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TABLE OF CONTENTS

1 Executive summary

Introduction

Monitoring humidity under transient conditions and at temperatures above 100 °C is a key factor in controlling drying processes. In Europe, every 0.1 % improvement in drying efficiency due to better process control can save around 30 M€/year. By improving the reliability of these humidity measurements, annual savings of millions of euros could be achieved in Europe. The aim of this project was to significantly improve the accuracy and efficiency of industrial humidity measurements at high temperatures up to 180 °C, pressures up to 6 bar, and under non-static conditions by developing improved humidity measurement and calibration techniques.

The Problem

The quality and shelf life of pharmaceutical products are significantly affected by the humidity of the ambient air during manufacture and storage. Pharmaceutical companies allocate significant resources in order to maintain high quality monitoring of humidity in their production premises and storage facilities. These companies were therefore seeking more efficient calibration methods for humidity sensors and improved methods for spatial humidity monitoring under transient conditions to achieve savings both in costs and materials.

The food industry is the second largest manufacturing sector in the EU with a total manufacturing turnover of over 900 billion euros. Drying and baking are key processes in this sector, and water activity – i.e. equilibrium relative humidity – is a key parameter in controlling the quality of food and feed products. In order to determine water activity, material samples are taken from the process and measured using laboratory analysers. A significant quantity of material is wasted, because of slow feedback, and the optimisation of energy consumption is limited by the larger safety margins that are required due to uncertainties, which are increased by sampling errors and transient conditions.

More than 180 accredited industrial laboratories in Europe calibrate hygrometers for their customers but, at the beginning of the project, none of them were able to perform calibrations at temperatures above 100 °C. In addition, calibration methods for transient humidity conditions were not available. Furthermore, there was no measurement technology available for monitoring fast transients in humidity in the temperature range above 100 °C. Dynamic humidity measurements are an integral part of the environmental tests for various industrial products, however no proper methods existed to estimate their uncertainty.

The Solution

A high-temperature mass fraction water vapour/steam generator and two new calibration/testing facilities were developed to provide traceable humidity standards at temperatures up to 180 °C and pressures up to 6 bar (abs). With the developed methods, the uncertainty in relative humidity calibrations was 1 % and 0.13 °C in dew-point temperature. New calibration schemes with an appropriate calculation method were developed to provide efficient approaches to perform humidity calibrations in a range of temperatures or using humidity ramps. A modular calibration setup was developed to provide a practical approach for carrying out humidity calibrations at non-static conditions in commercial temperature/climatic test chambers. Several capacitive and acoustic humidity sensors were tested to determine appropriate calibration procedures for industrial use. The calibration facilities and schemes have been applied to calibrate humidity sensors in industrial applications: spray drying in dairy manufacturing and lambda sensors in biscuit production.

Three prototypes of a field humidity calibrator were designed and tested. One of them was demonstrated in a pharmaceutical plant and in hands-on humidity training for industry and accredited calibration laboratories. Feedback was collected for further development and commercialisation of the device.

A new type of hygrometer based on direct Tuneable Diode Laser Absorption Spectroscopy (dTDLAS) was developed for non-static process environments with temperatures up to 180 °C with a response time well below 1 s. Validation measurements were performed in the temperature range between 40 °C to 180 °C. The target uncertainty, 5 %, was met in the temperature range 40 °C - 100 °C. The dTDLAS instrument was applied in an industrial demonstration on non-static in-line humidity measurement in a hazelnut drying and roasting process. A new measurement method for detecting the influence of microbiological processes on the transient humidity conditions within small samples was developed and demonstrated with sterile Petri dishes.

A new SI traceable in-line water activity measurement method was developed, validated and launched as a service. The method establishes the traceability link between water activity and water mass fraction measurements through sorption isotherm measurements. The uncertainty of the method is around 2 %. A software tool for modelling the temperature dependence of sorption isotherms and transient water activity in hazelnuts was also developed and reported as a journal article. The water activity measurements were demonstrated in pet-food manufacture and whole hazelnut drying.

Impact

This project delivered new and improved methods and techniques for humidity measurement and monitoring at high temperatures and under transient conditions enabling a wide range of industrial enterprises in the EU to enhance their competitiveness through reduced energy consumption and waste production and through more efficient and reliable quality assurance and new products. The outcomes have already been exploited by the industrial partners of the project, and manufacturing industry. To assist the industrial and scientific communities, new SI traceable measurement and calibration services in high dew-point and air temperatures as well as in sorption isotherm and water activity determination have been launched.

The calibration schemes developed in the project save 40 % of the calibration time in a typical calibration scheme. Faster and more automatic calibration schemes reduce the downtime of the devices under calibration, but they also make the calibration work more economically sustainable for the accredited laboratories.

More accurate measurements improve product quality in food, pharmaceutical and paper manufacturing, which results in reduced amounts of waste. On the other hand, energy savings can be expected, as the products will no longer be over-dried. In-line measurements of water activity applied in the food and feed industry will improve consumer safety by improved product safety and prolonged shelf-life of the food or feed products - in a cost-effective manner.

The procedures developed have been used as input in a draft guideline “A practical approach for estimating the uncertainty of humidity measurements at high temperatures (up to 180 °C) and under transient conditions in industry” to be published as a EURAMET guide. The partners have actively participated in two standardisation groups of DIN and ISO and in five metrology working groups of EURAMET and CIPM.

2 Need for the project

Prior to the start of the project, drying was estimated to cost European industry around 30 000 M€ per year in associated energy costs. Every 0.1 % improvement in drying efficiency due to better process control could save around 30 M€/year. Monitoring humidity under transient conditions and at temperatures above 100 °C is a key factor in controlling drying processes. Thus, by improving the reliability of these humidity measurements, annual savings of millions of euros could be achieved in Europe.

The quality and shelf life of *pharmaceutical products* are significantly affected by the humidity of the ambient air during manufacture and storage. Pharmaceutical companies allocate significant resources in order to maintain high quality monitoring of humidity in their production premises and storage facilities. These companies were therefore seeking more efficient calibration methods for humidity sensors and improved methods for spatial humidity monitoring under transient conditions to achieve savings both in costs and materials.

The *food industry* is the second largest manufacturing sector in the EU with a total manufacturing turnover of over 900 billion euros. Drying and baking are key processes in this sector, and water activity – i.e. equilibrium relative humidity – is a key parameter in controlling the quality of food and feed products. In order to determine water activity, material samples are taken from the process and measured using laboratory analysers. A significant quantity of material is wasted, because of slow feedback, and the optimisation of energy consumption is limited by the larger safety margins that are required due to uncertainties, which are increased by sampling errors and transient conditions.

More than 180 accredited *industrial laboratories* in Europe calibrate hygrometers for their customers but, at the end of the project, only one of them was able to perform calibrations at temperatures above 100 °C. In addition, calibration methods for transient humidity conditions were not available. Furthermore, there was no measurement technology available for monitoring fast transients in humidity in the temperature range above 100 °C. Dynamic humidity measurements are an integral part of the *environmental tests* for various industrial products, however no proper methods existed to estimate their uncertainty.

3 Objectives

The overall objective of this research was to improve the efficiency of material and energy usage and to improve quality control in industry by developing improved measurement and calibration techniques for humidity at high temperatures and under transient conditions. The main objectives were:

1. **To develop humidity calibration methods and procedures for industrial use for air temperatures above 100 °C and absolute pressures from 0.5 bar to 6 bar.** The uncertainty levels appropriate for relevant applications (typically less than 2 % relative humidity) will be achieved with the minimum additional work load and equipment costs.
2. **To develop humidity calibration methods and procedures for industrial use applicable to transient conditions,** including development of a prototype field humidity calibrator. The target is for the uncertainty of the calibration to be the same as when performed under steady state conditions (i.e. better than 2 % relative humidity) but for the time needed for the industrial calibration to be reduced by at least 50 % from up to one day.
3. **To develop humidity measurement techniques and procedures for the accurate monitoring of temporal and spatial humidity variations in selected applications**, including development of a new type of hygrometer based on direct Tuneable Diode Laser Absorption Spectroscopy (dTDLAS) for process environments with temperatures up to 180 °C and a new measurement method for detecting the influence of microbiological processes on the transient humidity conditions within small samples. The target relative uncertainty for the water vapour amount fraction at high temperatures is 5 %, and the uncertainty for the relative humidity measurements will be less than 2 %rh.
4. **To develop water activity measurement (equilibrium relative humidity) techniques for in-line measurement applications** - with a measurement uncertainty smaller than 0.02 - **and to develop methods for establishing the traceability link between water activity and water mass fraction measurements** including the development of tools for analysing error sources in water activity measurements which are an integral part of sorption isotherm measurements.
5. **To validate all of the methods developed in this project through demonstration in selected industrial applications** in order to facilitate the take up of the technology and measurement infrastructure developed by the project and to recommend what further actions are necessary to ensure the uptake.

