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NanoQI

Multimodal X-ray and Hyperspectral Thin-Film Nano-material Evaluation and Quality Imaging

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Common interface description for machine integration of in-line and in-situ metrology

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Executive Summary

This report represents deliverable D1.6 “Common interface description for machine integration of in-line and in-situ metrology”. It summarizes the objectives of the NanoQI project and describes the main achievement of the first nine months with particular focus on the X-ray analysis and Hyperspectral Imaging integration into thin film processing machines.

The report comprises a preliminary description of a common integration concept (with respect to machine integration and user interaction), which will be proved and demonstrated on three industrial applications in the course of the NanoQI project and which will be ideally applicable to a plurality of thin films coating systems.

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1. Introduction

This report represents deliverable D1.6 “Common interface description for machine integration of in-line and in-situ metrology”. It is a part of Task 1.3 (Specification for machine integration of XRD and HSI tools) of Work Package 1.
The goal is to describe the general concept for combining the measurement tools X-ray diffraction and reflection analysis (XRD and XRR) and hyper spectral imaging (HSI) and integrating the measurements to industrial thin film processing machines for both at-line and inline quality control.
The report will take examples of the three industrial application cases within NanoQI and discuss the potential use to other and diverse processes. The application examples in NanoQI are:

- Roll-to-roll vacuum thin film deposition of functional coatings on plastic webs such as transparent electrodes or gas permeation barriers
- Large-area perovskite layer deposition, curing and sintering for highly efficient solar cells
- Industrial atomic layer deposition of oxide nano-laminates for flexible electronics encapsulation

The report will further discuss the possibility for homogenizing the machine integration interface for the different processing machine types and characterisation tools.

1.1. Description of measurement element tools

X-ray Diffraction and Reflection Analysis

X-ray characterisation techniques such as X-ray diffraction analysis (XRD) or X-ray reflectometry (XRR) are widely used in research laboratories to determine nano-physical dimensions (*e.g.* thickness and surface roughness) and nano structural properties (*e.g.* solid-state phase, crystal orientation and size, electron and mass density) of nanomaterials. In particular:

X-ray diffraction analysis (XRD): determines x-ray diffraction patterns in periodic solid-state materials by measuring the specular reflectance under variable angle of incidence (0° … > 90°). XRD allows determination of crystal state (occurrence of diffraction peaks), crystal orientation (intensity ratio between peaks), crystallite size (full width at half maximum (FWHM) of peaks) and internal stress (angular position of the diffraction peaks).

X-ray reflectometry (XRR): measures the specular reflectance of x-rays under an oblique angle of incidence (typ. < 5°). XRR may determine the electron density (critical angle of total reflection); surface roughness (scattering loss at increasing angle of incidence) and thickness of ultra-thin films (optical interference pattern). XRR may separate individual layers in multi-layer stacks and XRR and XRD can be combined in one stand-alone measurement unit.

Hyper Spectral Imaging (HSI)

HSI measures a full transmitted or reflected light spectrum (from ultraviolet to near infrared wavelength) on each pixel on a CCD line or CCD array camera. Full spectra are much more sensitive towards the detection of small deviations and local defects on sample surfaces in contrast to state-of-the-art grey-scale or three-color CCD images. Deviations are made visible to the user as coloured image.

Typical examples of the two metrology tools are shown below: they have been used as development basis within the NanoQI project framework.

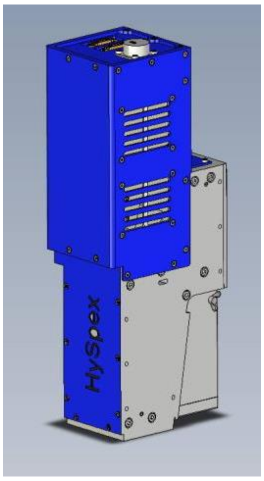


Figure 1: left: X-ray diffractometer (Bruker AXS), right: drawing of a hyperspectral camera (Norsk Elektro Optikk AS)

NanoQI distinguishes three different levels of integration of metrology and characterisation tools into thin film processing machines

Term	Measurement when?	Integration to machines
At-line	Characterisation is done **after** finishing processing and **after** taking the process batch out of the machine.	The measurement unit is separated from the processing machine but is located in proximity to the process unit for easy access by machine operators
In-line	Characterisation is done **after** finishing processing but **inside** the processing machine. Results are available before processing the next substrate; immediate parameter adjustment is possible	The measurement unit is integrated to the processing machine but in a separate chamber (located after the process).
In-situ	Characterisation is done **during** the process. Realtime results can be used to adjust the processing parameters before finishing the product.	The measurement unit must be integrated to the processing chamber respecting issues with disturbing the process through the measurement or the measurement through the process conditions (e.g. temperature; radiation.). Complex geometries are usually required to minimize interference between process and measurement.

