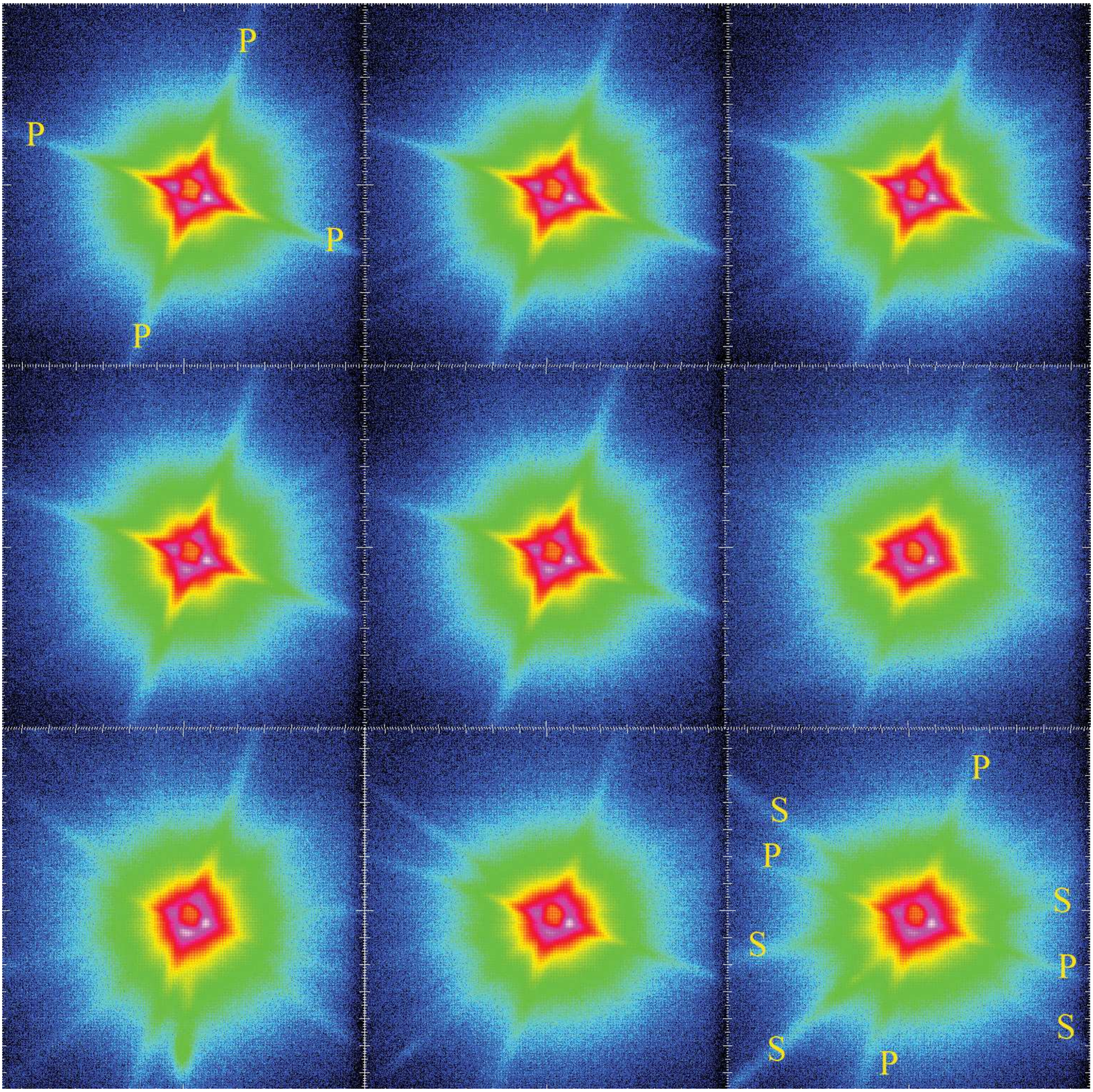
Report on the DUBBLE beamlines at the ESRF



Spring 2013

Report on the Dutch Belgian Beamlines at the ESRF (DUBBLE) covering the period 2007- 2012. Written for the 5 yearly beamline review by the European Synchrotron Radiation facility.

Previous DUBBLE project reviews have taken place in 2002 (5 yearly ESRF review) and 2007 (5 yearly ESRF review. In 2007 also an NWO/FWO initiated review was conducted which was followed by an NWO/FWO review in 2011. Based upon the outcome of the last review NWO and FWO have decided to continue the funding till at least end 2017.

Responsible author: W. Bras (project leader DUBBLE)

Front page: SAXS results from Dr. Gozde Dere and Dr. Eric Offermans on precipitates in metal foils. The streaks seen represent a problem already studied by Prof A. Guinier.

Contents

Acronyms

BENESYNC	Belgian Dutch consortium participating for 6% in the ESRF
CRL	Compound Reflective Lenses
CRG	Collaborative Research Group
DPI	Dutch Polymer Institute
DUBBLE	Dutch-Belgian Beamlines at the ESRF
ESRF	European Synchrotron Radiation Facility
FWO	Fonds Wetenschappelijk Onderzoek (Flemish Scientific Research Council)
HRPD	High Resolution Powder Diffraction
ID	Interface Diffraction
NWO	Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organisation for Scientific Research)
SAXS/WAXS	Small and Wide Angle X-ray Scattering
SR	Synchrotron Radiation
SWOT	Strengths, Weaknesses, Opportunities and Threats
XAFS	X-ray Absorption Spectroscopy (also XAS, XANES, EXAFS)

Summary

- The DUBBLE project consists of two synchrotron radiation beamlines at the European Synchrotron Radiation Facility that provide additional access to experimental techniques for which the ESRF public beamlines cannot provide sufficient beamtime to satisfy the requirements of the Dutch and Flemish user community.

- The DUBBLE project is co-funded by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek and Fonds Wetenschappelijk Onderzoek Vlaanderen.

- The DUBBLE beamlines have in the last 10 years produced around 750 publications and PhD thesis of a high qualitative level. The annual output can be expected to stabilize around 70-80 publications/year. The beamlines are responsible for around 50% of the total Dutch and Belgian materials science publications using synchrotron radiation based results.

- The instrumentation has been upgraded in the last 3 years and it can be foreseen that the technical state of the beamlines is such that another 7-10 years operation at an internationally recognized high level can be achieved without the requirement for major investments assuming that the ESRF upgrade program does not impose major investments or source degradation.

- The beamlines cover a very wide range of research fields but are especially an important research tool for Dutch, Flemish and high quality international research groups in the fields of catalysis, polymer and colloid research. There is a growing interest in the use of the beamlines by groups working with ‘energy’ related materials as well as environmentally relevant soil research.