4 Results

- **Objective 1: To develop humidity calibration methods and procedures for industrial use for air temperatures above 100 °C and absolute pressures from 0.5 bar to 6 bar. The uncertainty levels appropriate for relevant applications (typically less than 2 % relative humidity) will be achieved with the minimum additional work load and equipment costs.**

A high-temperature mass fraction water vapour/steam generator and two new calibration/testing facilities have been developed to provide traceable humidity standards at temperatures up to 180 °C and at pressures up to 6 bar (abs). Two acoustic and 5 capacitive humidity sensors have been tested with the developed set-ups to determine appropriate calibration procedures for industrial use. IH, Michell and Vaisala provided these sensors and gave industrial input to the measurement procedures that were developed.

A new humidity calibration set-up has been developed at INRIM. The operating principle is based on a mass fraction vapour/steam generator that delivers a humid gas stream to a sub-chamber that is accommodated in a climatic chamber. The water mass fraction of the outlet air is controlled by SI traceable mixing of a water mass flow evaporated into a dry air flow. This provides SI-traceability for humidity measurements at air temperatures up to 180 °C, dew-point temperatures up to 140 °C and pressures up to 6 bar (abs). A microwave cavity resonator, also hosted in the sub-chamber, is integrated into the system to provide accurate measurements of water vapour amount fraction.

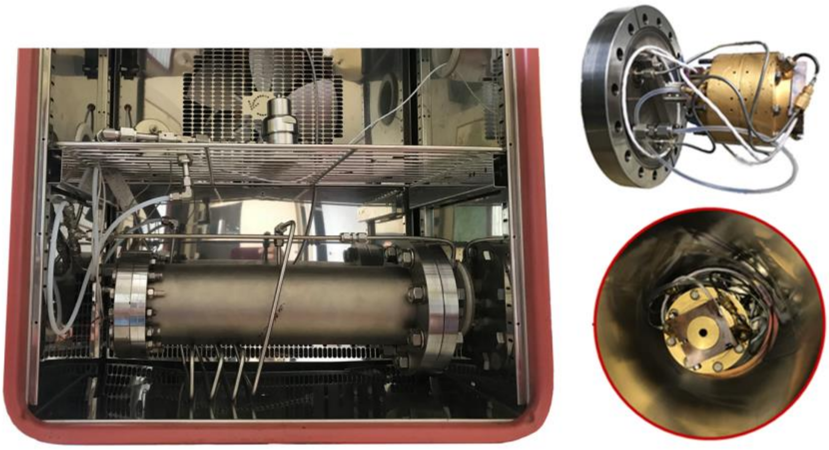


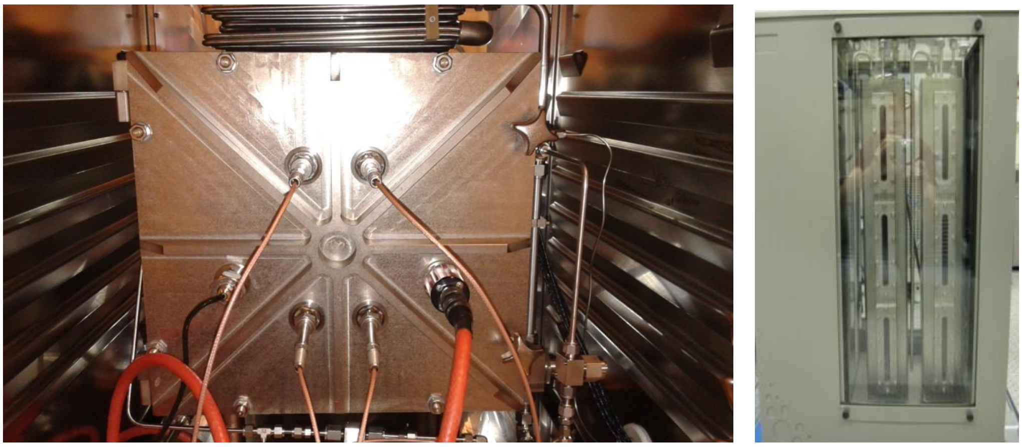
Figure 1. Calibration set-up. Sub-chamber hosted in a climatic chamber (left side) and microwave resonator hosted in the sub-chamber (right side)

The vapour/steam generator has been validated using absolute water vapour amount fraction measurements in the gas phase, both by exploiting microwave-based hygrometry and by comparison against a primary humidity standard in the relevant dew-point sub-range. Microwave hygrometry is a well-established method that is able to detect the vapour amount fraction of a binary mixture with high precision using the cavity microwave resonance frequencies. For a binary mixture of water vapour in nitrogen or air, the simultaneous measurement of pressure, temperature and dielectric constant provides a measurement of the composition of the mixture in terms of the water vapour amount fraction.

The measured water vapour amount fraction and the humidity generated by the mass fraction vapour/steam generator were compared over a temperature range from 80 °C to 180 °C and in the pressures from ambient air pressure up to 6 bar (abs). In the investigated range, the relative standard uncertainty of the microwave hygrometer was about 1 %. Data fusion techniques have been used to minimise the uncertainty levels.

At VSL, the operating temperature, dew-point and pressure ranges of the existing humidity chamber were extended to 180 °C, 150 °C and 6 bar (abs), respectively. These extensions to the operating ranges were achieved by further implementing an existing set-up, which was capable of achieving a carrier gas temperature of 180 °C and a pressure of 600 kPa, but it was limited by a maximum dew-point temperature of 95 °C by the saturation in the water bath.

The maximum dew-point temperature of the extended system, 150 °C, was achieved by replacing the water bath with a mineral oil bath, whose temperature can be controlled up to 150 °C.



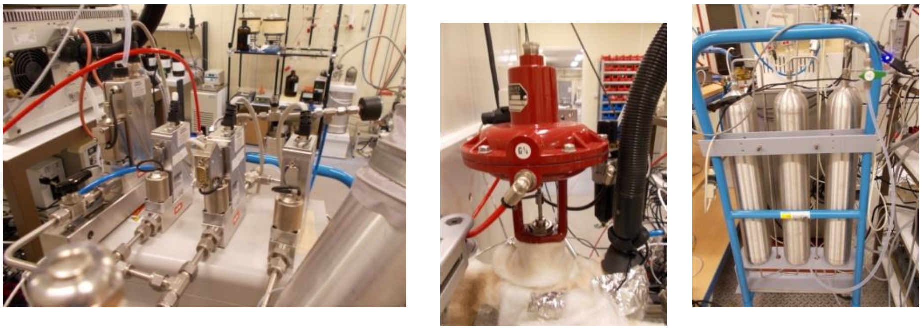


Figure 2. Overview of the high temperature relative humidity facility. Clockwise, the pressure chamber with industrial sensors mounted in the door, the saturator in the mineral oil bath, the pressure control valve, storage tanks for automatic refill of the saturator and the mass flow controllers for the dry and wet gas flow (far right in the picture) and the refill of the saturator (left in the picture).

The dew-point temperature of the gas in the sub-chamber is measured with Chilled Mirror Hygrometers (CMH's). Depending on the required uncertainty, the dew-point temperature can be measured inside the pressure chamber (at pressure) or, when high-level accuracy is needed, via a sample line downstream of the pressure control valve on the expanded gas. The CMH's are calibrated against VSL's primary High Temperature Saturator (HTS) up to 95 °C with uncertainties up to 0.08 °C. For the highest dew-point temperature of 150 °C at 600 kPa, the dew-point temperature of the expanded gas (to nearly atmospheric pressure) is 93.8 °C, which is within the range of the HTS. The uncertainty of the dew-point temperature measurement is 0.13 °C.

