Final Publishable JRP Summary for IND12 Vacuum
Vacuum metrology for production environments

Overview

Computer chips, flat-panel displays, mobile phones and solar cells all rely on production steps carried out in a vacuum environment, with the semiconductor industry alone making up about 40% of the vacuum market. Vacuum is also an important and indispensable tool for many other industrial processes such as lighting, pharmaceutical, coating and food packaging industries. This project has addressed the need of industry to reliably characterise both fast changing vacuum pressures and partial pressures of either desired or unwanted (outgassing) gas species, and also the need to facilitate the reliable measurement of leak rates. The project has enabled vacuum measurement gauges to be compared against a standard where the pressure is varied from 100 kPa to 100 Pa (or vice versa) within 20 ms or 1 s and partial pressure and outgassing rate measurements can now be made traceable for the first time. By applying new technologies and hydrodynamic approximations it has been possible to provide new leak standards with predictable leak rates for many gas species applicable to typical industrial environments.

Need for the project

Vacuum technology has developed and matured a lot over the last 50 years, but there are still important challenges to overcome for industrial applications in order to ensure the reliability and quality of vacuum processed products and to improve the cost efficiency of vacuum processes. Industry often requires a complete characterisation of the vacuum; including the total pressure, knowledge of the gas species in the vacuum (partial pressures) and the time-dependent changes of the partial and the total pressures.

Prior to the project existing vacuum measurement standards provided traceability from 10^{-9} Pa to 10^{5} Pa for pure gases under equilibrium conditions. Industrial processes, however, very rarely work with pure gases (e.g. physical and chemical vapour deposition for electronics manufacture or coatings for hardening tools etc.) and often take place in non-equilibrium environments (e.g. physical and chemical deposition for optical disc manufacture), where the pressure is changing dynamically. These fast vacuum processes require reliable vacuum gauge data during cycling times lasting a couple of seconds.

The “cleanliness” of the vacuum – or more technically the absence (or sufficiently low partial pressure) of specific gas species or vapours – is also vital. Lack of control of the partial pressures of certain gas species (for example water) may result in unwanted oxidisation of surfaces and the creation of inappropriate insulating layers. In-situ methods for the calibration of quadrupole mass spectrometers (QMS), the main instrument used to measure partial pressures, and other vacuum gauges in a real environment are also required.

Although oil-free vacuum pumps and other components have improved in recent years, in terms of cleanliness, components in particular can still be a source of unwanted gases that get released or “outgassed” into the vacuum. Outgassing rates are therefore a vital quality-assurance figure in vacuum technology, but historically there has been no established way to measure such rates and the results obtained have not been traceable to the SI system of units.

Leak detection is crucial to ensure the safety and reliability of vacuum systems and products. For example vacuum leak detection prevents refrigerants in air-conditioning systems or toxic, radioactive and environment polluting substances in containment systems from escaping and industry requires faster and cheaper leak detection methods in this and other areas. Nowadays, the most sensitive, versatile and accurate method for leak detection is either to flush the outside of the system with helium and then measure how much of the helium leaks into the evacuated system or alternatively to pressurise the system with helium and measure

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how much helium leaks out of it. However, in industrial applications leaks usually occur under different pressures, temperatures and temperature gradients, gas species and mixtures or can even involve liquid flows through the leak instead of gas. Thus, research was needed to provide industry with traceability for leak measurements under real industrial conditions.

Scientific and technical objectives

The key aims of the project were therefore to:

- Develop a novel vacuum calibration facility for dynamically changing pressures including
  - Development of a vacuum system capable of producing very fast pressure changes over three orders of magnitude from 100 kPa to 100 Pa and vice versa within a second or less
  - Development of vacuum gauges with very fast response times of a few ms
  - Evaluation of the feasibility of optical methods for the detection of fast pressure changes
  - Provision of data on the relaxation time of vacuum gauges for fast pressure changes
- Provide traceability for partial pressure measurements in industry by
  - Identification of the key parameters needed for the calibration of QMS
  - Provision of information on the metrological characteristics of industrial QMS
  - Establishment of a measurement standard to calibrate QMS from 10^{-8} Pa to 10^{-2} Pa
  - Development of a draft for a technical specification for the calibration of QMS (for consideration by ISO TC 112 Vacuum Technology)
  - Provision of methods to compare results obtained with QMS from different manufacturers
  - Evaluation of the feasibility of using optical methods as an alternative for partial pressure measurement
- Provide traceability for industrial outgassing rate measurements and material characterisation by
  - Development and testing of an industrially applicable method for outgassing rate measurement from 10^{-8} Pa L/s to 10^{-1} Pa L/s
  - Provision of reference materials for the validation of outgassing rate measurements in industrial environments
  - Provision of information on the comparability of different outgassing methods in terms of reliability, accuracy, and practicability for industry
  - Development of a draft for a technical specification for a traceable outgassing rate method (for consideration by ISO TC 112 Vacuum Technology)
- Improve capabilities for leak measurement and testing in industrial environments by:
  - Development of methods better suited to leak measurements under realistic industrial conditions
  - Specification of alternative methods to mass spectrometer leak detectors in order to detect leaks within a second or less (dynamic measurements)
  - Production of a practical guide for industry on the metrological performance of commercial leak detectors