1.2. XRR/XRD - HSI Integration

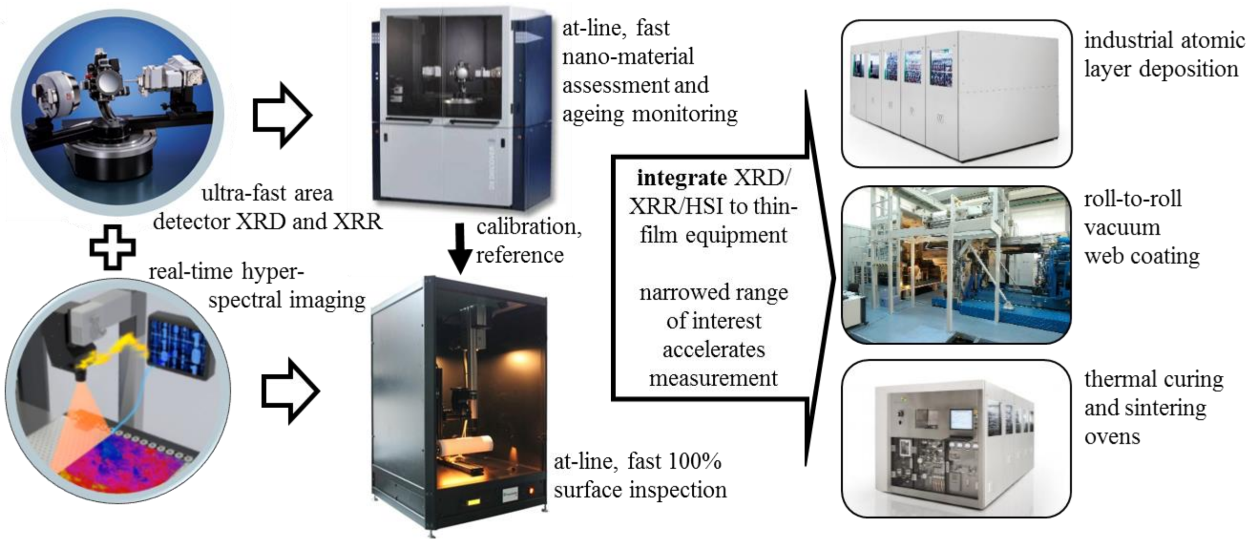


Figure 2: NanoQI concept

The schematic picture, above, represents, synthetically, the NanoQI project concept, whose main objective and ambition is to demonstrate feasibility and to create an integration platform for combining X-rays analysis and Hyperspectral camera imaging through a robust data exchange and correlative algorithm. The project aim is to take advantage from the synergy of the two systems respective characteristics that is, powerful thin film analysis capability by X-rays and fast, large area resolved and highly defined imaging of spectral camera.

1.3. Three industrial application to demonstrate the multi modal integrated metrology tool.

1.3.1. Curing oven for Perovskite slot die coatings for photovoltaic glass plates:

The curing oven is a pilot line consisting of modular drying, quenching, annealing chambers connected to each other and with driven rollers for sample transport. The machine is able to handle substrates with a width up to 300 mm. Both an XRD system and a hyperspectral camera shall be integrated directly to the machine into the process chain **as inline metrology systems**.

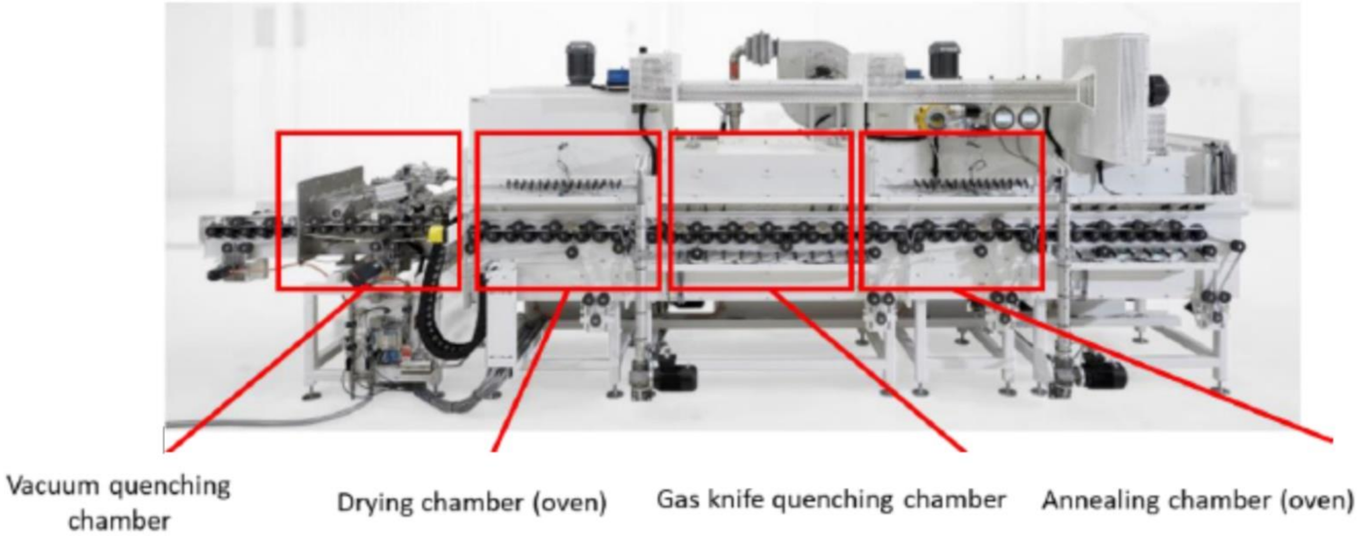


Figure 3: Sheet-to-sheet Curing Oven for Perovskite Layers for highly efficient solar cells.