The calibration rig at CETIAT (France) is based on a closed loop or complete recirculation principle. The humid air generator can be used according to two configurations: one is dedicated to dew/frost point temperature calibration and the second one to relative humidity calibration (Figure 3). The air flows through a coil, acting as a heat exchanger, and then through the equilibrium chamber of the saturator containing a sheet of water, which can be liquid or solid, depending on the working temperature. In transient conditions, an exchange of water molecules takes place between water and air. As soon as thermal and mass equilibrium is reached, the exchange rate of water molecules between water and air is balanced. The partial pressure of water vapour is directly linked to the equilibrium temperature reached in the saturator, which reflects the dew/frost point temperature. Thus, the traceability to the SI units is ensured by the calibration of the temperature probe inside the saturator which represents the dew/frost point temperature when the system is at equilibrium. The SI traceability of the relative humidity is ensured by the calibration of the temperature probes inside the saturator and inside the testing chamber. The first one represents the dew/frost point temperature, and the second one the dry temperature, when the system is in equilibrium.

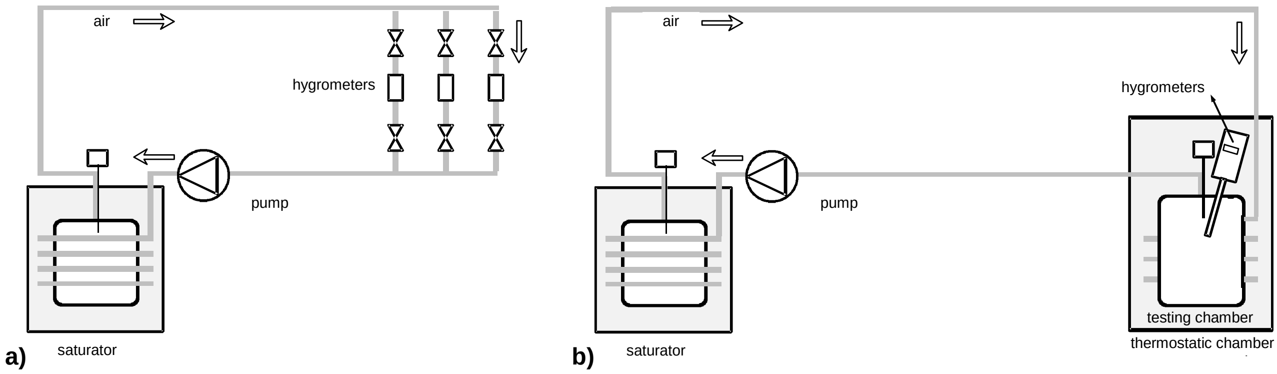


Figure 3. The Humid Air Generator of CETIAT, working in a) dew-point calibration and b) relative humidity calibration mode

Before the HIT project the dew/frost point temperature calibration range at CETIAT was from -80 °C to +80 °C. During the project, the calibration range was extended up to +90 °C. An interlaboratory comparison was carried out to validate this extension. The comparison included two CMHs, MBW 573 type, within the range from -20

°C up to +90 °C. The extension up to +90 °C in dew point temperature required upgrade in order to avoid unwanted parasitic condensation in the tubes delivering the humid air. An additional heating element was added to the head of the pump (Figure 4), as well as a new cover for reaching high temperatures (Figure 5).

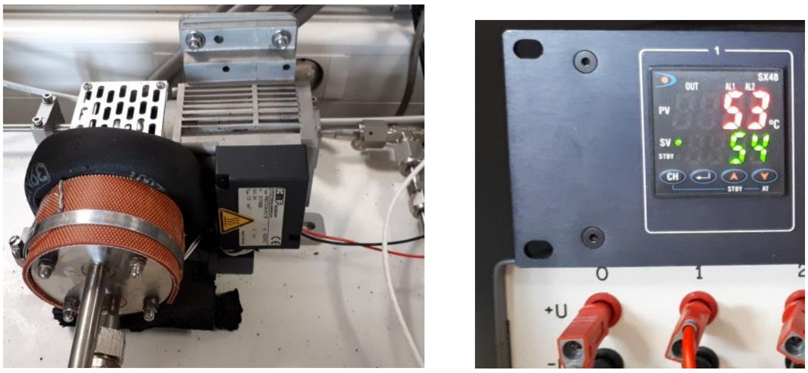


Figure 4. The heating element associated with its temperature controller

Figure 5. Cover that prevents unwanted condensation at dew-points up to +90 °C

A test facility to characterise humidity sensors at elevated relative humidity levels and temperatures up to 100 °C was set up at PTB, Germany. The facility has been qualified as a secondary national standard humidity standard in Germany. This facility was used to validate a dTDLAS (direct Tuneable Diode Laser Absorption Spectroscopy) based hygrometer developed by TU-DA. The dTDLAS is capable of operating as a humidity transfer standard and in process environments up to 180 °C with a response time well below 1 s, therefore it is suitable for non-static humidity measurements.

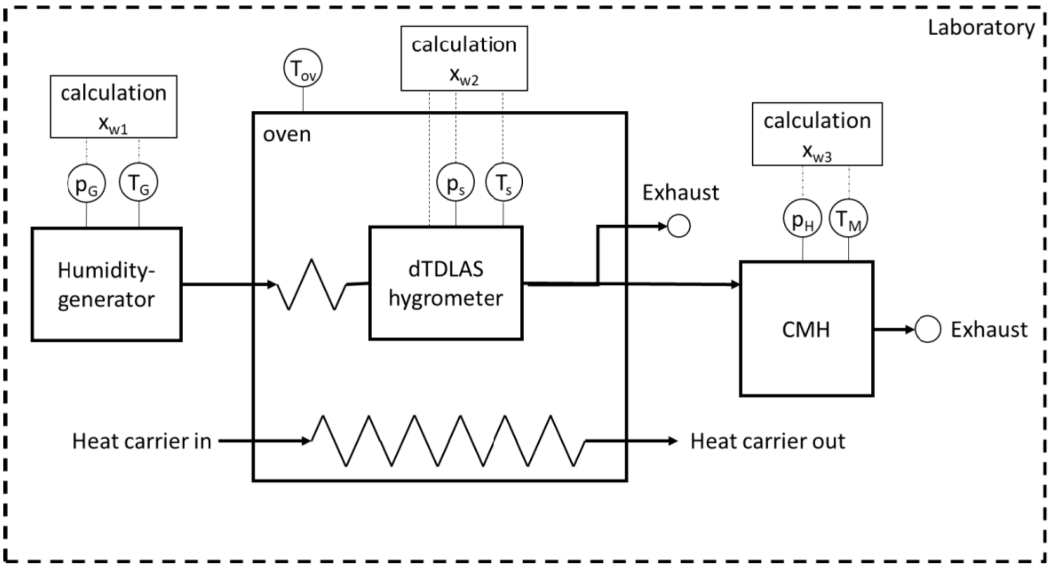


Figure 6. Schematic of the validation setup of the dTDLAS hygrometer at PTB

Validation measurements were successfully performed at ambient pressure and temperatures between 40 °C to 100 °C: The developed dTDLAS hygrometer has shown relative deviations of less than 5 % compared to the reference value, confirming its capability to be used as a transfer standard.

The outcomes of Objective 1 were:

- A high-temperature mass fraction water vapour/steam generator and two new calibration/testing facilities were developed to provide traceable humidity standards at temperatures up to 180 °C and pressures up to 6 bar (abs). Five capacitive and 2 acoustic humidity sensors were tested to determine appropriate calibration procedures for industrial use. With the developed methods, the uncertainty in relative humidity calibrations was 1 % and 0.13 °C in dew-point temperature.
- The dDTLAS spectrometer developed by TU-DA was validated with the PTB humidity characterisation facility. The target uncertainty of 5 % was met.
- Based on the experiments, VSL, CETIAT, INRIM, PTB, VTT, IH, Michell, TU-DA and Vaisala prepared recommendations for humidity calibrations in the temperature range 100 ^{0} C - 180 ^{0} C. The procedures developed were used as input to a draft guideline “A practical approach for estimating the uncertainty of humidity measurements at high temperatures (up to 180 °C) and under transient conditions in industry”.
- This objective was fully achieved.