Results

Novel dynamic vacuum calibration facility

A dynamic vacuum calibration facility was established which generates predictable pressure changes from 100 kPa to 100 Pa in less than one second by expansion of gas from a small volume into a large volume through exchangeable orifices or ducts. A fast opening gate valve that opens to its full diameter of 40 mm within 4.6 ms initiates the gas expansion and was specially manufactured for this purpose.

The fastest pressure reduction possible in the facility is a decrease in pressure from 100 kPa to 100 Pa within 23 ms, when the full opening between the two expansion volumes is used, and this exceeds the most demanding current requirements of industry. The pressure reduction can be made to occur over a longer period if nozzles or ducts are installed between the small volume and the fast opening valve. If the orifice is

used the reduction is reached within 600 ms, and if two different Laval nozzles are used the corresponding times are 450 ms and 900 ms.

The most accurate pressure decrease is achieved when the fast response vacuum gauges developed by the project partner INFICON AG are used, in which case the pressure can be determined during the pressure reduction with an uncertainty of 5 %. By changing the orientation of the fast opening valve, it is also possible to produce very fast pressure rises from high vacuum to 100 kPa.

Using this facility, the relaxation or response time of vacuum gauges was tested for fast increases or decreases of pressure, which was not previously possible. The response times of the vacuum gauges developed by INFICON AG were less than 1.3 ms.

Using the new fast gauges as secondary standards it was also possible to verify that optical methods measuring refractive index changes are suitable to measure fast pressure changes.

Provision of traceability for partial pressure measurement in industry

A survey of end users undertaken by the project identified leak monitoring, leak rate measurements and outgassing rate measurements as the main industrial applications of quadrupole mass spectrometers (QMS) requiring more reliable and improved measurements and accuracy.

A primary standard for partial pressures suitable for characterising and calibrating quadrupole mass spectrometers was established at PTB and validated. It also includes a module to traceably measure outgassing rates. Using this primary standard, partial pressures of three gas species can be generated simultaneously in the range from $2x10^{-9}$ Pa up to $1x10^{-2}$ Pa, with uncertainties of typically a few percent. The partial pressures are generated by gas flow from a reservoir through newly developed leaks into the main calibration chamber where the quadrupole mass spectrometers are installed, the gases then exit this chamber via an orifice of known conductance. The new leak components developed in the project are based on ducts that enable molecular flow to be achieved for pressures up to 1 kPa and higher, which greatly simplifies the gas dynamics and hence the determination of gas flow for any gas species.

A two year long term study to investigate the performance of nine different commercial quadrupole mass spectrometers and to identify the key factors / parameters affecting their performance was undertaken. The results showed that the adjustable operational parameters of the instruments have quite different influences on the instrument's metrological behaviour. While the mass scale parameters were reasonably stable, the stability of the sensitivity exhibited typical changes of 60 % over a 3 month period. Today's quadrupole mass spectrometers therefore require clear standardised procedures for their calibration and characterisation, and additional in-situ calibration is also required to achieve the best results. A draft Technical Specification for the calibration of QMS was therefore developed by the project for consideration by ISO TC112 Vacuum Technology. This draft includes the methods to compare results obtained with different QMS.

The feasibility study of optical methods highlighted that these methods provide more reliable results than those obtained with QMS, but are efficient only at pressures > 0.1 Pa and have the disadvantage that the gas species must be known before the measurement.

Provision of traceability of industrial outgassing rate measurement and material characterisation

Three outgassing rate reference systems utilising different methods were developed and validated; two of them use primary techniques to determine the partial outgassing rate i.e. with independent traceability directly to SI system of units. PTB's system uses the continuous expansion method, IMT's system uses gas accumulation and Karlsruher Institut für Technologie's system uses a modified version of the throughput method where a difference of gas flux from two chambers (one empty and one containing the sample) is measured. The different methods, including an industrial system, were compared in terms of reliability, accuracy, and practicability for industry using reference samples.