1.3.2. Roll-to-roll vacuum coating

Roll-to-roll vacuum coating is used to deposit single or multi-layer thin film for different applications including optical filters, low-e and heat protection films, packaging, security, flexible electronic and sensors on plastic web substrates. NanoQI will establish XRD and XRR as fast and efficient **at-line** quality control methodology to determine layer thicknesses in multi-layer stacks, surface roughness, electron density and solid-state phase. The XRD results will be used to calibrate a **hyperspectral imaging system which is integrated directly to the coating machine** for **in-line observation** of deviations of the abovementioned properties on 600 mm coating width.

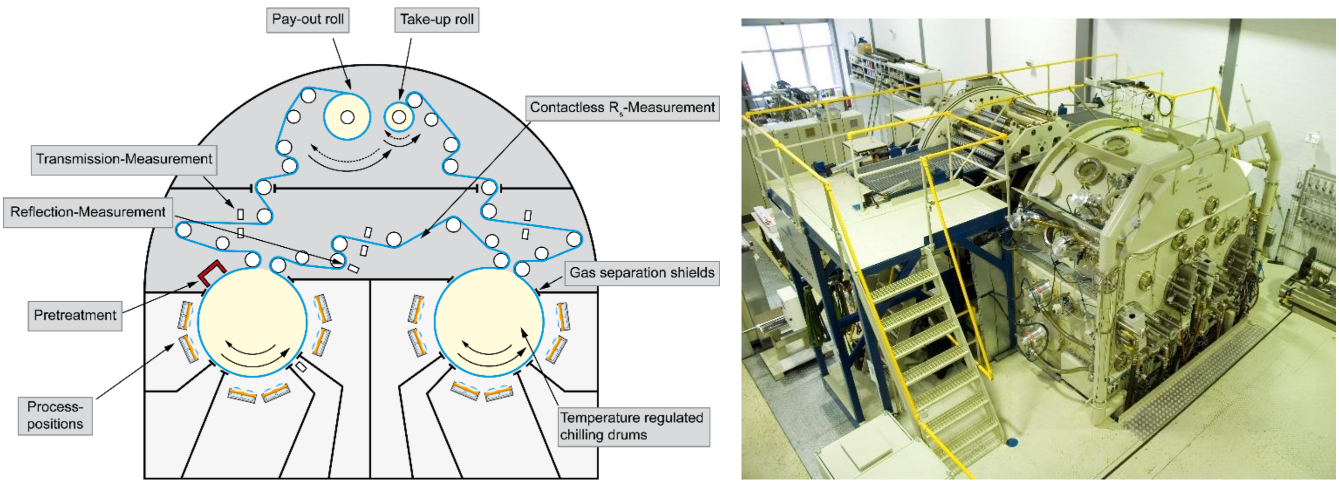


Figure 4: coFlex® 600 vacuum roll coater at FhG-FEP

1.3.3. Pilot scale Atomic Layer Deposition (ALD)

The ALD deposition chamber used in NanoQI is part of a S2S pilot line for the production of opto-electronic devices and ultra-high barrier coatings. The line is composed of an inert box system with a wet deposition chamber with printing and drying units (left part), an evaporation chamber (middle part) for thermal and e-beam evaporation of metals, oxides or organics and an encapsulation chamber which is connected to the ALD line for thin film encapsulation. Within NanoQI, a hyperspectral imaging system will be integrated to the machine in the substrate mounting chamber next to the ALD chamber for inline evaluation of layer thickness and structure homogeneity.

Figure 5: Printed Electronics Pilot Line including a deposition chamber for Atomic Layer Deposition (ALD) for device encapsulation.

2. Results and Discussion

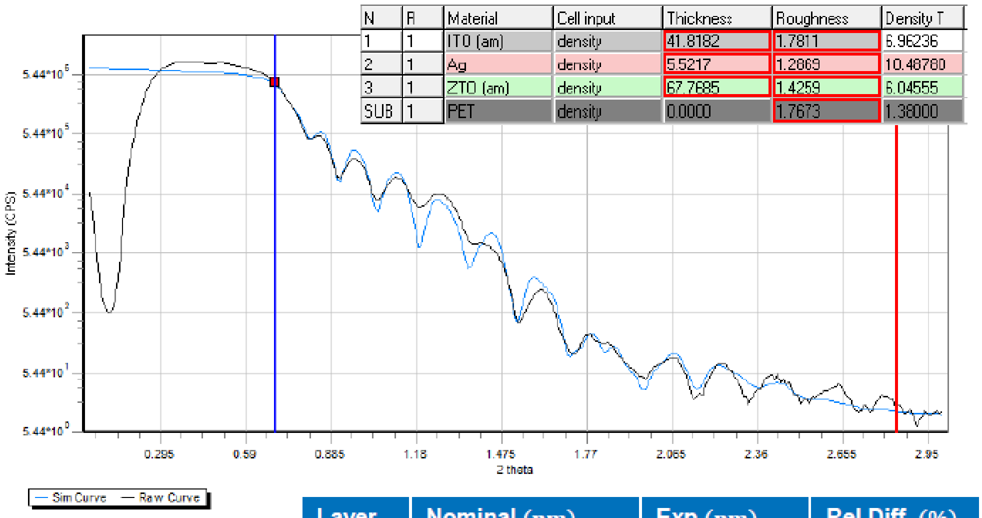
The following section will describe the understanding of measuring tools integration gained at the current NanoQI project progress (Month 9): this will represent the basis to envisage possible integration scenarios which will be confirmed and implemented with more practical solutions during the next step of project development.