- **Objective 2: To develop humidity calibration methods and procedures for industrial use applicable to transient conditions, including development of a prototype field humidity calibrator. The target is for the uncertainty of the calibration to be the same as when performed under steady state conditions (i.e. better than 2 % relative humidity) but for the time needed for the industrial calibration to be reduced by at least 50 % from up to one day.**

At CETIAT, the calibration facility described in Objective 1 was also upgraded to generate controlled humidity steps. At VTT, a measurement setup for calibrations under non-static conditions and a procedure for applying continuously changing conditions was developed and validated. Six different capacitive humidity sensors were tested in transient conditions. Michell and Vaisala provided humidity sensors for the tests.

VTT’s modular system comprises a humidifier and a measurement chamber assembly (Figure 6a) that can be easily installed in any commercial temperature test chamber with an inner volume of 200 dm^3 or larger (Figure 6c). The humidity generation is based on mixing dry and humidified air using mass flow controllers located outside of the temperature chamber (Figure 6b). The humidifier is based on the bubbler principle, in which air is humidified by passing it through a water filled cylinder. To compensate for evaporation induced cooling, the water is heated. The generated humid air is thereafter mixed with dry air before it enters into the measurement chamber from the back of the chamber. In order to properly mix the air inside the measurement chamber, and thus to minimise temperature and humidity gradients, a fan is placed inside the chamber close to the humid air inlet (Figure 6d). Quick couplings are used for easy assembly of the sample lines. A dedicated LabView program was developed for controlling the calibration system to enable automatic calibrations in static and non-static conditions.

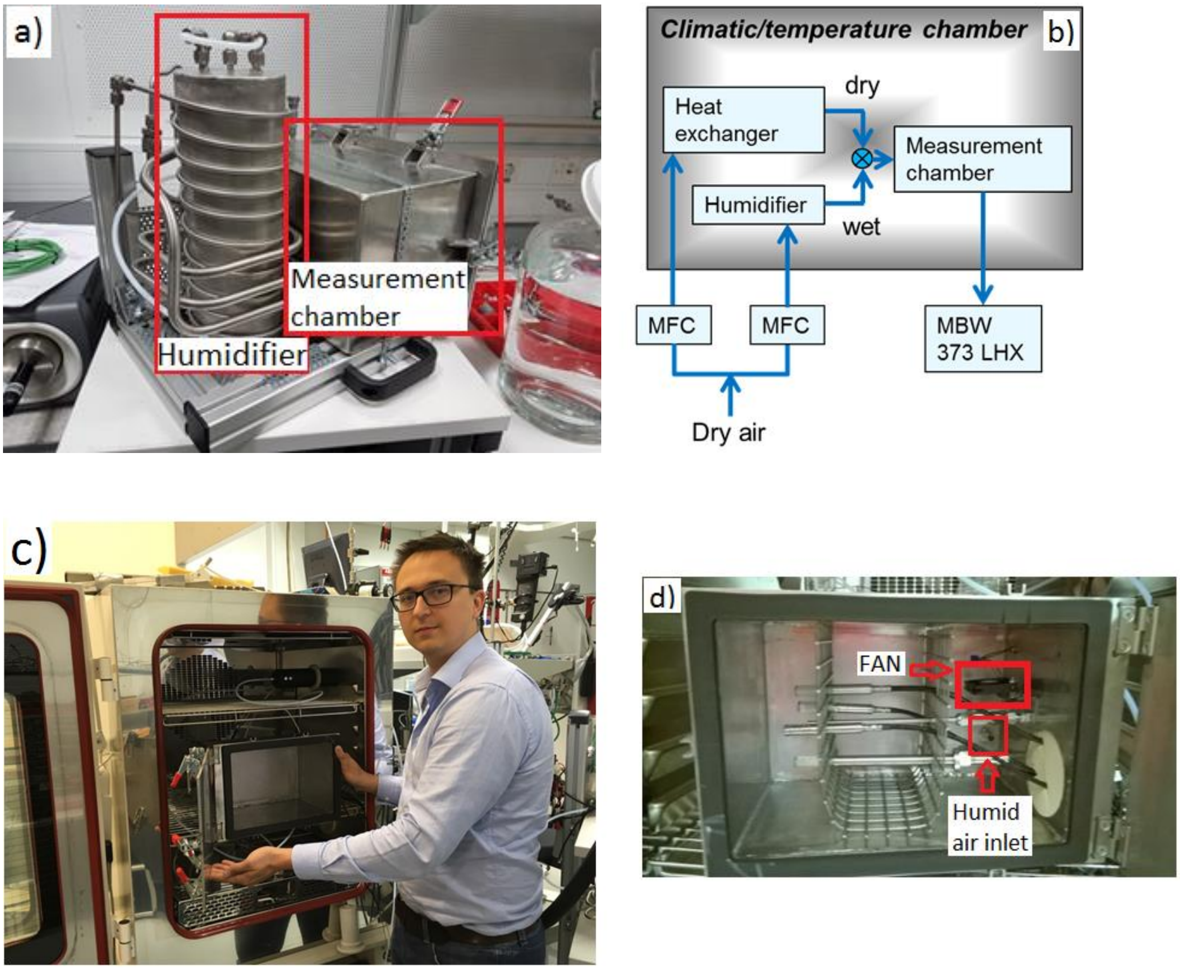


Figure 7, VTT measurement setup. a) Humidifier and measurement chamber. b) Schematic of the humidity generation principle. c) The modular system in figure a) inside a climatic chamber. d) Humidity sensors fitted inside the measurement chamber.

A calibration method for relative humidity hygrometers based on a non-static calibration method was developed. The method provides equivalent calibration results compared to the typical method based on calibrations at stable humidity points. The main advantage of this new method is that the calibration can be performed faster and more data on the sensor behaviour can be attained than with the conventional point-wise calibration. For example, a typical 5 point static calibration with a 1 h stabilisation time at each calibration point, will take up to 10 hours (each point is measured twice) compared to a dynamic calibration of 6 hours (two 1 h static points and two 2 h ramps). In the case of more than five measurement points, the benefit is even larger.

Three prototypes of a field humidity calibrator, based on dynamic humidity control and with an operating range of 10 %rh to 90 %rh, were developed: a large volume calibrator, a small volume calibrator with manual operation, and a small volume calibrator with computer controlled operation.

The large volume calibrator includes unique features, e.g. arbitrary RH/T calibration profile. Unfortunately, there was a malfunction in the system that could not be repaired during this project, and the calibrator could not be applied in demonstrations.

Figure 8. Large volume calibrator

Two prototypes of the small volume calibrators were set up: the first one with manual operation, and the second one with computer controlled operation. The size of the calibrator is 30 cm x 25 cm x 15 cm (Figure 8). It consists of a water chamber, a desiccant chamber, two fans and a measurement chamber. The air flow through the water and desiccant chamber is controlled with fans. The desired humidity level entering the measurement chamber is obtained by mixing dry and moist air in a specific ratio. This ratio can be controlled by controlling the individual fan speeds. Another fan was placed inside the measurement chamber to ensure sufficient air speed (> 0.2 m/s). In order to ensure sufficient humidification at the relatively high flow rates, a resistive heating wire was applied for heating the water.

Figure 9. Field calibrator prototype developed at VTT

The relative humidity is controlled with a Labview-program making use of PID-algorithms and reference sensor readings. In this way humidity can be controlled within ±0.5 %rh. The prototype was validated by comparison with a CETIAT humidity calibration setup that uses humidity sensors as transfer standards. The prototype was also tested with a sensor provided by Vaisala. Vaisala also provided input in the further development of the prototype.

Besides humidity control, different kinds of calibration cycles can be chosen. If the reference sensor has a fast response time, these excitation schemes can be used to find the response times of the device under test. The current prototype is only operating at ambient temperature, which is sufficient for most humidity calibrations on site.

