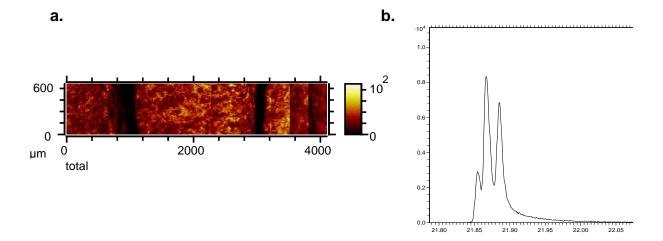


Figure S-7: (a) Total image from the scan of a 0.6*4 mm² of the sample with the three post-it sheets. (b) Na peaks distribution extracted from left to right: upper layer, middle layer, lower layer. Z is adjusted at the middle layer, Surface potential 0V, minimum scan raster size is $300*300 \, \mu m^2$.

3 Post-it $^{\circ}$ sheets have been adhered in a way to form a sample with 3 layers (2 interfaces) and a difference of 180 μ m between the lower and the upper layers. In the presence of the flood gun electrons with a surface potential of 0V, we were able to have 3 peaks of Na from the three different layers. This was achieved only when Z is adjusted on the middle layer. Once Z is adjusted on one of the two outermost layers the other one was not detectable. Thus, we implicitly considered that 180 μ m is above the maximum height which can be measured using this type of protocol.

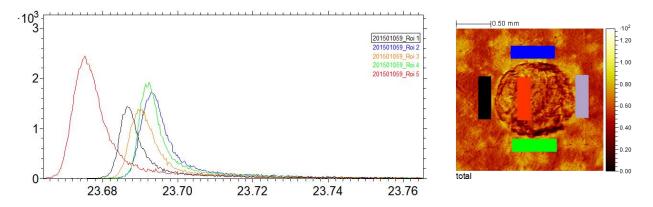


Figure S-8: The Na spectral signature of different R.O.Is taken from the corresponding presented total ion image of the Braille dot and its surrounding, of a brand new 1000 L.L. (scan mode with a minimum raster size of $500*500\mu m^2$).

A Braille dot taken from a brand new 1000 L.L. was analyzed using also the scan mode with a surface potential of -65 V and an extraction gap at to the upper layer. ROI were taken from the analyzed area and their Na peaks were compared. The heights of the surrounding area of the dot, are much closer to each other with no variation higher than 2 ns. These results affirm that the

Braille dots are properly printed using the raised ink technique. However, during circulation (as shown in figure 5.c) this feature is losing its uniqueness when it is compared to its surrounding.

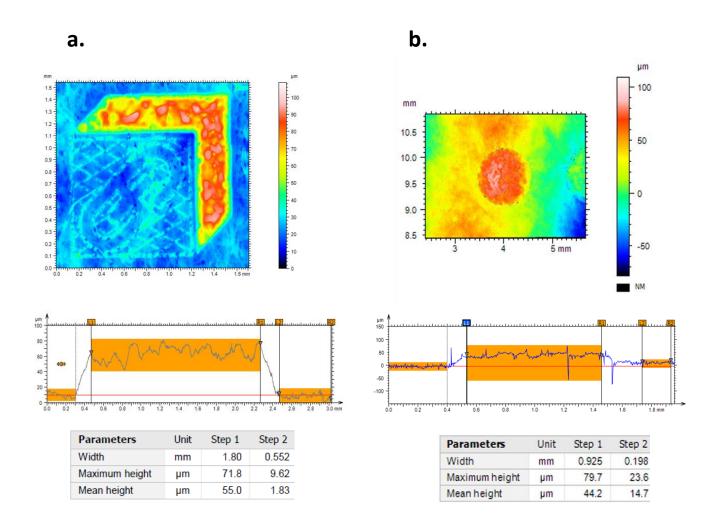


Figure S-9: Profile distribution and step height measurements of (a) the G&D wall from the Franz Schubert-Fitaglio $^{\circ}$ specimen and (b) the braille dot from a circulated 1000 L.L. using a Sensofar S Neox, from Schaefer Technologie GmbH. Recording data is done using the confocal system with different objectives, pixels resolution of 576x768 and data were treated using the MountainsMap $^{\circ}$ Premium 7.4.8495 software