1 Introduction and brief history

1.1 Introduction

The Dutch Belgian Beam Lines at the ESRF (DUBBLE) project is a shared initiative between the Dutch and Flemish research councils (NWO/FWO)[1] to operate two beamlines at the European Synchrotron Radiation Facility (ESRF). The remit is to provide experimental possibilities for the Dutch and Flemish research communities in addition to the beam time that is available to the Dutch and Belgian researchers via the BENESYNC participation in the central ESRF laboratory. The beam lines have been in operation for about 12 years and in this period produced data which have been used in more than 700 refereed publications and over 100 PhD theses as well as formed the basis for several patents.

The core funding of DUBBLE is an operations budget that is provided on a 75:25, NWO/FWO basis and the beam time is, in this ratio, made available to Dutch respectively Flemish user groups.

The beamlines are specialized in two techniques for which the ESRF, via the public beam line access program, cannot provide sufficient access to satisfy the Dutch and Flemish demands. The two main techniques are combined Small and Wide Angle X-ray scattering (SAXS/WAXS) and X-ray absorption spectroscopy (XAFS). There is a strong emphasis on time-resolved experiments and the simultaneous combination of different techniques.

1.2 Brief history

The collaboration between NWO and FWO regarding the construction of two beam lines at the ESRF started in 1987. The beamlines were designed and constructed in close collaboration with the drawing offices and workshops of AMOLF and NIKHEF. The first beam line started generating experimental results in 1999 followed by the second in 2003. Originally it was foreseen to implement 4-5 experimental techniques. Due to technological developments, user pressure for different techniques and a gradual changes in the research landscape in the Netherlands and Flanders it was decided to limit this to two main techniques (SAXS/WAXS and X-Ray spectroscopy) and one minor (Interface Diffraction).

In 2008, after the positive reviews of 2007 a program to refresh and upgrade the beamlines and sample handling environments was initiated. At the time of writing this program is nearly completely implemented. This was achieved within the DUBBLE budget with additional funds obtained via the NWO Middelgroot program. There has been no user down-time due to this upgrade program.

[1] NWO Nederlandse Organisatie voor Wetenschappelijk Onderzoek, FWO Fonds Wetenschappelijk Onderzoek Vlaanderen

2 Mission, Objectives and Vision

This section follows the style dictated by reviews performed by the Netherlands Organisation for Scientific Research (NWO). This in order to leave the reader with an idea about what the constraints are within which DUBBLE is operating, not only from the ESRF side but also from the funding agency side.

2.1 Mission

The core mission of the DUBBLE project is to provide synchrotron radiation beam time for the Dutch-Flemish research community in experimental techniques which are not sufficiently available via the participation in other synchrotron radiation laboratories, particularly the ESRF. The experimental facilities offered to the research community should be of a high qualitative level.

2.2 Objectives

The objectives of DUBBLE are :

provide 70% of the available experimental time to the Dutch-Flemish user community

provide 30% of the available experimental time to the international community

provide experimental facilities at an internationally recognized high level

provide a low threshold synchrotron access for Dutch and Flemish users

2.3 Vision

An efficient and high level of experimental opportunities should be offered to the users thus enabling a large scientific output at an internationally recognized qualitative high level.

The beam lines are continuously developed driven by interplay of user demands, novel technological developments and imaginative use of combinations of existing technologies. The scientific staff, with the broader oversight of both the multidisciplinary scientific demands as well as the technological possibilities, fulfils a pro-active role in this process.

In the 30% of the international beam time it is aimed to attract and retain a user base of high level international user groups and stimulate collaborations between these user groups and the Dutch-Flemish community.

3 Organisation

3.1 General

DUBBLE is a Collaborative Research Group (CRG) at the ESRF. This means that 70% of the experimental time (beam time) is made available for research groups within the remit of the funding agencies NWO and FWO. The 70% of the available beam time is on a time-averaged basis divided in 75% NWO and 25% FWO in proportion to the financial contributions from the funding agencies. The remaining 30% of the beam time is, according to ESRF regulations, made available to general ESRF users. The core funding for operations/maintenance without manpower is around 650 keuro. Manpower is budgeted separately due to different overheads in Flanders and the Netherlands. There is a core funding for 7 staff members.

DUBBLE is part of the Dutch and Flemish research infrastructure. Belgium and the Netherlands do not own any national synchrotron radiation source but Dutch and Belgian users have access to the ESRF public beam lines via the Benesync consortium. This provides access to a larger portfolio of techniques. The overall share of 6% to which Benesync users are entitled is at present not fully used although the beamlines that offer comparable techniques to DUBBLE are also for the full 6%, or more, used by Dutch and Belgian users.

With the Swiss-Norwegian CRG at the ESRF there is an exchange agreement in which a limited amount of beam time on their powder diffraction, single crystal diffraction and combined Raman/XAFS set-ups can be used in exchange for access to the DUBBLE facilities for their user community.

3.2 Administrative

The main governing body of DUBBLE is the steering committee. This is made up of NWO and FWO appointed members. The chairman of the steering committee is the formal representative of DUBBLE to the ESRF. NWO is the legal contracting party to the ESRF.

The user community is represented by the program committee with members appointed by the steering committee. Together with the project leader they advise on the scientific direction of the project.

Beam time proposals are assessed by 4 referees which for reasons of the NWO ‘code of conflict’ rules are not allowed to apply for beamtime via the CRG channels. Effectively this means that there is an international committee.

The project leader is based in Grenoble, is responsible for the beamline operation, maintaining scientific contacts with the existing user community, attracting new user groups, his own research and the supervision and the in-house scientific program.

3.3 The local team

The staff in Grenoble maintains, operates and develops the beamlines and provides user support in the broadest sense. This ranges from establishing contacts with potential new user groups, investigate technique improvements, discus the scientific issues with the users, help and train users in data analysis and find user groups in hitherto unexplored research areas etc.

There is no differentiation between user support for international ESRF and 'CRG' users. Wherever meaningful the staff tries to initiate informal collaborations between the different groups using DUBBLE in order to benefit from the synergy off collaborating teams in shared expertise, financial resources etc.

Each of the scientific staff members is involved in research activities as well. This both via the in-house research program but as well as via collaborations with user groups.

Although not part of their remit the local team is also often assisting new or potential user groups from the Benesync area to identify which ESRF public beam line is providing the best solution if experiments require techniques that cannot be carried out on DUBBLE and mediate in the contacts with other beam line scientists.

4 Technical

A brief technical description of the beam line optics is given in Appendix 1. More detailed descriptions can be found in the previous review reports.