The outcomes of Objective 2 were:

- A new calibration scheme with an appropriate calculation method was developed to provide an efficient approach to perform humidity calibration at several temperatures, in the temperature range from 100 °C to 180 °C. Another scheme based on linear humidity ramps was designed for calibrations at a single temperature. These calibration schemes save 40 % of the calibration time compared with a typical calibration scheme. The schemes were validated with a static method, and the agreement was good.
- A modular calibration setup was developed to provide a practical approach for carrying out humidity calibrations at non-static conditions in commercial temperature/climatic test chambers. Six different capacitive humidity sensors were tested with different calibration scheme parameters to analyse the effect of non-static conditions on calibration results.
- Three prototypes of a field humidity calibrator were designed and tested.
- The outcomes of the developments were reviewed, and recommendations for humidity calibrations in non-static conditions were prepared by VSL, CETIAT, INRIM, PTB, VTT, IH, Michell, TU-DA and Vaisala. The procedures developed were used as input to a draft guideline “A practical approach for estimating the uncertainty of humidity measurements at high temperatures (up to 180 °C) and under transient conditions in industry”.
- The typical calibration time saving obtained was 40 % instead of the original aim of at least 50 %, but otherwise this objective was achieved.

- **Objective 3: To develop humidity measurement techniques and procedures for the accurate monitoring of temporal and spatial humidity variations in selected applications, including development of a new type of hygrometer based on direct Tuneable Diode Laser Absorption Spectroscopy (dTDLAS) for process environments with temperatures up to 180 °C and a new measurement method for detecting the influence of microbiological processes on the transient humidity conditions within small samples. The target relative uncertainty for the water vapour amount fraction at high temperatures is 5 %, and the uncertainty for the relative humidity measurements will be less than 2 %rh.**

The dTDLAS spectrometer developed by TU-DA was validated with the PTB humidity characterisation facility (see Objective 1) in the temperature range 40 ^{0} C - 100 ^{0} C, and at INRIM in the temperature range 100 ^{0}C - 180 ^{0}C.

Based on the first validation, the dTDLAS hygrometer could subsequently be applied at an industrial site within the project (Objective 5). The industrial validation was carried out in GBV’s tunnel drying and roasting oven facility. The industrial validation of the dTDLAS was done with the DTI and UNICLAM/INRIM water activity and equilibrium moisture content measurement facilities.

However, the first validation trial in the temperature range 100 ^{0}C - 180 ^{0}C at INRIM showed larger deviations than the targeted 5 % at higher pressures. The reason was tracked to a possible mismatch of the applied fitting approach and the spectral line data in the HITRAN database: Line data parameters reported in HITRAN are valid for gas mixtures of the measured gas in air and typical temperatures in between -50 °C and +50 °C. During the experiments at INRIM the mole fraction and the pressure were increased to a level were the combination of those parameters in the fitting, i.e. in particular self- and air pressure broadening coefficients and its temperature dependence above 50 °C, produced a bias.

A method to measure local gradients of 1 %rh/cm was developed by UL. The method was demonstrated with an experimental setup of a stack of 6 Petri dishes. The goal was to measure unwanted high humidity occurrences in transient conditions within sterile Petri dish samples stacked in a large climatic sterile room. The system was used to perform root cause analysis of the influence of microbiological processes to the transient humidity conditions within the samples.

In Figure 10, temperature and humidity probes are placed in individual Petri dishes and between them. When performing such a measurement, the thermal influence of the measurement system should be minimised and temperature and humidity probes with a small heat mass should be applied.

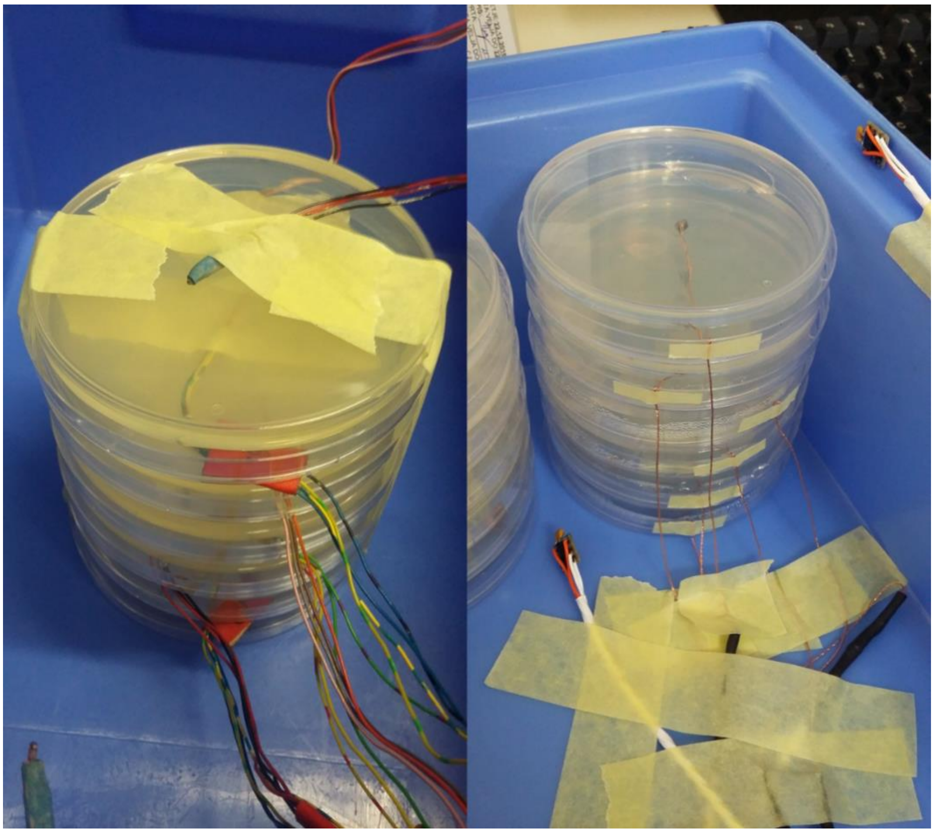


Figure 10. Temperature probes in Petri dishes

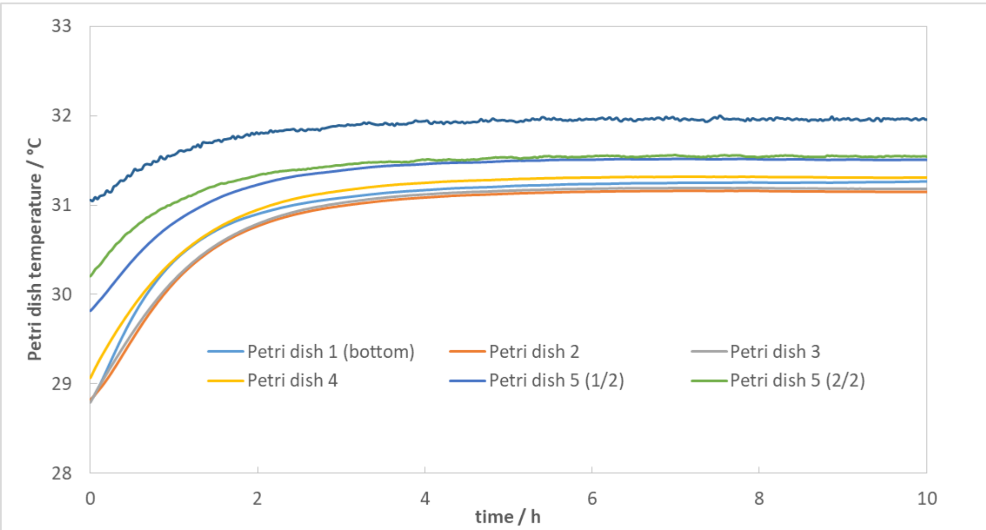


Figure 11. Temperature transitions inside individual Petri dishes

The rate of condensation growth depends on the temperature and humidity gradients. As the temperatures inside individual Petri dishes rise during a transition, temperature uniformity improves, which means that the gradients decrease. Figure 11 shows the decrease in (standard) deviation of the temperatures in the batch of Petri dishes.

In this case, the temperature deviation between Petri dishes finally stabilised at 0.28 °C, which corresponds to a relative humidity deviation of approx. 1.6 % rh. Taking into account the 95 % probability with coverage factor *k*=2, the expanded uncertainty due to the uniformity of the batch of 6 Petri dishes, which is used as a target for profile measurements amounts to 0.54 °C and 3.2 % rh, respectively.