2.1. Integration of measurement tools (Technology demonstration)

The first NanoQI project action has been the collection of representative samples from all partners with definition of the characterization parameters, relevant for each application. The initial measurement done by XRD and HSI indicated that the basic idea underlying the entire project is feasible: using the x-rays analysis as a reference to calibrate the hyperspectral camera and using its output to monitor and ultimately control process stability and properties uniformity. The graphs in Figure 6 show results of both methods on a transparent electrode coating on plastic web. In WP4, NanoQI will develop models and algorithms to link the reference measurement by XRD and the HSI on-line or in-situ output to allow the operational use of the integration concept for processes and machine control. Using this initial example of "multi-modal analysis", the method of the NanoQI innovative approach would include following main steps:

1. Measurement and analysis of the thin film properties of interest by XRD and/or XRR or another suitable characterisation method on a set of representative calibration samples.
2. Measurement of the same sample by Hyperspectral camera.
3. Use of a (machine learning based) algorithm linking the properties of XRD and the HSI imaging
4. Using the fast and spatially resolved response from the camera to detect properties deviation on a 2-D-sample with larger dimension and on moving samples in an industrial and production environment.

The final and key result will be to set up methods to integrate the systems to be used for industrial processing units of diverse coating technologies and application and to devise a structured training program to enable average skilled operators to use the system



Layer	Nominal (nm)	Exp (nm)	Rel Diff. (%)
ITO	40	41.8	4.5
Ag	5	5.5	10
ZTO	75	67.8	-9.6

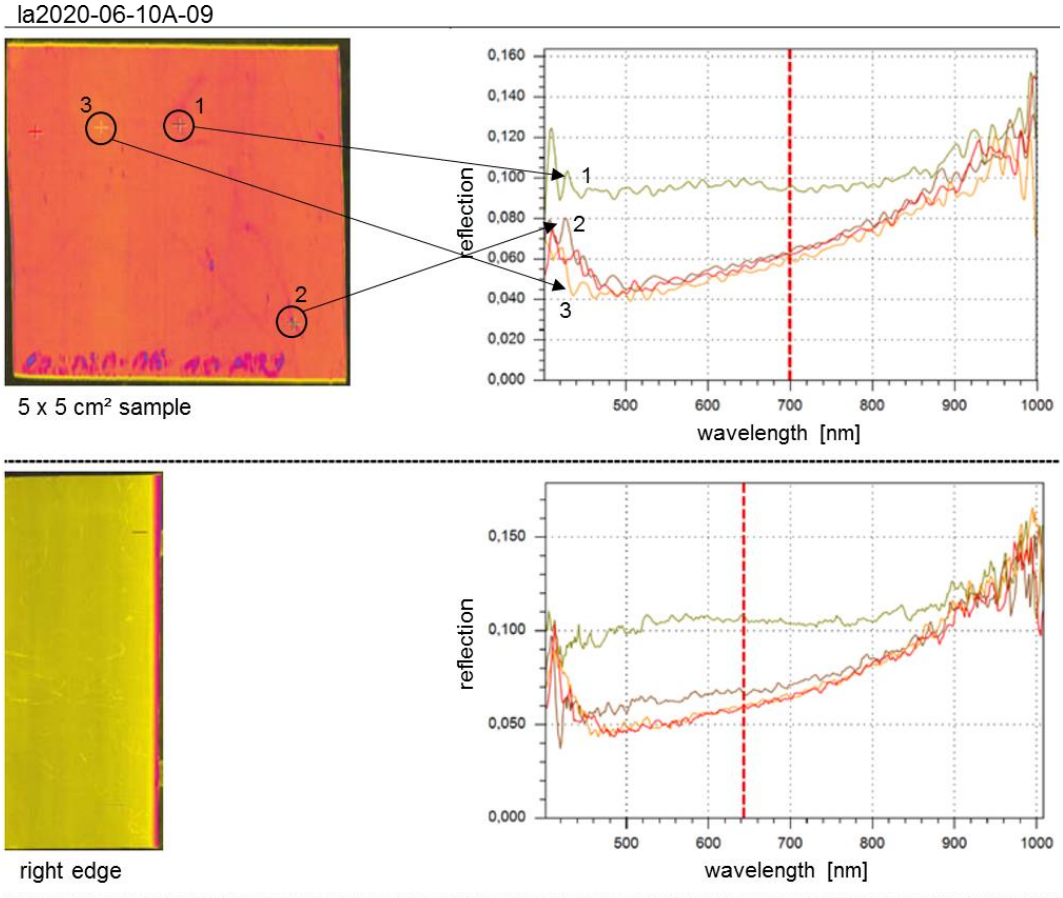


Figure 6: top: XRR spectrum and result and bottom: HSI result of a coated PET film with a three layer transparent electrode stack. The left side shows a false-colour image of a 5 x 5 cm² sample, the right side shows a spectrum for selected spots on the surface – clearly showing visible differences that would not have been detected with a camera based inspection system. Ideally the false colour image will show the spatial distribution of the relevant parameters identified by different colours, here a good representation is given for a thickness decrease on the edge.