Three types of reference outgassing artefacts were developed and assessed: reference outgassing materials (Viton (FPM-fluoropolymer) and PDMS (poly-dimethyl siloxane), which were identified as having sufficient repeatability and reversibility of gas absorption and gas release), a permeation reference outgassing artefact and an in-situ leak artefact based on nanoholes in a thin SiN membrane. A patent application for the easy to handle permeation type outgassing artefact for water vapour, gas mixtures and dodecane was submitted to

the German Patent Office. In addition two prototype devices with nominal outgassing rates of water vapour 3×10^{-3} Pa l/s and 6×10^{-5} Pa l/s were tested and demonstrated good performance.

The permeation and in-situ nanohole reference outgassing devices can also be used to calibrate quadrupole mass spectrometers.

Reference outgassing samples/artefacts are now available for some important industrially relevant gas species such as water vapour and developments in the project enable outgassing rate measurements to be traceable and comparable. The reference outgassing samples can be placed in an outgassing rate measurement facility at the same location where the samples under test are measured, thus ensuring that the gas dynamics in the system are the same for both the reference and test samples. By comparing their results with the certified reference value the industrial outgassing rate measurement facilities can be calibrated or validated. Certified reference samples can also be used in a round robin test for the proficiency testing of systems in different laboratories.

A draft Technical Specification for the traceable measurement of outgassing rates was developed by the project for consideration by ISO TC112 Vacuum Technology.

Improvement of leak measurement and testing in industrial environments

Three different types of leaks were produced within the project and assessed: short ducts in metal discs, modified glass capillaries and nanometre sizes holes in silicon nitride membranes. In addition, two types of leaks, i.e. long micro machined channels and sintered material with porous channels for gas flow that were not developed within the project were also investigated.

Ducts were drilled in vacuum compatible metal discs using laser and focused ion beam technology. The six most regular artefacts were investigated and the gas flow rate generated from the leaks with respect to vacuum and to atmosphere pressure measured at different inlet pressures and with several industrially relevant gas species. Using these data it was possible to develop and verify models which describe the gas flow through the leaks. As a result when a short tube with circular cross section is calibrated for one gas species, it is possible to predict the flow rate for many other gas species with an uncertainty of less than 15 %. The modified glass capillaries were found not to be fully suitable because their geometry could not be determined with sufficient accuracy. Holes of 100 nm diameter were drilled in 100 nm and 200 nm thin SiN-membranes using focused ion beams. The experiments showed agreement between the predicted flow rate and the measured flow rate within the combined uncertainties of about 10 %. The long capillaries (0.5 m) of polyamide material and glass prepared by micromachining, and the porous body leak also turned out to have predictable flow rates with typical deviations between experiment and theory of less than 10 %, with a maximum deviation of 15 %.

A practical guide for leak measurements using commercial leak detectors, targeted at both for users of leak detectors and standard committees, was produced and is available on the IND12 website.

The feasibility of a fast optical method to detect leaks in a few seconds was demonstrated for leak rates between $7.2\text{x}10^{-2}$ Pa L/s and 1.3 Pa L/s, but further improvements are needed.

Actual and potential impact

The project team undertook a wider range of activities to disseminate the project's outputs to the relevant industrial and scientific communities. The project organised a "Workshop on measurement characteristics and use of quadrupole mass spectrometers for vacuum applications" in April 2012 and a final workshop "Vacuum Metrology for Industry" that took place from 25-27June 2014 in Berlin with 54 participants. The results of the project were discussed with the stakeholders from industry, including the companies Singulus Technologies AG (a manufacturer of optical disc production lines) with their interest in dynamic vacuum measurements, ASML (the largest supplier in the world of photolithography systems for the semiconductor industry) and VAT vacuum valves AG (a major manufacturer of valves for vacuum applications) for outgassing rate measurements. 16 peer-reviewed papers on the project results and outputs have been published and a further 5 are in progress.