4. 1 Radiation source

The radiation sources for the beam lines are the 0.4 T bending magnet spectrum for the XAFS beam line and the 0.85 T spectrum for the SAXS/WAXS line. The working energy range as specified in the original concept was for both beam lines from 5-30 keV. This is still the case for the SAXS beam line but in response to user demand it has been extended from 4.5 - 42 keV for the XAFS beam line.

4.2 BM26A/XAFS beamline

Here we describe briefly the beamline optics, the technical developments since the previous review and some ideas for technical changes to the beamlines in the coming years. It should be remarked that since we are in the final stages of the last upgrades the details for future developments have not necessarily crystallized out yet.

4.2.1 Basics

This beamline has a flux of around 10^{11} photons/sec which in the normal operation mode can be focused in a 300 micron spot. It is possible to perform transmission, fluorescence and electron yield spectroscopy. The original optics were a sagittal focusing monochromator[2] equipped with Si111 crystals and a vertical focusing mirror which is also used for harmonics rejection.

Standard detection systems are conventional ion chambers and a 9 element monolithic Ge detector with digital electronics. The monolithic design ensures a large detection solid angle and is crucial in many of the time-resolved experiments with more complicated sample environments.

Routine sample environments like furnaces and cryostats are available.

This beam line receives on average 1-2 visiting research groups/week.

[2] The sagittal focussing was a relic from the time that this beam line was supposed to do both EXAFS as well as MX/powder diffraction. This option was never used for EXAFS experiments due to loss of energy resolution.

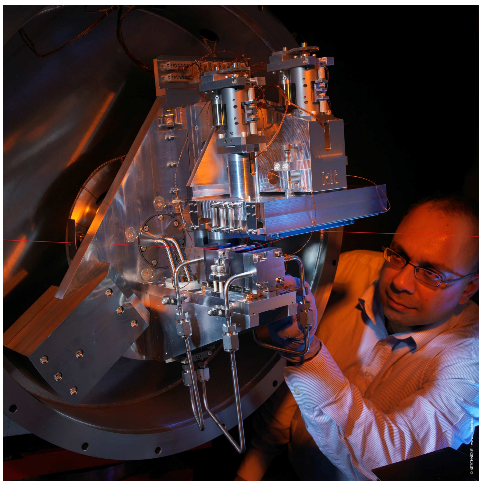


Figure 4.1
The newly installed monochromator with both Si111 and Si311 crystals. This monochromator will further enhance the stability of the beam and allows us to extend the energy range to around 45 keV.

4.2.2 Developments since previous review

In the original optical lay-out of the beamline a collimating mirror was foreseen and the vacuum chambers and mirror purchased. This mirror has been installed during the 2012 ESRF upgrade shutdown. The old sagittal focusing monochromator has been replaced by a more conventional double crystal monochromator (December 2012) with an on-axis encoder. By sideways translation it is now possible to choose between Si111 and Si311 crystals. The energy range is extended beyond 40 keV.

The original improvised experiments table has been replaced by a table which is more stable and versatile than the old system. This allows the installation of heavier sample environments, equipment for auxiliary techniques like XRD and, due to its low vibration level, the installation and use of microfocus optics.

The 9 element fluorescence detector had deteriorated. It has completely been refurbished in 2011. This was done without interruption to the user program thanks to the (identical) loan detector from STFC/Diamond.

A beam position/profile monitor was installed with feedback to the mirror control system. This has eliminated longer term drifts due to slow optics (Compton) heating as well as it has allowed a more accurate control over which part of the sample is illuminated by the X-ray beam[3]. This results in a beam positional stability of around 2 micron.

Due to the large number of experiments in catalysis it was decided to install a gas mixing system to safe on installation time and reduce the number of safety checks. This gas rig can accurately mix most of the catalytically relevant gasses. The gas flow can be controlled over three different ranges by mass flow controllers. Pressures up to 50 bar are possible and there is a steam generator available[4].

The possibility to perform EXAFS/WAXS quasi simultaneous was done by using a position sensitive curved Inel detector. The data quality of this detector was especially at higher energies not very satisfactory and the system is been replaced with a small diffractometer equipped with a position

[3] The input from Dr. Roelof van Silfhout, Manchester is gratefully acknowledged.
[4] The system has cost around 100 keuro of which 50 keuro was externally financed by interested groups. A PhD student from University College London (Vladimir Martis) based at DUBBLE was responsible for the implementation.

sensitive Mythen detector. Tests of this new system have been delayed somewhat due to delays in the data acquisition software that should enable this to be implemented in a more transparent fashion.

The large interest in nucleation and growth type of experiments has also initiated the development of a set-up in which SAXS/WAXS and XANES can be carried out simultaneously. This allows to follow a whole structure forming chemical reaction from the initial stages where only pre-cursors of reaction products are present, via the early stages where crystalline order is still too low but the longer range density variation is visible with SAXS, to the growth of crystallites once they have sufficient size or crystallographic order.

During the last period also an electron yield detector was adapted which can detect the photo electrons energy resolved. This allows one to obtain Xanes spectra in a depth resolved way. The emitted electrons which have been generated deep inside the sample will have on average a lower energy than the photons generated in the surface/near-surface area. This method is not completely new but has been adapted for operation with more realistic catalysts. This project was carried out by an UCL sponsored student in collaboration between UCL/Diamond and DUBBLE.

The introduction of polycapillary microfocus optics which have been used for microfocus XAFS[1] Confocal microfocus tomography has been developed by the group of L. Vincze in collaboration with DUBBLE staff. Voxel resolution of 10 micron has been achieved[5] and not only fluorescence data but also real XAFS data has been collected. This microfocus for the use of XAFS is rather rare at the ESRF and worldwide.

Section references:
Nikitenko, S.; Beale, A. M.; van der Eerden, A. M. J.; Jacques, S. D. M.; Leynaud, O.; O'Brien, M. G.; Detollenaere, D.; Kaptein, R.; Weckhuysen, B. M.; Bras, W., Implementation of a combined SAXS/WAXS/QEXAFS set-up for time-resolved in situ experiments. *Journal Of Synchrotron Radiation* **2008,** *15*, 632-640.
G. Silversmit, B. Vekemans, S. Nikitenko, S. Schmitz, T. Schoonjans, F. E. Brenker and L. Vincze, *Spatially resolved 3D micro-XANES by a confocal detection scheme*, Physical Chemistry Chemical Physics **12** (2010), no. 21, 5653–5659.
R. van Silfhout, A. Kachatkou, N. Kyele, P. Scott, T. Martin and S. Nikitenko, *High-resolution transparent x-ray beam location and imaging*, Optics Letters **36** (2011), no. 4, 570-572.