2.2. A common platform for measurement integration

As described in the previous paragraph, the first step to compare and to develop links between the two different measuring method (XRR/XRD and HSI) is to **integrate the relevant sensors in a single platform**: Within NanoQI, a standard XRR/XRD cabinet (Bruker D8 Discover) has been modified to mount an HSI camera. This solution gives the advantage of measuring the sample with both methods from the same position, to provide a multimodal measuring centre and to use a configuration, already qualified with all the safety protection for X-rays analysis. The picture of the XRD system with the indication of the main parts and provided with camera mount is shown below. The standard diffractometer housing (bottom right) will be modified to integrate the HSI camera.

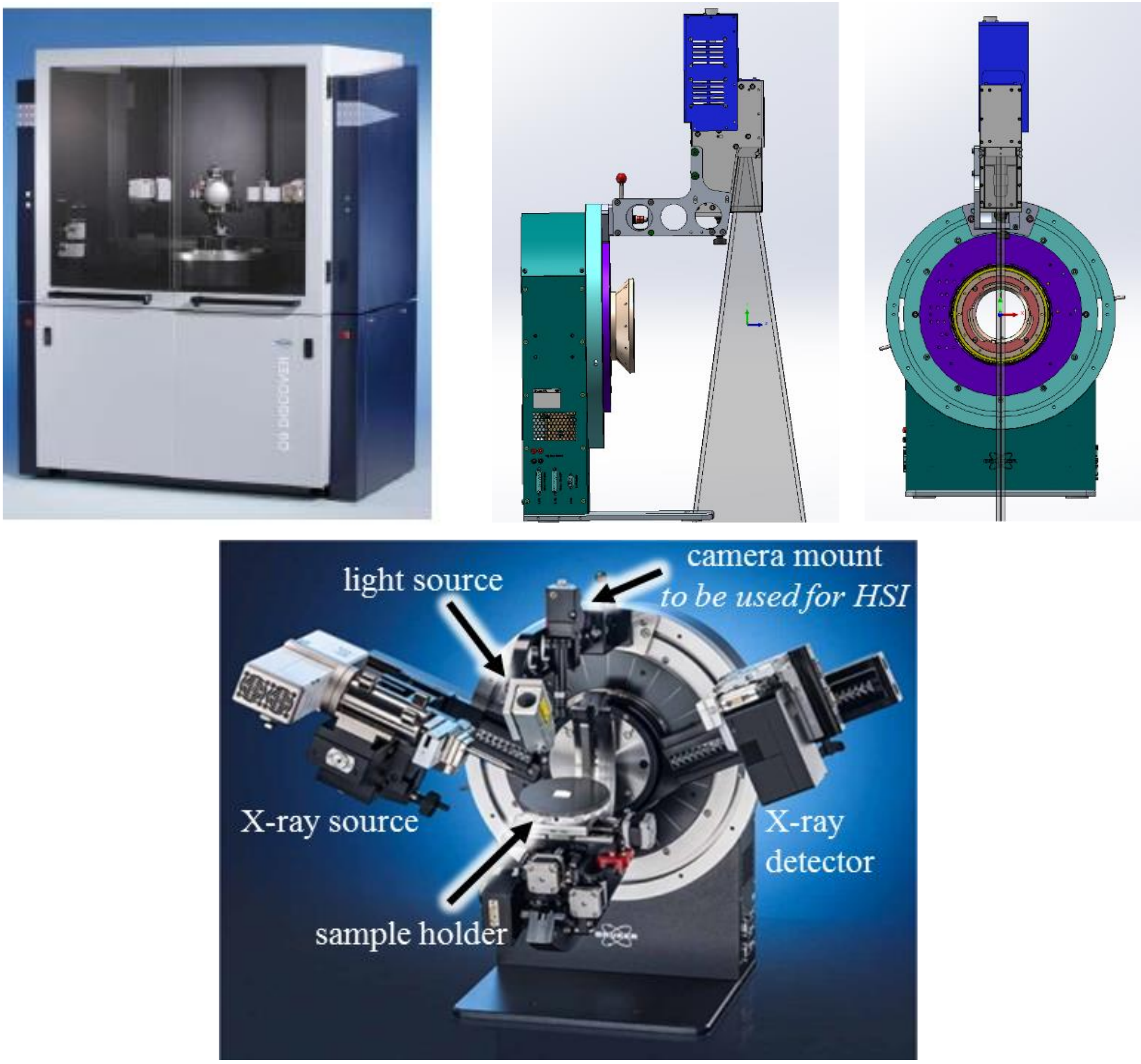


Figure 7: Top Left: Illustration of a Bruker D8 Discover Casing for XRD/XRR Systems, Top Centre and Top Right: Schematic illustration of the HSI Integration into the XRD unit; Bottom: Positioning of the HSI Camera in relation to the other components of an XRD unit.