The project has enabled the European vacuum industry and end users to improve their products, process and services in a number of ways:

- Faster capacitance diaphragm gauges were developed within the project by INFICON AG and their response times evaluated to be as low as 1.3 ms by comparison with the newly developed fundamental standard. Three patents applications were submitted for these new gauges which are now available as a commercial product (the "Stripe" gauge series by INFICON AG) and INFICONAG have received an R+D prize for their development.
- The calibration standard for dynamic vacuum established in the project to determine the response time of vacuum gauges enables the vacuum industry to test and improve their developments and products in this respect for the first time. Companies outside of the project have already expressed interest in having their products tested.
- A calibration system for partial pressures to characterise and calibrate quadrupole mass spectrometers is now available for the first time in Europe. The system provides the manufacturers of quadrupole mass spectrometers with the opportunity to improve those aspects of the metrological performance of their products that were found to be rather poor during the project. During the final project workshop, users of quadrupole mass spectrometers expressed their satisfaction that the project had confirmed in a metrologically sound way their past experiences related to the performance of quadrupole mass spectrometers. The draft for a Technical Specification of the characterisation of quadrupole mass spectrometers developed within the project was discussed with ISO TC 112 and the relevant German standardisation committee and was identified as particularly beneficial and has subsequently been approved as new work item within ISO TC 112.
- Reference outgassing samples are now available for some important industrially relevant gas species as water vapour and developments in the project enable outgassing rate measurements to be traceable and comparable. A draft for a Technical Specification of outgassing rate measurement procedures was discussed with ISO TC 112 and the relevant German standardisation committee and was identified as particularly beneficial and has subsequently been approved as new work item within ISO TC 112. The nanometre sized holes have already been successfully introduced into the research community as leak elements with fully predictable leak rates for any gas species. When the mounting technology for the leak elements is sufficiently robust for a commercial product - some necessary improvements were identified during the project - this will have a significant impact on the market of leak elements. As a result of this project it has become possible to predict the gas flow rate through a leak element for several gas species by just calibrating the leak using one gas species. This, in general, makes calibration service for users cheaper and more efficient.

The project outputs will also lead to longer term benefits in the future:

- Implementation of the new fast capacitance diaphragm gauges into production systems will enable processes such as optical disc metallisation to be shortened in a reliable manner resulting in a more economical process with higher profit. The performance of vacuum pumps can also be improved as the fast capacitance diaphragm gauges will enable fast transient processes within the pump to be observed.
- The methods developed for the simulation of fast pressure changes in the intermediate pressure regime will be applicable for improving the performance of vacuum systems and pumps and in reducing their energy consumption.
- Stakeholders and end users have already indicated their appreciation that, the project output, a Technical Specification for the characterisation of quadrupole mass spectrometers will be developed at the ISO level. This will help them to choose the instrument with suitable performance for their application, avoid inappropriate purchases and avoid incorrect interpretations of the measurement data obtained with the instruments.
- The availability of reference outgassing samples will significantly improve the unfortunate situation for outgassing rate measurements, where due to the lack of traceability and standardised

procedures, "local" solutions were developed which were costly and not comparable to other "local" solutions.

- In the near future it will be possible to reliably measure the outgassing rate of vacuum components that are needed to maintain clean vacuum environments in industrial processes. Developers of new materials with lower outgassing rates will also benefit from the new traceability and reliability, as it will enable the proper characterisation of new materials with well-defined uncertainties. In this area the new standard will have indirect impact on the reduction of energy consumption and increased productivity.
- The leaks developed with predictable leak rates for many gas species and suitable for industrial environments will make it easier to reliably identify and measure leaks within vacuum systems. This will reduce the costs associated with leak testing and also the potential risks to the environment and associated costs due to leaks of toxic or polluting gases. The mathematical model developed will allow the gas flow rate delivered by the standard leaks with defined geometry developed during the project to be predicted for several typical gas species based on knowledge of the gas flow rate for one gas species. Knowledge of the performance of the leak artefacts under industrial conditions as well as the metrological characteristics of helium and refrigerant detectors will enable a critical view of the European standards devoted to the calibration of detectors. A guide on a procedure for leak detector calibration and uncertainty evaluation will be available to the users where the main results obtained during the project are summarised.

List of publications

Articles about the project results (popular press)

[1] Nanopores boost leak detection, Physics World Focus on: Vacuum Technology, August 2013, page 3

[2] Vacuum measures up for vacuum, Physics World Focus on: Vacuum Technology, August 2014, p. 33-34.

Scientific journals

[1] D. Pražák, J. Zůda, J. Tesař, L. Peksa, M. Vičar, *Perspectives of atmospheric reference leaks calibration by gravimetric method*, Measurement 46 (2013), 621-627.