4.2.3 Work in progress

New GDA software is nearing completion. This will allow us to replace the existing obsolete Sun workstations and has as another major advantage that it will become easier to implement the data acquisition strategies required for the combined technique experiments.

The orders for the mechanical improvements of the complementary SAXS and diffraction set-up have been placed.

The position sensitive Inel detector that was used for the time-resolved XRD experiments had too limited a limited spatial resolution and was inefficient at higher photon energies. A new diffractometer system is being installed and will be fully commissioned when the GDA software is completely functional.

[5] Nominal beam size is 0.3 x 1 mm^2 (VxH)

In the light of the many studies that are taken place on both BM26A/B on the onset of crystallization in various systems (polymers, zeolites, bone mineralisation etc.) it might be interesting to invest in the possibility to make PDF scattering experiments possible. This technique can shed light on the amorphous/poorly crystalline state of very small clusters of atoms. To this effect the monochromator energy range has already been extended. Collaboration with University College London, in which a PhD student will be based at DUBBLE will have this development as one of the core targets.

4.2.4 Future developments

The main occupation of the station in the coming years will be to utilize and benefit from the improvements that have been implemented in the last number of years.

In collaboration with L. Vincze from Gent University the micro imaging and fluorescence work using confocal methods will be extended and made available to a larger user community. Vincze has been granted some priority access to the beamline for this by the DUBBLE steering committee and will base personnel with DUBBLE as well as allow access to a newly developed two dimensional energy dispersive detector See also the Capita Selecta in chapter 10.

The gas system for XAFS should be extended with a mass spectrometer for measuring the output during on-line catalysis experiments. This will be purchased as soon as the DUBBLE budget allows and can be brought forward in the spending profile with contributions by interested user groups.

Even though the monochromator crystal cage has just been renewed there are still improvements feasible. One of the potential plans is to replace the existing monochromator axis with an air bearing system. This would reduce the mechanical noise level even further and it would allow for more rapid angular scanning. For normal and even time resolved exafs the latter is not required. However, it might be interesting for Pair Distribution Function (PDF) experiments where the PDF data is obtained at higher energy and the XAS data at the appropriate edge. A rapid and reliable scanning between these two energy ranges will be required.

Since 2007 this station has seen an increase in geochemical experiments related to soil contamination (Wageningen, Utrecht, Gent, Diepenbeek) as well as energy related research (CO_2 storage, H_2 storage and battery materials). Research efforts from Dipanjan Banerjee will be focused in this direction.

At present there are several studies about possible improvements/extensions to the BM26A station under investigation. These will maybe be implemented if the user community in the Netherlands-Flanders is sufficiently interested.

- In order to increase the time resolution of the experiments it could be considered to implement a fast scanning monochromator (Frahm type). However, this will have major consequences for the already existing optics and will require outside funding.

- One could consider obtaining a Vortex Si detector for energies up to 20 keV. This might be an interesting addition to the Ge fluorescence detector and could be helpful for the further development of the microfocus work.

4.3 BM26B/SAXS/WAXS beamline

4.3.1 Basics

The beam line has been in operation since 2000. In the normal operation mode the SAXS/WAXS beamline generates a flux of 10^{11} photons/sec in a 300 micron spot. Diffraction distance from 0.15-200 nm can be resolved. Time-resolved experiments can be carried out down to a msec/frame. The beamline was designed to be able to install large scale sample handling equipment in order to follow for instance polymer processing techniques on-line.

There is a variety of sample environments available. Among else there are available:

- Furnaces ranging from room temperature to 1100 C
- Differential Scanning Calorimetry (Linkam) -150 – 550 C
- Tensile tester
- Rheometer (Anton Paar)
- Parallel plate shear cell (Linkam)
- 4 and 7 Tesla superconducting magnets
- Stop-flow cells (Biologic)

There is also a range of detector systems available. Pilatus detectors (300k, WAXS; 1M SAXS) for high countrate low noise applications and Frelon and Photonic Science detectors where the detector pixels size is more important and the noise level of lesser importance.

In this experimental hutch also an Interface Diffractometer (ID) is installed. This instrument has effectively not been used in the last two years.

This beamline receives on average 2 research groups/week.

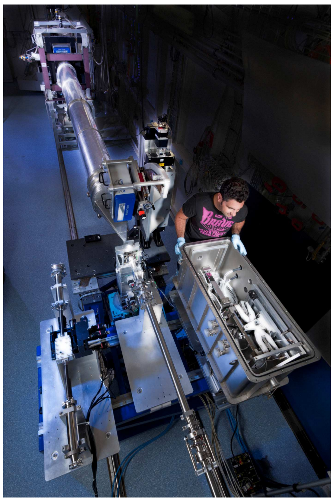


Figure 4.2
The SAXS/WAXS beamline with table for the two microfocus options

4.3.2 Developments since previous review

Two post-focusing microfocus systems are available. A low background Kirkpatrick-Baez system which generates a spot size of 15 x 5 microns with a low angle resolution of 30 nm (real space) and 20x20 micron with low angle resolution of 70 nm for soft condensed matter experiments. The flux is around 10^{10} photons/sec. The second is a microfocus Compound Refractive Lenses system (CRL) which is used extensively for the studies of silica based colloidal systems (Lekkerkerker, Utrecht). This focus the beam down to around 10 microns. The latter option preserves the beam coherence and allows diffraction distance larger than 1 micron to be studied at the expense of a somewhat elevated background. Both systems have been used extensively and recently a new table was installed to allow the rapid changeover between those systems.

The data acquisition software has recently been upgraded to the GDA system developed at the Daresbury/Diamond laboratories.

Within the DUBBLE budget space was found to obtain a Pilatus 300k detector which replaced the old MSGC WAXS detector. With the help of a successful bid for funds from the 'NWO Middelgroot' program a Pilatus 1M detector was obtained in order to replace the gas filled wire chambers.

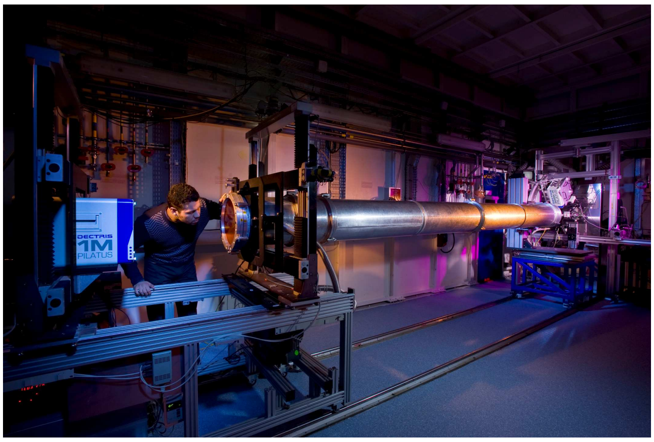


Figure 4.3
The SAXS/WAXS beamline equipped with the two new, state of the art, Pilatus detectors

The detector mountings have been adapted so that it is now possible to have overlap between the SAXS and WAXS detecting ranges.