In order to analyse the behaviour and to estimate the measurement uncertainties of impedance humidity sensors, the effects of direct thermal irradiation, response times, heating/purging features and responses to cyclic and stepwise temperature and humidity changes were studied at VTT, FORCE and Vaisala. Different types of sensors from different manufacturers were studied. All of the studied sensors were shown to be sensitive to transient conditions, causing deviation in their displayed values. This applies to both temperature and humidity readings. Cyclic exposure and transients may also cause permanent changes in the sensor readings. Response times are taken into account so that sensors typically require time until they have reached their stable and correct value. Sensors from different manufacturers and with different functionalities perform differently. Therefore, when exposed to high temperatures and /or temperature or humidity transients, it is important to characterise the behaviour of the sensor and/or the measurement target for vulnerability to saturation and oscillation, response time, thermal and humidity gradients, and thermal radiation. The observations made on the uncertainty contributions in non-static conditions also apply to Objective 4.

Based on the observations, a draft guideline “A practical approach for estimating the uncertainty of humidity measurements at high temperatures (up to 180 °C) and under transient conditions in industry” was prepared. It will be developed to a EURAMET guide.

Figure 12. Set-up for studying the effect of direct thermal irradiation on the humidity sensors

The outcomes of Objective 3 were:

- An experimental setup with precision sensors was developed and tested for the analysis of unwanted high humidity occurrences in transient conditions within sterile Petri dish samples.
- The behaviour of impedance humidity sensors, as well as the estimation of the measurement uncertainties due direct thermal irradiation, the response times of the sensors, the heating/purging feature and the response to cyclic and stepwise temperature and humidity changes, were studied by VTT, FORCE and Vaisala. Based on these results, a draft guideline “A practical approach for estimating the uncertainty of humidity measurements at high temperatures (up to 180 °C) and under transient conditions in industry” was prepared for publication as a EURAMET guide.
- A new type of hygrometer based on direct Tuneable Diode Laser Absorption Spectroscopy (dTDLAS) was developed for process environments with temperatures up to 180 °C. Validation measurements were successfully performed at ambient pressure and temperatures between 40°C to 100°C, however the first validation trial in the temperature range 100 ^{0} C to 180 ^{0} C showed a larger deviation than the targeted 5 % at higher pressures. The reason was a mismatch of the line data and the fitting approach that was used.
- The target uncertainty of dTDLAS, 5 %, was not met during the first validation trial in the temperature range above 100 ^{0} C, but otherwise this objective was achieved.

- **Objective 4: To develop water activity measurement (equilibrium relative humidity) techniques for in-line measurement applications - with a measurement uncertainty smaller than 0.02 - and to develop methods for establishing the traceability link between water activity and water mass fraction measurements including the development of tools for analysing error sources in water activity measurements which are an integral part of sorption isotherm measurements.**

The water activity (aw) is a measure of how much free water there is available for chemical reactions and for the growth of micro-organisms such as bacteria and fungi. Food and feed manufacturers use the water activity to control the shelf-life or texture of their products. Within the HIT project, a new in-line water activity measurement method was developed. The tests were carried out in GBV's tunnel drying and roasting oven facility. DTI (Denmark) completed and validated an experimental setup for the SI traceable water activity measurements. The validation was carried out in comparison with the gravimetric steady-state equilibrium moisture content determination by UNICLAM and INRIM, using saturated salt solutions. As a result, the water content of a 500 g sample containing 10 % water can be measured with 0.25 % uncertainty. A sample having 80 % water activity can be measured with an uncertainty of less than 1 % uncertainty.

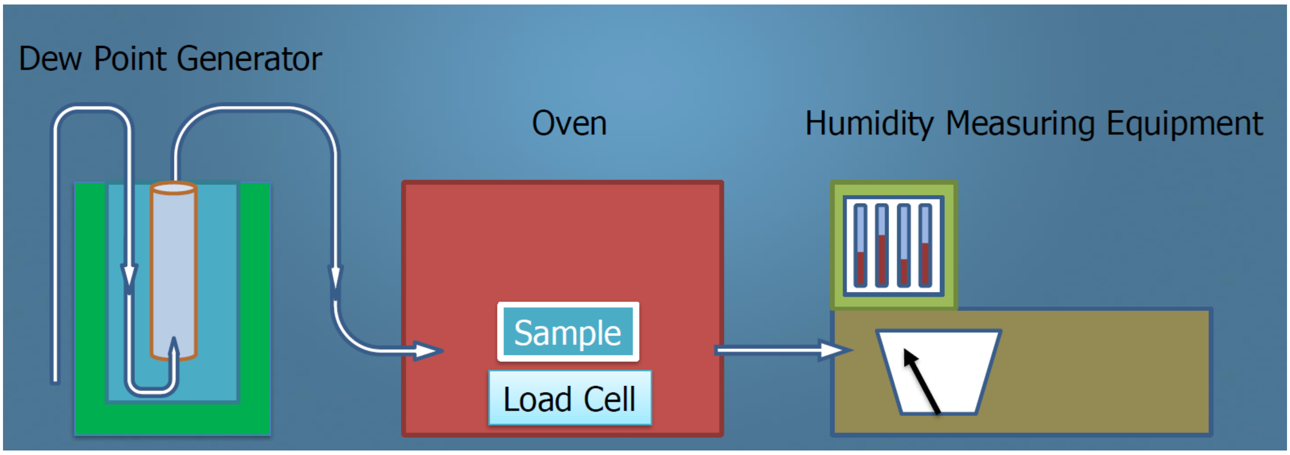


Figure 13. DTI water activity measurement setup

Figure 14. Hazelnut sample in a sample cell of the DTI setup

UNICLAM developed a one-dimensional numerical transient tool for modelling the temperature dependence of sorption isotherms and transient water activity in hazelnuts. The tool was practically realised with Visual Basic for Applications (VBA) of Microsoft Excel, and it offers a practical and simple tool to study the transient water accumulation in hazelnuts until the thermodynamic equilibrium conditions are reached. It consists of several spreadsheets used for input data, numerical calculations, postprocessing and uncertainty budged determination.

The numerical transient tool was validated with experimental data from the DTI and UNICLAM setups, and it was applied to predict water activity. The uncertainty of the simulation results derives from the uncertainty of the experimental input parameters. Therefore, the uncertainty methodology and budgets of DTI and UNICLAM were applied in the numerical tool development. There was good agreement with the water activity data in the scientific literature. A journal article describing the model was prepared and submitted to Journal of Food Engineering.

Using the outcomes from the techniques for in-line water activity, UNICLAM performed a sensitivity analysis giving information about the accuracy of the developed numerical model. The tool was applied in water activity prediction in transient conditions, and an uncertainty confidence interval was associated to it.

The developed method establishes the traceability link between water activity and water mass fraction measurements through sorption isotherm measurements. Material dependent modifications are needed when applying the software to other materials.

The developed methods were applied in industrial sites (see Objective 5). Measurements were carried out for clients in pet food manufacturing and drying/roasting determination of the adsorption isotherms in the whole hazelnut drying and roasting process. CNR assisted in applying the developed methods and tool in a food-industry use case. The measurements were carried out at different temperatures, in steady state and in transient conditions. In addition, the dTDLAS developed by TU-DA and PTB in Objective 3 was also applied in these in-line measurements.

T**he outcomes of Objective 4 were:**

- A new SI traceable in-line water activity measurement method was developed and validated.
- A sample-based system for sorption isotherm measurement was developed, tested and launched as a service.
- A software tool for modelling the temperature dependence of sorption isotherms and transient water activity in hazelnuts was developed.
- The outcomes of this objective were also reviewed by VTT, FORCE and Vaisala, to be included in the EURAMET draft guideline “A practical approach for estimating the uncertainty of humidity measurements at high temperatures (up to 180 °C) and under transient conditions in industry”.
- This objective was fully achieved.

- **Objective 5: To validate all of the methods developed in this project through demonstration in selected industrial applications in order to facilitate the take up of the technology and measurement infrastructure developed by the project and to recommend what further actions are necessary to ensure the uptake.**

An IH acoustic humidity sensor calibrated and characterised with the VSL facility developed in this project up to a temperature of 120 °C, pressures up to 500 kPa and dew-point temperatures up to 117 °C was successfully demonstrated in a dairy manufacturing process in the Netherlands. Commercial baby milk powder is produced in large spray dryers. The capacity and the efficiency of the dryer depends on the moisture uptake by the air. Therefore, it is necessary to have reliable measurements of the air flow and the humidity.