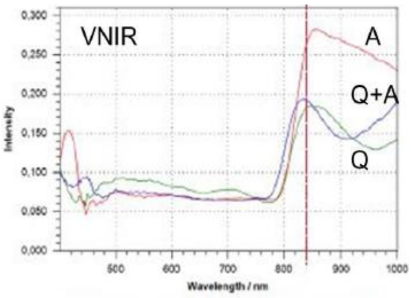
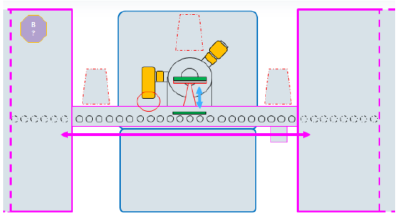
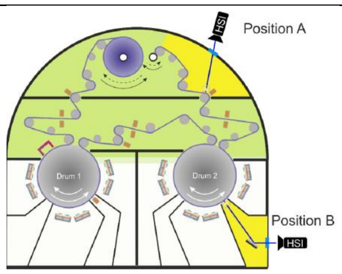
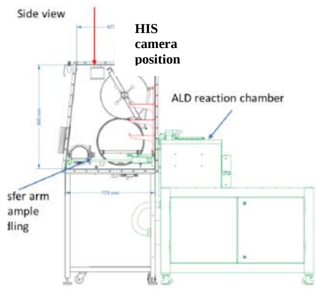
2.3. Interfaces for metrology tools on typical vacuum coating machines

The main objective of the NanoQI project is the multimodal metrology tools (XRD and HSI) integration on three different thin film manufacturing systems: for their nature, they represent a more universal spectrum of thin film deposition technologies and applications. It is the ambition of NanoQI to demonstrate the possibility of using the combined and integrated instruments to characterize, in real time, thin films with the ultimate aim of process and production control on industrial machines. In the paragraph 1.3 the three technologies and machines (existing pilot units located at three project partners premises) were described, they are summarized with their instrument integration characteristics in the table below.

Application	Partner	XR	HSI
Slot die coating curing oven	TNO	in oven	in oven
R2R coater	FhG FEP	at line	in coater
ALD barrier coater	FhG IAP	at line	at line in inert glove box

The following table represents, schematically, the typical features of each single pilot unit, which dictates the choice of the indicated metrology configuration.

	Slot die coating curing oven	R2R Coater	ALD barrier coater
representative products	Perovskite photovoltaic coating on glass plates	Multilayer coating on plastic films in rolls	Conformal, pinhole-free gas barrier coatings and high-k dielectric passivation layers
process features (relevant to metrology)	Organo metal halide Perosvskite light absorbers, applied by slot die coating are dried, quenched and annealed. These three steps are carried out inside a modular linear oven with one section for each process. The sample is transported from one section to the next one by means of programmable driven rollers. NanoQI targeted substrate width: 300 mm	Multiple nano-sized layers (thickness < 500 nm) are stacked on a substrate to achieve specific functionality. Particularly, optical coatings, thin-film sensors and electronic layers require precise thickness and surface roughness control. They are deposited on web type plastic films by diverse technologies, including thermal evaporation, sputtering, PECVD etc. Target web width: 600mm Typical web speed. 2 – 5 m/min	ALD ("Atomic Layer Deposition") allows conformal layer-by-layer growth of virtually defect free layers. The most peculiar aspect of this technology is the sequence, in cycles, of thin film formation and deposition followed by a gas-purging phase. The main quality controlling parameters are the film growth and the presence of residual components which conta-minate the layer formation.
properties of interest	The crystalline phases in the perovskite layer after quenching determine the quality of the PV cell absorber. Of interest are also surface properties and thickness.	The main properties of interest are thin film thickness (single layer and stack), surface roughness, com-position, crystallinity etc. If detected in line, they can guide process control for quality consistency.	Film thickness, roughness, electron density, crystallinity, residual carbon and hydrogen are main quality parameters: ideally, they are measured in situ after each cycle.
point of detection and samples handling	At this project stage, there are different options for XRD/HSI location: **"in-situ"**: inside a quen-ching chamber or **"in line"** inside a separate measuring section (module) installed *e.g.* between the quenching and annealing chamber. Most likely, the XRD measurement will be in-line in a separate measuring section as shown below, while the HSI measurement will be in-situ measuring through an observation window in the quenching section.	For this type of process, the NanoQI project features "at line" XRR detector and "in-line" HSI. The hyperspectral camera will preferably be installed outside the vacuum chamber to inspect the web through a glass window. The optimized location on the existing pilot machine will be decided seeking the best trade-off among field of view (and related scanned web width), sample illumination, web stability and resolution. The schematic below represents a possible solution ideally suited and adaptable to any roll-to-roll equipment, designed with a web inspection window.	The pilot ALD system is provided with a "glove-box" chamber connected to the process unit for inert sample handling. The Hyperspectral camera will be installed in the glovebox chamber. The XRR/XRD system will be "at-line". The schematic, below, represents the most likely HSI position: final configuration will be decided depending on the selected camera cooling solution. It is ideally adaptable to any coating machine having a geometrically separated but functionally process connected measurement zone.