A new sample stage has been developed which gives micrometer precision translation and allows single crystal type work to be carried out. For an illustration see the cover page of this report.

4.3.3 Work in progress

A set of beam position monitors will be installed. A feedback loop to the mirror and possible to the sagittal focusing mechanism will be introduced. This will allow for more stable performance but it will also be possible to suppress vibrations down to around 60 Hz (part of these are known storage ring vibrations and part are due to the DUBBLE optics)

With DPI funds the Anton Paar rheometer will be upgraded to a new model (MCR302) suitable for the Couette, plate-plate and extensional geometries.

Successful Grazing Incidence SAXS experiments have been performed with improvised set-ups but in order to release this option to general users some mechanical components have to be modified. This is in progress at the moment and will be released to users around November 2013. However the first manuscripts have been submitted.

Cédric Renaud, Sébastien-Jun Mougnier, Eleni Pavlopoulou, Cyril Brochon, Guillaume Fleury, Dargie Deribew, Giuseppe Portale, Eric Cloutet, Sylvain Chambon, Laurence Vignau and Georges Hadziioannou. Block Copolymer as a Nanostructuring

Agent for High-Efficiency and Annealing-Free Bulk Heterojunction Organic Solar Cells, *Advanced Materials (2012)DOI: 10.1002/adma.201104461*
Interaction, structure and surface morphology of gold nanoclusters deposited on SiOx/Si substrates
*Giuseppe Portale,*1 Francesco Giannici2 , Luisa Sciortino2, Antonino Martorana2, Cristiano Albonetti4, Fabio Biscarini4, Wim Bras1 and Alessandro Longo*,3 Submitted for publication*

4.3.4 Future developments

There is an extensive program in polymer processing experiments. In this it has been possible to perform crystallization experiments under shear and with industrially relevant cooling rates. Also extrusion, film blowing and deformation rates have been performed which mimick the real industrial conditions. In collaboration with TU Eindhoven/DPI and with support from SABIC, DSM and EXXON the existing program will be enhanced by the addition of a PhD student and a post-doc position. The PhD student will be involved in further developments in this field and the postdoc will be involved in a project to model the on-line processing results. (This is a field in which TU Eindhoven has made great progress in recent years and which has attracted industrial interest).

The GISAXS set up will be further developed and this most likely will open up new research areas in blockcopolymers and devices.

5 Staffing

The core funding for the Dubble team consists of one projectleader, three beam line scientists and three technical support staff. In addition there are at present three externally funded postdoc positions and three PhD positions.

The main activity for the core personnel at DUBBLE is user support and beam line development.

For the last 7 years a postdoc position has been funded by the Dutch Polymer Institute (DPI). This has recently been renewed and an extra post has been funded. The DPI postdocs are involved in the support of DPI related projects but further have no beam line responsibility. Another two year postdoc position is funded by the Alistor project.

In the period 2007-2012 one PhD student funded via University College London (Bras/Sankar/Catlow collaboration) successfully finished his work. A second UCL student (Bras/Catlow) is foreseen to start in 2013 (Bras/Beale). A DPI funded student has just been recruited.

Time averaged there are visiting scientist whom spend time with DUBBLE. Dr. Roelof van Silfhout has spend a sabbatical and has been involved in the development of beam conditioning devices and now a collaboration using GISAXS methods on devices. Dr Alessandro Longo also is spending a sabbatical year and is involved in several projects dealing with scattering techniques.

	2007	2008	2009	2010	2011	2012	2013
Tenured staff	1	1	3	3	3	3	3
Non tenured staff[6]	4.16	4.16	2.16	2.16	2.16	2.16	2.16
PhD students			1	1	1	1	3
Technical staff	3	3	3	3	3	3	3
Visiting fellows		0.5	0.5	0.5	0.5	1.5	0.5
Staff via ESRF	0.6	0.6	0.6	0.6	0.4	0.4	0.4

Table 5.1 staffing levels period 2007-2013

The DUBBLE budget does not allow the further recruitment of core staff for user line support. It has already been mentioned in several reviews that this is in fact an untenable situation.

Core staff:

Dr. W. Bras

Project leader, overall responsibility, instrumentation development (both optics as well as sample environments and technique combinations), attracting new user groups, contact with existing user groups about research directions. Carries out own research and supervises the research from the team members and the students/postdocs. Spends 2 x 1 month/year in Lawrence Berkeley Laboratory

Ir. D. Detollenaere

Senior engineer, data acquisition electronics, computer support, vacuum specialist, supplier contacts

Mr. S. Nikitenko

User support XAFS, beam line operation, beam line scheduling.

Dr. G. Portale

User support SAXS, beam line operation, beam line scheduling, instrumentation development, polymer research. Carries out own research.

Dr. D. Banerjee

Instrumentation development, User support XAFS. Carries out own research.

Programmer

Vacancy

Mr. F. Ledrappier

Mechanical engineer, technical user support

Postdocs:

Dr. D. Hermido

DPI postdoc, DPI user support, Carries out own polymer research.

Dr A. Brownrigg

Alistor postdoc, research in Li-ion battery materials

[6] Dr. W. Bras spends two months/year in Lawrence Berkeley Laboratory and is during this period partially replaced by Dr. G. Luijckx whom then takes responsibility for the operations.