Figure 15. HumiFlow and spray dryer

The methods for humidity calibrations and measurements at high temperatures and under transient conditions, developed at INRIM, GBV and DTI, were applied to characterise GBV's tunnel drying and roasting oven facility, which was developed for processing whole hazelnuts. CNR performed chemical and microbiological profile measurements on raw and processed foods produced in GBV's tunnel drying and roasting oven facility. At the final stage a correlation between the food characteristics and the process humidity-temperature profile and with the final product quality was estimated. TU-DA's dTDLAS hygrometer was characterised with the PTB humidity characterisation facility (described in Objective 3), and applied to demonstrate traceable transient high-temperature humidity measurements at different locations in the hazelnut drying / roasting process.

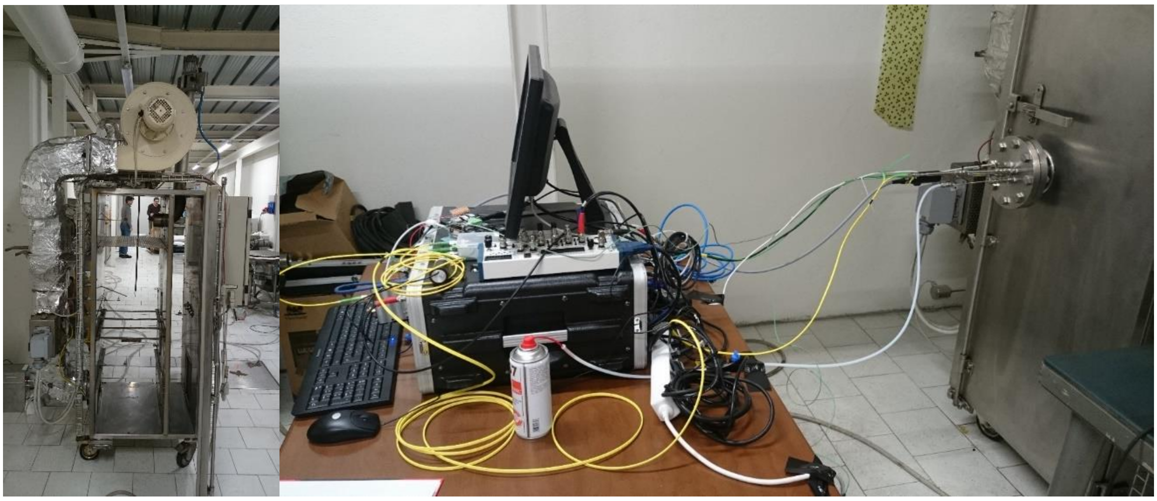


Figure 16.d TDLAS measurement setup at GBV

In order to obtain a good shelf-life, for roasted hazelnuts, the residual water content should not exceed 5 %. The goals were to demonstrate the possibility to control the water loss of a batch of hazelnuts using dew-point real-time measurements during a roasting process, to characterise the different roasting procedures by means of a metrologically-sound temperature and humidity control approach, and to correlate humidity/air-temperature measurements and the quality of processed hazelnuts. Good reproducibility was found between drying process repetitions, which allowed us to model the drying profile at different oven temperatures and heating methods. Capacitive relative humidity probes were found to be unsuitable for real time measurement due their low resolution, but they were suitable for measurements with steam introduction during the process. A good correlation was found between the hazelnuts' water loss after roasting and real time water loss measurements during roasting. Preliminary results on the correlation between humidity/temperature and roasted hazelnut quality are promising. Novel evidence on the link between humidity/temperature during roasting and the level of hazelnut allergenicity were found.

The methods for humidity calibrations and measurements at high temperatures have been applied by CETIAT to the humidity probes used in the food industry for measuring low humidity at high temperature. The aim was to demonstrate the benefits of the sensor calibration at high temperatures using the new methods developed in the project. CETIAT had the opportunity to collaborate with CTCPA (a technical centre working on the baking processes that are applied to cookies) as well as with CENELEC's working group CLC/TC/59X/WG18 "Electric ovens for commercial use", which is working on a project for a new European standard for energy efficiency in professional ovens with a steam mode. One instrument that could be used for measuring temperature and humidity during the process is equipped with temperature sensors, thermocouples, and a zirconium oxide humidity sensor (lambda sensor).

In cooperation with a collaborator, Orion Oyj, VTT tested the field humidity calibrator developed in a pharmaceutical plant. In the tests, humidity sensors were calibrated at room temperature from 20 %rh to 70 %rh using the field humidity calibrator. The prototype calibrator with two Vaisala sensors (HMP233, HMT330) was operated with different measurement schemes: single calibration with ascending ramp, and at arbitrary steps. The stability of the device (±0.3 %rh at static calibration points in the range from 20 % to 70 % rh) was found to be sufficient, and the dynamic calibration approach useful, at least in some applications. With the current approach, a 30 % to 50 % reduction in the overall calibration time could be expected. The demonstrated prototype operated only at ambient temperature. If temperature calibration options were added, the reduction is expected to be even larger. The potentiality of the calibrator was also discussed with Vaisala.

Figure 17. Demonstration of the field humidity calibrator at Orion

A method to measure local gradients of 1 %rh/cm developed by UL and described in Objective 3, was demonstrated at a client's site.

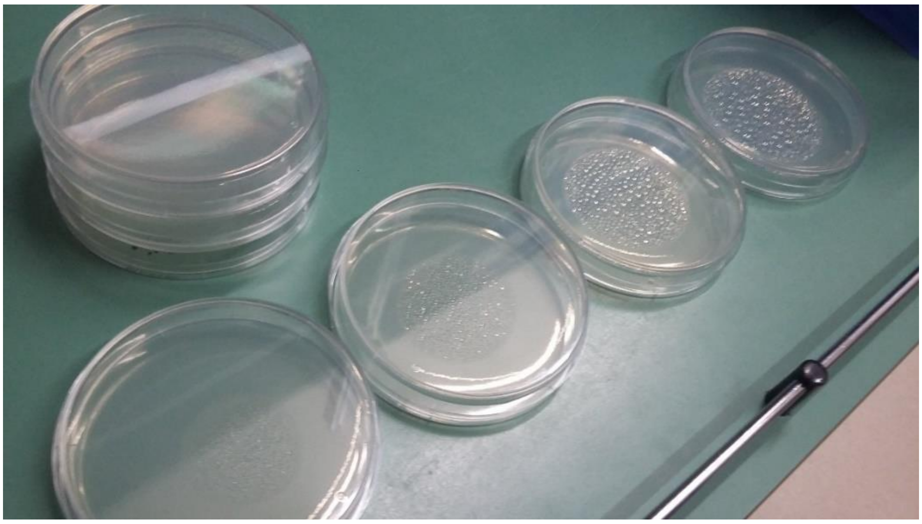


Figure 18. Condensation on the inner wall of the Petri dishes

VSL and Michell carried out a demonstration in a papermaking company in The Netherlands. The demonstration was related to the importance of calibrated humidity measurements during the drying process, which in turn reduces the energy consumption and improves safety. In the demonstration, the new, calibrated polymer sensors were compared with the existing humidity measurement system of the factory.

The purpose of the demonstration was to make sure the energetic dry hood is in balance. The output ventilator adjusts how much hot air is sucked out of the hood. When too much air is sucked out, the pressure under the hood decreases, with the negative effect of cold air from the environment (production area) being sucked in. The burner is then switched on to heat up the air, which causes loss of energy. This is measured and a low value of gH2O /kg of air can be seen at that time. When not enough hot air is sucked out, the pressure under the hood increases, and warm air blows into the environment, causing energy loss and danger of fire. This can be seen when a high value of gH2O /kg is measured. Also, a too high value of gH2O /kg, drying of the product/process reduces and condensation occurs inside the hood. The balance is demonstrated with humidity measurement. When the hood is in balance, a drying optimum is reached with as little as possible cold air being sucked in, which turns to energy saving. In practice the adjustments are not always optimal, so there are moments when the hood does not dry to a 100 % optimum.

The existing calibration process of the sensors did not correspond to the conditions of actual use, and it was not traceable to SI units. This test demonstrated that the polymer sensing elements are fit for the measurement (after the heat exchanger), they are cheaper than the existing method, and they are available with SI traceable calibration certificates.