	Slot die coating curing oven	R2R Coater	ALD barrier coater
XRR/XRD characteri-zation capability	The XRD pattern shows the presence of formed crystallization phases; target time :<12 sec for a limited angle interval for monitoring the presence of selected (required or unwanted) phases.	The graph shown in section 2.1 represents an example of thin film stack characterization by XRR; in particular it measures layer thicknesses and roughness. It is a single spot detection with detection time <12 sec. Deviations over a large substrate area are not accessible in real-time using XRR as each spot must be measured individually.	XRR may precisely (accuracy ±1 monolayer) determine the thickness, surface roughness and density of ALD layers on a single spot.
HSI integration capability	HSI can detect the differences among treatments (in the example A = annealing; Q=Quenching) : through correlative properties algorithm, HSI can represent phases presence, layer thickness and surface properties on a 2D sample ("calibrated" from XRD spot measurement) 	Hyperspectral camera spectra and/or colour images represented in the picture in section 2.1 show reflection diagrams on a few sample locations and on the visible and near infrared (VNIR) and on Short Wave Infrared (SWIR) spectra. The NanoQI program ahead has the objective of creating a correlation model between the sample "at line" measurement by XRD and HSI output.	Hyperspectral camera spectra and/or colour images represented in Figure 8 below, refers to the same sample type (Al_2O_3 on glass) characterized by XRR before. It gives a 2-D image (the sample must be moved under the fixed camera to get sample scanning) showing properties deviation. NanoQI will create a correlation model between the sample "at line" measurement by XRR and "in-situ" HSI output.
Choice of Metrology tool configuration	The XRD unit will be inserted in-line to the processes between two oven modules as a separate chamber including a conveyor belt for continuous substrate movement. HSI measurement "in situ" will be done through a window in a quenching section (chamber).	XRR/HSI measurement cabinet "at line". HSI hyperspectral camera will be installed at the selected spot on the R2R machine and will have a basic software interface and link with the coater operation. The hardware integration makes use of standard vacuum flanges (DN 250 – ISO KF).	Hyperspectral camera "in line" inside a measuring box (in the pilot ALD example, it will be located inside the glovebox chamber) "At-line" XRR-XRD cabinet will provide "calibration" data to the HSI
Schematic diagram			

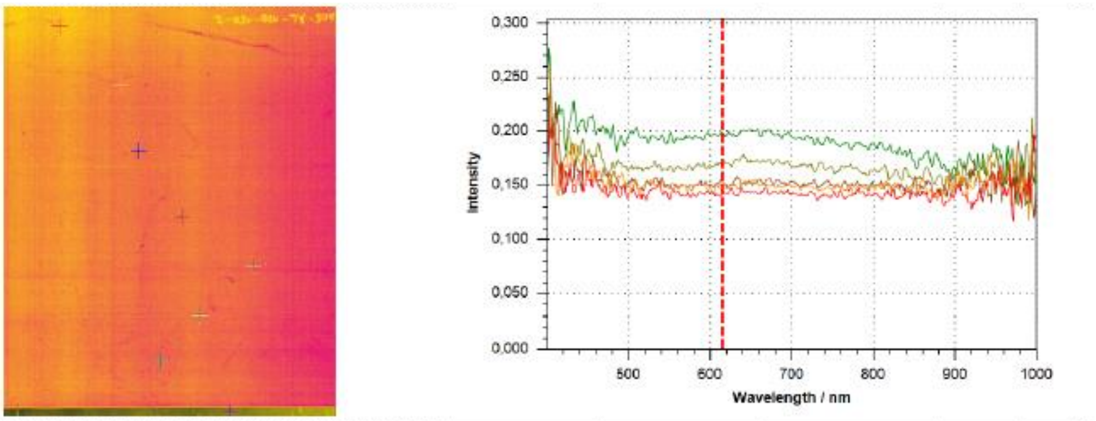


Figure 8: Hyperspectral imaging result on Al_2O_3 layers grown by ALD showing a thickness gradient from left to right. The false colour images show a ≈ 10 x 15 cm² section of a 15 x 15 cm² large sample.

2.4. Common interface for in-line and in-situ metrology integration – Summary

The previous paragraphs have described the solutions for the integration of X-rays analysis and Hyperspectral camera with the aim of developing an innovative and advanced metrology tool for thin films deposition systems. In the NanoQI project framework, these solution will be applied and demonstrated on three pilot units representing specific processes and applications: due to their diverse technologies and equipment configurations, the same concepts can be potentially used on a much broader ranges of nanomaterials deposition systems. The general concepts is visually shown in the following picture:

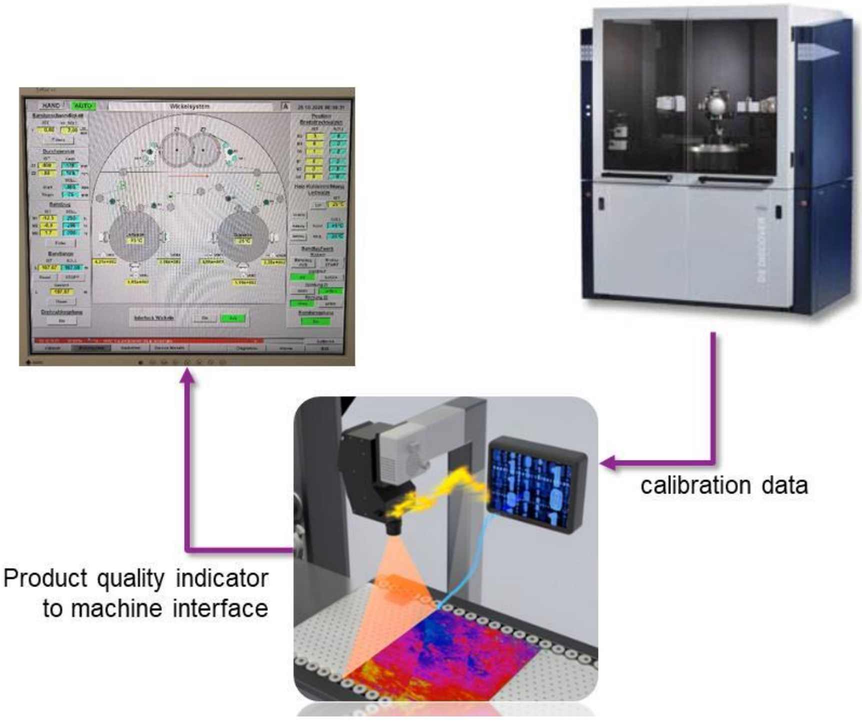


Figure 9: Data processing and coating machine integration workflow: The XRD / XRR system will deliver calibration information to a machine learning algorithm integrated to the HSI data evaluation software. This software shall create a quality indicator quantity and a visualization of large area homogeneity that are passed to and displayed within the machine operation software so that the operator is immediately able to react in case the product properties are moving out of the specified range.

The metrology tool set comprises two units:

a) An x-ray diffractometer/reflectometer housed inside a cabinet designed and certified for radiation safe operation. The system will be provided with fittings and clamping to mount a hyperspectral camera to measure both XRD and HIS on the same spot for highly accurate calibration data preparation. The design/dimensions of the cabinet can be adapted to the specific machine and application. The X-ray measurement determines the application relevant thin film properties efficiently and delivers calibration data to the in-line / in-situ HSI system.

b) A hyperspectral imaging unit (comprising a camera and adapted optical path) with wavelength range and optical features selected according to the process and machine conditions, designed for atmospheric installation. The hyperspectral imaging system will visualize deviations of the application relevant thin film properties on large product surface areas [up to 600 mm width in NanoQI] based on calibration data received from XRD / XRR measurement or other characterisation tools. NanoQI will develop a standardized data exchange strategy / format (e.g. based on HDF5, CHADA or other suited data formats).

2.4.1. Commonalities in machine integration interfaces

Within the first 9 month of the NanoQI Project it became already clear that a common interface for machine integration for **in-line** integration of **optical metrology units** (such as HSI) has to be discussed on machine type level. In vacuum coating machines and in thermal curing oven, camera mounts can be based on standard DN-ISO KF/CF flanges (DN 63 … DN 250). However, optical paths and illumination have to be adapted individually to each machine layout.
XRD and XRR integration to machines requires specific adaption to the concrete machine layout. The strategy to design the XRD System as an attachable machine module (as NanoQI will demonstrate with the curing oven) has the strong advantage of being able to use the radiation proof casing of the XRD and so simplifying radiation safety requirements for the machine. In-situ integration requires specific design and adapted machine layouts to fit the goniometer configuration.

2.4.2. Commonalities in data exchange and operator/machine intefaces

The correlation of XRR/XRD and HSI measurements represents a very important NanoQI project area, and implies thin film properties modelling, data evaluation algorithms and big data processing. At the end of this project, we can expect a typical use of the integrated metrology system as follows (e.g. the multi-layer R2R coater but the other applications can be handled in equivalent manner):

- XRR/XRD as reference for job set-up: The operator will search from a recipe database (previously prepared by specialist of XR-analysis) the simulated characteristics of the layer stack and will compare with a produced product prototype on the "at-line" or "in-line" metrology unit.
- The XRR unit will indicate the actual measured parameters and deviation from the set specification: the operator can adjust the process parameters on the coater until reaching the required properties.
- The at-line unit will calibrate the "in-situ" or "in-line" instrument (HSI) to control the continuous coating process by representing and alerting the operator on deviation from the specifications. Depending on the process complexity and as a further development after the NanoQI project, the HSI output can drive a process control variable for automatic feedback control.

The described metrology tool could be used for a plurality of other operations, such as quality control, product development, thin layer characterization etc., all, in general, implying the data exchange through a robust and user's friendly interface between the x-ray diffractometer, the source of thin film properties detection and analysis and highly dynamic and sensitivity Hyperspectral cameras. The HSI will enable real time data acquisition on moving or stationary samples at industry-suited spatial resolution to detect small properties deviation using reference data delivered by XRR/XRD.
The NanoQI ambition is to create data processing, and algorithms including machine learning procedures to allow use of the multi-modal metrology interface by operators not specifically skilled in the data analysis after a limited training period, included, for the three prototypes, in the NanoQI program.

3. Conclusions

The measures of multiple samples representative of different thin film applications has confirmed the feasibility of x-rays analysis and Hyperspectral camera integration. A common platform, consisting of XRR/XRD diffractometer, upgraded for accelerated thin film analysis and of a Hyperspectral camera, of improved high speed detection and sensitivity and mounted on the diffractometer frame and housing, will be customized to suit different thin film technologies requirements. They can be used “at line”, “in-line” or “in situ” depending on the machine design and functions. Once that correlative algorithms will be developed within the NanoQI project program (WP 4) and tested on the three demonstration thin film lines, the same integrated metrology concept can be used for a plurality of other processes and applications where thin film real-time characterization is a key quality requirement.

4. Degree of progress

Degree of fulfilment of Deliverable D1.6 is 100 %.

5. Dissemination level

This Deliverable is a Public (“PU”) Report and can be made available to free accessible resources. It will be shared with all members of the consortium and with the Commission Services.

6. References