Postdoc

DPI postdoc. Vacancy. Partially based in TU Eindhoven. Involved in modeling of the data generated by the on-line polymer processing experiments that are generated on DUBBLE. (collaboration W. Bras/G. Portale and G. Peters/H. Goossens, project in start-up phase)

Students:

H. Islam

Catalytic aspects of greenhouse gasses sequestering. Academic home institution University College London (collaboration W. Bras and R.C.A. Catlow)

Vacancy

Development of technique combinations for on-line catalysis research with XAS as basis. Specifically the development of Pair Distribution Function Analysis in combination with XANES/XRD to be able to study the very early stages of structure forming. Academic home institution University College London (collaboration W. Bras and A. Beale, project in start-up phase)

Vacancy

Development of on-line polymer processing experiments. Academic home institution TU Eindhoven, DPI funded. (collaboration W. Bras/G. Portale and G. Peters/H. Goossens, project in start-up phase, post has been offered to candidate)

The visiting fellows:

Dr. Allessandro Longo

Palermo University. Involved in the technical development of GISAXS hard- and software

Dr. Jim Torbet

Free lance. Involved in biophysics projects. Teaching local staff about biological fibre diffraction

Dr. Roelof van Silfhout

Development of X-ray beam position monitors. This work has resulted in 5 publications co-authored with DUBBLE staff. This work has also resulted in a patent. Also a beam stabilization unit on BM26A was developed which uses a feedback loop between the beam position monitor and the mirror position. Regular consultant to DUBBLE on optical systems

Dr. Halina Stanley

Glass devitrification and radiation effects on physical and chemical processes

Dr. Guy Luijckx

Replacement of the project leader for 2 x 2 weeks/year. Also contacts with engineering companies in Netherlands/Belgium.

6 In-house research

Besides the beam line support duties most scientist working on the DUBBLE beamlines are also involved in in-house research. Part of this work is instrumentation research and part of this is wider ranging. Apart from W. Bras and D. Banerjee all scientists working at DUBBLE can be considered to be 'early career' researchers. There are many collaborative efforts with external user groups. Overall supervision is by W. Bras.

Some references to representative publications are given.

Core staff

W. Bras
Research interests in:
- bio-polymers under external fields (shear, magnetic, tensile)
- vitrification processes in glass ceramics
- on-line catalysis and difference between bulk/surface catalysis
- on-line polymer processing
- use of on-line pulsed and static magnetic fields (also involved in the Euromagnet network, i.e. Nijmegen, Grenoble, Dresden and Toulouse)
- influence of radiation on crystallization and chemical processes

(co-)author on 44 publications and 2 book chapters in the period 2007-2012

G. Portale
His research focuses mainly on polymer crystallization and polymers for energy.
Polymer crystallization under processing conditions: he is involved actively in the development of ultra fast cooling system for mesophase studies on polymers (polypropylenes and polyamides).
Polymers for energy: he works on proton and electrical conducting polymers. He is involved in the study of the nanostructure of proton conducting polymeric membranes for fuel cell applications. Within this framework, he collaborates with Dr. KD Kreuer from the Max Plank in Stuttgart. He develops the GXD/GISAXS method for the study of electron conducting polymers for solar cell applications. He is also working on novel liquid crystals membranes with the group of Prof. D. Broer (TU/e Eindhoven), Dr. T. Dingemans (TU/e Delft) and Dr. O. Francescangeli (University of Ancona).

J. C. Gielen, M. Wolffs, G. Portale, W. Bras, O. Henze, A. F. M. Kilbinger, W. J. Feast, J. C. Maan, A. Schenning and P. C. M. Christianen, Molecular organization of cylindrical sexithiophene aggregates measured by X-ray scattering and magnetic alignment, Langmuir 25 (2009), no. 3, 1272-1276.

Cédric Renaud, Sébastien-Jun Mougnier, Eleni Pavlopoulou, Cyril Brochon, Guillaume Fleury, Dargie Deribew, Giuseppe Portale, Eric Cloutet, Sylvain Chambon, Laurence Vignau and Georges Hadziioannou. Block Copolymer as a Nanostructuring Agent for High-Efficiency and Annealing-Free Bulk Heterojunction Organic Solar Cells*, Advanced Materials (2012)DOI: 10.1002/adma.201104461*

Appel, W. P. J., Portale, G., Wisse, E., Dankers, P. Y. W., Meijer, E.W., *Aggregation of Ureido-Pyrimidinone Supramolecular Thermoplastic Elastomers into Nanofibers: A Kinetic Analysis*, MACROMOLECULES, 44, 17, 6776-6784

D. Banerjee
He has joined DUBBLE only recently. In his previous employment he was employed by the ROBL CRG where he was involved in the geochemistry of transuranes. In his position with DUBBLE the focus will be slightly moved to geochemistry of non-radioactive materials and geo-catalysis.

Abrasonis G., Wintz S., Liedke M., Aksoy F., Krause M., Kuepper K., **Banerjee D.**, Liu Z., Gemming S. (2012) Environment controlled de-wetting of Rh-Pd bilayers: A route for core-shell nanostructure synthesis. *Journal of Physical Chemistry C*, **116**, 14401-14407.

Hennig C., Weiss S., **Banerjee D.**, Brendler E., Honkimäki, V., Cuello G., Ikeda-Ohno A., Scheinost A.C., Zaenker H. (2013) Solid-state properties and colloidal stability of thorium(IV)-silica nanoparticles. *Geochimica et Cosmochimica Acta*, **103**, 197-212.

Postdocs

D. Hermida

DPI postdoc. Develops instrumentation for on-line supercritical CO_2 processing with the intention to utilize this for self assembly processes of core shell particles (Luik University, TUE and Nottingham). Is responsible for polymer deformation studies in collaboration with H. Goossens and templated crystallization in collaboration with N. Sommerdijk

Ivana Vukovic, Thomas P. Voortman, Daniel Hermida Merino, Giuseppe Portale, Panu Hiekkataipale, Janne Ruokolainen, Gerrit ten Brinke, and Katja Loos, *Double Gyroid Network Morphology in Supramolecular Diblock Copolymer Complexes*, Macromolecules, 45, **8**, 3503-3512

N. Ricardo, F. Costa, C. Chaibundit, G. Portale, D. Hermida-Merino, S. Burattini, I. W. Hamley, C. A. Muryn, S. K. Nixon and S. G. Yeates, *The effect of n-, s- and t-butanol on the micellization and gelation of Pluronic P123 in aqueous solution*, Journal of Colloid and Interface Science **353** (2011), no. 2, 482-489

Alex Brownrigg

Alistor postdoc. Develops the XAFS electron yield system for on-line electrochemistry. This will be applied to materials relevant for battery research.

Postdoc vacancy (2 year)

DPI funded. The work of this postdoc will be to further refine the link between the data obtained with realistic polymer processing experiments and the existing models for polymer flow and crystallization.

Students

V. Martis

Phd student (University College London)

Has developed the ‘dry’ electron yield system in order to be able to distinguish between bulk and surface catalytic activity of materials in a single experiment. To this effect he has also co-designed the high pressure gas system to be used with the electron yield experiments. These systems have been used for Fischer Tropsch catalysts. (obtained PhD in 2012)

Vladimir Martis, Richard Oldman,Ross Anderson, Martin Fowles, Tim Hyde, Richard Smith, Sergey Nikitenko, Wim Bras and Gopinathan Sanka, *Structure and Speciation of Chromium ions in Chromium doped Fe2O3 Catalysts*, Physical Chemistry Chemical Physics 2013 **(**accepted**)**

V. Martis, S. Nikitenko, S. Sen, G. Sankar, W. van Beek, Y. Filinchuk, I. Snigireva and W. Bras, *Effects of X-rays on crystal nucleation in lithium disilicate*, Crystal Growth & Design **7** (2011), 2858-2865

This thesis work will yield 6 manuscripts in total.