The outcomes of Objective 5 were:

- The outcomes of the project were successfully demonstrated in 6 industrial applications.
    - An acoustic humidity sensor calibrated and characterised up to 120 °C, at pressures up to 500 kPa and at dew-point temperatures up to 117 °C, was successfully demonstrated in a dairy manufacturing process in the Netherlands.
    - The water activity measurement was demonstrated in a) pet-food manufacture and in b) whole hazelnut drying process monitoring. The measurement of water activity during drying and roasting improves the product quality and thus their shelf life.
    - The field calibrator was demonstrated at a pharmaceutical plant to calibrate their humidity sensors.
    - The importance of sensor calibration at high temperatures using the new methods developed in the project was demonstrated in biscuit production.
    - Applicability of a new sensor type and the importance of calibration were demonstrated in a papermaking company.
- This objective was fully achieved.

5 Impact

This project delivered new and improved methods and techniques for humidity measurement and monitoring at high temperatures and under transient conditions, enabling a wide range of industrial enterprises in EU to enhance their competitiveness through reduced energy consumption and waste production and through more efficient and reliable quality assurance and new products. The outcomes are available to be exploited by the industrial partners, manufacturing industry, industrial test and calibration service providers, instrument manufacturers and the wider stakeholder community. To assist the industrial and scientific communities, new SI traceable measurement and calibration services in high dew-point and air temperatures as well as in sorption isotherm and water activity determination have been launched.

The project has already delivered 3 articles published in scientific journals, 11 presentations in conferences, 13 training events and workshops, and 33 presentations and articles in various meetings and trade/professional magazines. 7 abstracts have been submitted to TEMPMEKO 2019 conference, and 6 of them will be published as peer-reviewed scientific articles after the conference. One article for a peer-reviewed scientific journal has been submitted for review, and another is being drafted phase. The project partners have liaised with standardisation bodies or the equivalent and demonstrated their work at 16 meetings. Fourteen companies have invited the partners to demonstrate the project's developments.

The partners communicated actively with metrological communities through participation in three metrology working groups at European level and two others at global level representing both physical and chemical metrology. Presentations were given in the conferences CIM 2015 and 2017, TEMPMEKO 2016, THERMACOMP 2016, ICHMT 2017, ICWPS 2018 and submitted to TEMPMEKO 2019. The project has 6 collaborators. In ResearchGate, this project has 27 followers and 375 reads.

Impact on industrial and other user communities

A wide range of industrial enterprises in the EU will benefit from the outcomes of this project through improved humidity control in drying, baking, storage and testing and through more efficient calibration methods. As a result, the enterprises will be able to improve their productivity through reduced energy consumption and waste production and through more efficient testing and calibration.

The project had significant direct impact on the application of humidity measurements in industrial process and quality control through new humidity calibration methods and procedures, the new type of transfer standard hygrometer and a field humidity calibrator, new measurement and uncertainty estimation methods in applications with significant temporal and spatial humidity variations and the novel in-line water activity measurement techniques.

The new type of field humidity calibrator developed in the project significantly reduced the time needed for humidity calibrations on-site by 40 % or more, thus reducing costs and downtime for instruments. Extending calibrations to dynamic humidity measurements will reduce the calibration time, e.g. in pharmaceutical companies and in humidity sensor manufacturing and will improve humidity control e.g. in environmental testing. The field calibrator has been demonstrated to industrial partners. The feedback obtained will be exploited in its future commercialisation attempts.

New services for traceably determining sorption isotherms and water activity at temperatures up to 70 °C have been launched. This supports the food and feed industry in their product quality assurance in a cost-effective manner.

Active communication with industrial partners, collaborators and other stakeholders has been maintained to ensure efficient two-way exchange of information. This includes face-to-face meetings with 14 companies, contributions to 11 training events, 9 conference presentations in 7 conferences and 17 workshops/seminars, 4 articles in professional magazines, a LinkedIn group, project website (www.empir-hit.eu) and e-Newsletter. The partners have extended their measurement, consultation and training services for industry in the fields of high temperature humidity measurements and water activity measurements.

Impact on the metrology and scientific communities

The high-temperature mass fraction water vapour/steam generator and two new calibration/testing facilities developed in this project have extended the reference humidity measurement capabilities of European NMIs to dew-point temperatures up to 150 °C, air temperatures up to 180 °C and air pressures up to 6 bar (abs). One of the facilities has already launched the service, while two others are progressing towards it. This enhances the wider availability of NMI level traceability services (i.e. calibration of reference instruments and inter-laboratory comparisons) which are vital in enabling industrial calibration laboratories.

By applying the calibration methods developed in this project, other NMIs/DIs will also be able to efficiently extend their capabilities according to national needs to cover humidity calibrations at temperatures above 100 °C, as well as dynamic humidity measurements. The outcomes of the work on relative humidity calibrations above 100 °C underpins the task of CCT WG Humidity related to internationally harmonised humidity terms and definitions. The project provides direct input to EURAMET in the form of recommendations on humidity calibration procedures for high temperatures and under transient conditions, and the draft EURAMET guide on the Calibration of humidity measuring instruments.

As a result of this project, the customer offering at the NMI level has been extended to water activity. New services for SI traceable determination of sorption isotherms and water activity at temperatures up to 70 °C have been launched. This development, combined with the numerical tools in estimating uncertainty in water activity measurements and the determination of sorption isotherms, enables food scientists to improve the characterisations of raw and processed food materials.

Impact on relevant standards

The partners contributed actively to the work of several CEN, IEC, ISO, AFNOR, and DIN standardisation groups together with metrology committees. The most authoritative guidance documents related to calibration and traceability are prepared and published by the Joint Committee for Guide in Metrology (JCGM) (which includes BIPM, IEC, IFCC, ILAC, ISO, IUPAC, IUPAP and OIML) and regional metrology organisations, such as EURAMET e.V. in Europe. The quality assurance of measurements is often evaluated in certification (ISO9001:2000; GMP etc.) and accreditation (ISO17025:2017) in Europe using the requirements set in the JCGM and EURAMET guidance documents. Until now there has been no European standard or guideline on humidity calibrations available for industry. This project developed a draft version of a EURAMET Guide cg NN “Calibration of humidity measuring instruments” based on the advances in the project in consultation with EURAMET TC-T WG Best Practice.

The consortium has been contributing to two standardisation groups of DIN and ISO and to five metrology working groups of EURAMET and CIPM. Experiments have been carried out and results analysed for formulating recommendations to EURAMET and CIPM and the draft EURAMET guide. The recommendations are about applying the new calibration methods developed in this project.

Longer-term economic, social and environmental impacts

Due to improved humidity calibration methods for temperatures above 100 °C and for non-static conditions, the calibrations can be carried out in conditions that correspond to the actual use of the humidity sensors. More accurate measurements improve product quality in food, pharmaceutical and paper manufacturing, which results in reduced amounts of waste. On the other hand, energy savings can be expected, as the products will no longer be over-dried. Faster and more automatic calibration schemes reduce the downtime of the devices under calibration, but they also make the calibration work more economically sustainable for the accredited laboratories. In-line measurements of water activity applied in the food and feed industry will improve consumer safety by improved product safety and prolonged shelf-life of the food or feed products - in a cost-effective manner.

6 List of publications

1. N. Massarotti et al., New benchmark solutions for transient natural convection in partially porous annuli, International Journal of Numerical Methods for Heat & Fluid Flow, Vol. 26 No. 3/4, 2016 pp. 1187-1285. https://doi.org/10.1108/HFF-11-2015-0464

2. F. Arpino, G. Cortellessa, N. Massarotti, M, Scungio, Two-phase explicit CBS procedure for compressible viscous flow transport in porous materials, Journal of Numerical Methods for Heat & Fluid Flow, Vol. 28 Issue: 2, pp.336-360. https://doi.org/10.1108/HFF-02-2017-0080

3. R. Bosma, R.J. Pouw, W. van Schaik and A. Peruzzi, Climatic chamber for dew-point temperatures up to 150 ^{0} C, Metrologia 55 (2018) 597-608. https://doi.org/10.1088/1681-7575/aacecc

7 Website address and contact details

The project website is: www.empir-hit.eu

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