[2] Pantazis S, *Hybrid Continuum Particle Simulations of Unsteady Flows*, Proceedings of 28th International Symposium on Rarefied Gas Dynamics (2012), Zaragoza, Spain, AIP conference proceedings, 1501 (2012), 451-456, http://dx.doi.org/10.1063/1.4769567

[3] Kurt Sonderegger, Michael Dür, Joachim Buthig, Sarantis Pantazis, and Karl Jousten, *Very fast-opening UHV gate valve*, J. Vac. Sci. Technol. A 31, 060601 (2013); doi: 10.1116/1.4813836

[4] Karl Jousten, Sarantis Pantazis, Joachim Buthig,Regine Model, Martin Wüest, Jaroslaw Iwicki, *A standard to test the dynamics of vacuum gauges in the millisecond range*, Vacuum 100 (2013), http://dx.doi.org/10.1016/j.vacuum.2013.07.037

[5] Jousten, K., Kim Jin Tae*, The work of ISO TC 112 towards standardization for specification and calibration of quadrupole mass spectrometers*, Vacuum (2014), pp. 457-461; DOI information: 10.1016/j.vacuum.2013.07.013

[6] V. Ierardi, G. Firpo, U.Valbusa, *Nano-holes for vacuum applications*, Journal of Physics: Conference Series 439 (2013), June 2013 doi 10.1088/1742-6596/439/1/012033

[7] Sarantis Pantazis, Joachim Buthig, Karl Jousten, *Conjugate heat transfer simulations of a thermocouple sensor in a low temperature nitrogen gas ambient*, International Journal of Heat and Mass Transfer 70C (2014), pp. 536-544; DOI information: 10.1016/j.ijheatmasstransfer.2013.11.033.

[8] Manuel Vargas, Steryios Naris, Dimitris Valougeorgis, Sarantis Pantazis, Karl Jousten, *Hybrid modeling of* time-dependent rarefied gas expansion, J. Vac. Sci. Technol. A 32(2), Mar/Apr 2014, 021602, http://dx.doi.org/10.1116/1.4830283.

[9] L. Peksa, T. Gronych, M. Jeřáb, M. Vičar, F. Staněk, Z. Krajíček, D. Pražák, *Implementation of multi-opening orifices in the primary metrology of vacuums and small gas throughputs*, Vacuum, 2013, DOI: 10.1016/j.vacuum.2013.10.014

[10] Sarantis Pantazis and Karl Jousten, *Computational and experimental study of unsteady gas flow in a dynamic vacuum standard*, Vacuum 109 (2014), 373-384, http://dx.doi.org/10.1016/j.vacuum.2014.05.005.

[11] Sarantis Pantazis and H. Rusche, *A hybrid continuum-particle solver for unsteady rarefied gas flows*, Vacuum 109 (2014), 275–283, DOI: 10.1016/j.vacuum.2014.06.022.

[12] Manuel Vargas, Steryios Naris, Dimitris Valougeorgis, Sarantis Pantazis, Karl Jousten, *Time-dependent rarefied gas flow of single gases and binary gas mixtures into vacuum*, Vacuum 109 (2014), 385-396, http://dx.doi.org/10.1016/j.vacuum.2014.06.024.

[13] Vincenzo Ierardi, Ute Becker, Sarantis Pantazis, Giuseppe Firpo, Ugo Valbusa, Karl Jousten, *Focused ion beam produced nano holes as leak elements*, Measurement (2014), http://dx.doi.org/10.1016/j.measurement.2014.09.017

[14] D. Mari, M. Bergoglio, M. Pisani, M. Zucco*, Dynamic vacuum measurement by optical interferometric technique*, Meas. Sci. Technol. 25, 125303, doi:10.1088/0957-0233/25/12/125303

[15] U. Becker, D. Bentouati, M. Bergoglio, F. Boineau, W. Jitschin, K. Jousten, D. Mari, D. Prazak, M. Vicar, *Realization, characterisation and measurements of standard leak artefacts*, Measurement 61 (2015), 249-256, http://dx.doi.org/10.1016/j.measurement.2014.10.045

[16] M. Bergoglio, D.Mari, J. Chen, H. Si Hadj Mohand, S. Colin, C. Barrot, *Measurement of gas flow rates through rectangular micro channels with three different experimental setups*, Measurement 73, 2015, 551–562, doi:10.1016/j.measurement.2015.06.011

[17] V. Hauer, K. Battes, M. Flämmich, V. Ierardi, K. Jousten, J. Šetina, *Outgassing rate measurements with the difference method in the framework of EMRP IND12*, Vacuum 2015 doi:10.1016/j.vacuum.2015.03.033

[18] M. Vičar, D. Pražák, M. Bergoglio, D. Mari, S. Pasqualin, F. Boineau, D. Bentouati, U. Becker, K. Jousten, B. Ongun, R. Kangi, W. Grosse-Bley, C. Laganà, B. Farangis *Metrology of leak detection - Practical guide*, http://www.ptb.de/emrp/ind12-publications.html

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