H. Islam
Phd student, 2nd year (University College London)
Catalysis related research with the natural sequestering of greenhouse gasses as topic.

PhD vacancy (University College London)
Project approved February 2013. Further development of methods for on-line catalysis in specific the investigation into PDF techniques in combination with XAFS

PhD vacancy (Dutch Polymer Institute)
Project approved January 2013. Further development of methods for on-line polymer processing in close collaboration with the DPI postdoc for modeling these processes and in close collaboration with G. Portale.

Visitors

A. Longo
Solution scattering and methodology development. Involved in collaboration with Jansen Pharmaceuticals.
A. Longo, G. Portale, W. Bras, F. Giannici, A. Ruggirello and V. Turco Liveri, *Structural characterization of frozen n-heptane solutions of metal containing reverse micelles*, Langmuir **23** (2007), no. 23, 11482-11487
L. Sciortino, F. Giannici, A. Martorana, A. M. Ruggirello, V. T. Liveri, G. Portale, M. P. Casaletto, and A. Longo, Structural characterization of surfactant-coated bimetallic cobalt/nickel nanoclusters by XPS, EXAFS, WAXS, and SAXS, Journal of Physical Chemistry C 115 (2011), no. 14, 6360-6366

R. van Silfhout

Beam position and beam profile monitors. Feedback mechanisms for optics control. Thin film structures for sensor development.
R. van Silfhout, A. Kachatkou, N. Kyele, P. Scott, T. Martin and S. Nikitenko, *High-resolution transparent x-ray beam location and imaging*, Optics Letters **36** (2011), no. 4, 570-572
Around 10 manuscripts based upon DUBBLE data and in collaboration with DUBBLE staff

H. Stanley
Devitrification of glasses and formation of Pb clusters under the influence of 10 keV X-rays
Manuscript in preparation

J. Torbet
Mechanical behaviour of fibrinogen linked to molecular deformations
2 manuscripts in preparation

7 Representative publications

References to representative publications with data from the DUBBLE beamlines are shown below. It is chosen to show publications per technique with a predominantly Dutch or Flemish author list as well as references to work by international users.

EXAFS

1. Di Vece, M.; Bals, S.; Verbeeck, J.; Lievens, P.; Van Tendeloo, G., Compositional changes of Pd-Au bimetallic nanoclusters upon hydrogenation. *Physical Review B* **2009,** *80* (12), 4.
2. Beale, A. M.; van der Eerden, A. M. J.; Jacques, S. D. M.; Leynaud, O.; O'Brien, M. G.; Meneau, F.; Nikitenko, S.; Bras, W.; Weckhuysen, B. M., A combined SAXS/WAXS/XAFS setup capable of observing concurrent changes across the nano-to-micrometer size range in inorganic solid crystallization processes. *Journal Of The American Chemical Society* **2006,** *128* (38), 12386-12387.
3. Van Damme, A.; Degryse, F.; Smolders, E.; Sarret, G.; Dewit, J.; Swennen, R.; Manceau, A., Zinc speciation in mining and smelter contaminated overbank sediments by EXAFS spectroscopy. *Geochim. Cosmochim. Acta 74* (13), 3707-3720.
4. Bruggeman, C.; Maes, N., Uptake of Uranium(VI) by Pyrite under Boom Clay Conditions: Influence of Dissolved Organic Carbon. *Environmental Science & Technology 44* (11), 4210-4216.
5. Walspurger, S.; Cobden, P. D.; Haije, W. G.; Westerwaal, R.; Elzinga, G. D.; Safonova, O. V., In Situ XRD Detection of Reversible Dawsonite Formation on Alkali Promoted Alumina: A Cheap Sorbent for CO2 Capture. *Eur. J. Inorg. Chem.* (17), 2461-2464.

SAXS

1. Rissmann, R.; Groenink, H. W. W.; Gooris, G. S.; Oudshoorn, M. H. M.; Hennink, W. E.; Ponec, M.; Bouwstra, J. A., Temperature-induced changes in structural and physicochemical properties of vernix caseosa. *Journal of Investigative Dermatology* **2008,** *128* (2), 292-299.
2. Pal, A., Besenius, P., Sijbesma, R. P., *Self-sorting in rodlike micelles of chiral bisurea bolaamphiphiles*, Journal of the American Chemical Society, 133, 12987-9, 2011, DOI 10.1021/ja205345e
3. Besenius, P.; Portale, G.; Bomans, P. H. H.; Janssen, H. M.; Palmans, A. R. A.; Meijer, E. W., Controlling the growth and shape of chiral supramolecular polymers in water. *Proceedings of the National Academy of Sciences of the United States of America 107* (42), 17888-17893.
4. van den Pol, E.; Petukhov, A. V.; Thies-Weesie, D. M. E.; Byelov, D. V.; Vroege, G. J., Experimental Realization of Biaxial Liquid Crystal Phases in Colloidal Dispersions of Boardlike Particles. *Physical Review Letters* **2009,** *103* (25), 4.
5. Cédric Renaud, Sébastien-Jun Mougnier, Eleni Pavlopoulou, Cyril Brochon, Guillaume Fleury, Dargie Deribew, Giuseppe Portale, Eric Cloutet, Sylvain Chambon, Laurence Vignau and Georges Hadziioannou. *Block Copolymer as a Nanostructuring Agent for High-Efficiency and Annealing-Free Bulk Heterojunction Organic Solar Cells*, Advanced Materials (2012)DOI: 10.1002/adma.201104461

International users

1. Detlefs, C.; Duc, F.; Kazei, Z. A.; Vanacken, J.; Frings, P.; Bras, W.; Lorenzo, J. E.; Canfield, P. C.; Rikken, G., Direct observation of the high magnetic field effect on the Jahn-Teller state in TbVO4. *Physical Review Letters* **2008,** *100* (5).

2. Masciocchi, N.; Galli, S.; Colombo, V.; Maspero, A.; Palmisano, G.; Seyyedi, B.; Lamberti, C.; Bordiga, S., Cubic Octanuclear Ni(II) Clusters in Highly Porous Polypyrazolyl-Based Materials. *Journal of the American Chemical Society* **132** (2010), no. 23, 7902-7904

3. Lei Li, Martin Rosenthal, Heng Zhang, Jaime J. Hernandez, Markus Drechsler, Kim Hô Phan, Stephan Rütten, Xiaomin Zhu Dr. Dimitri A. Ivanov, and Martin Möller, Light-Switchable Vesicles from Liquid-Crystalline Homopolymer–Surfactant Complexes, Angew. Chem. 2012, 51, 1 – 5, DOI 10.1002/anie.201205660

4. C. Travelet, G. Schlatter, P. Hebraud, C. Brochon, A. Lapp, D.V. Anokhin, D.A. Ivanov, C. Gaillard, G. Hadziioannou, Multiblock copolymer behaviour of alpha-CD/PEO-based polyrotaxanes: towards nano-cylinder self-organization of alpha-CDs, Soft Matter, 4 (2008) 1855-1860.

5. Giorgi Titvinidze , Klaus-Dieter Kreuer , Michael Schuster , Carla C. de Araujo , Jan P. Melchior , and Wolfgang H. Meyer, *Proton Conducting Phase-Separated Multiblock Copolymers with Sulfonated Poly(phenylene sulfone) Blocks for Electrochemical Applications: Preparation, Morphology, Hydration Behavior, and Transport*, Adv. Funct. Mater. 2012, DOI: 10.1002/adfm.201200811

8 Performance parameters

In this section not only the application and pressure and publication output is discussed but also a sketch is made of how DUBBLE fits into the framework of Dutch-Belgian use of the ESRF.

8.1 Application pressure

Applications for beamtime on DUBBLE come from two sources. The largest part (70%) is available for user groups with affiliations to Dutch and/or Flemish universities and other public research laboratories. The remaining 30% of the time is available for general ESRF users. It should be noted that due to the participation in the ESRF Dutch and Belgian users are also entitled to a 6% of the beamtime on the ESRF public beamlines.

In fig 8.1 the application pressure from the Dutch/Flemish community is given. Also indicated is the amount of time that on average is available per scheduling period. The ESRF operates both an EXAFS as well as a SAXS/WAXS beamline. An indication of the amount of time that could be available to the Dutch/Belgian community is also given in the figure. In general this amount of beamtime is also used. It should maybe be noted that most industrially highly developed countries with a national source have several SAXS and XAS beamlines installed on these national facilities.

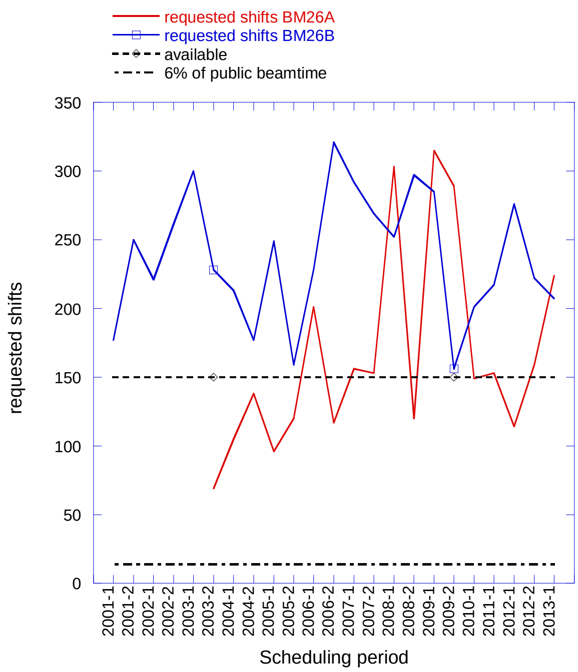


Figure 8.1
The application pressure on BM26A and BM26B from the Dutch/Flemish community. The dotted lines indicate what amount of time is roughly available per scheduling period and what amount of time would be available for the same users on the ESRF public beamlines.

Time-averaged the oversubscription rate hovers around a factor of 1.75 – 2.00 on both beamlines[7]. There are some strong fluctuations which are correlated with the numbers of PhD students starting/finishing in a certain year. An oversubscription in excess of two is in general detrimental for the quality of the research since PhD supervisors loose the trust that their students will have a reasonable chance of success in their applications and therefore consider to divert the students to (most times) second best techniques with more change of success.

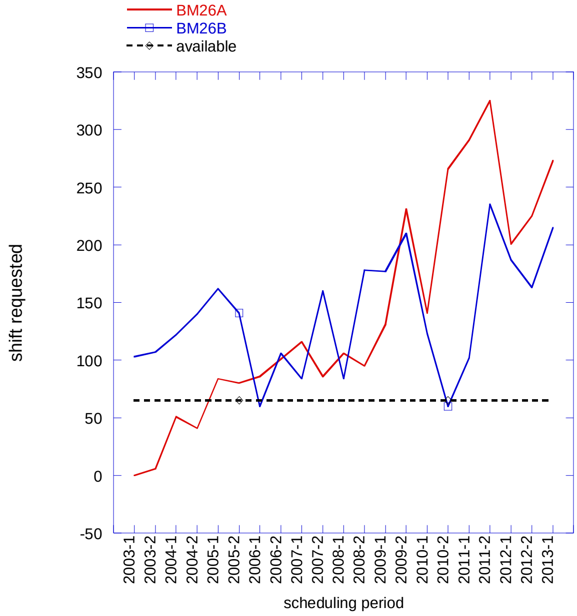
In figure 8.2 the application pressure from the ESRF general users as well as the average amount of available beamtime is given. The very high oversubscription on the EXAFS station in 2011 is probably correlated with the lengthy shutdown and rebuilding of the ESRF BM29/BM16 public beamline. Part

[7] According to the American Physical Society report: 'Access to major international X-ray and Neutron scattering facilities' an oversubscription of 1.5 – 2.25 is the natural equilibrium state of beam lines.

of the user groups are overlapping. A similar effect one can expect on BM26B when ESRF ID02 will also be closed for thanks to the upgrade program (2013).

Figure 8.2

The application pressure on BM26A and BM26B from the ESRF general users. The dotted lines indicate what amount of time is roughly available per scheduling period.



The recent large oversubscription for the ESRF part on BM26A is associated with the temporary closure of ESRF station BM29. The total oversubscription rate for both stations is manageable when the ratio remains between 1.5 – 2x. If the oversubscription increases above a factor 2x many users find the access too erratic in time and do not want to risk PhD/postdoc programs and consequently look for alternative beam lines or ‘second best’ techniques.

It should be noted that in the spirit of the ESRF it is stimulated that the 30% of the beamtime reserved for general ESRF users is indeed used by international users. There is a strong belief that in the long run the cross fertilization between the international and the Dutch-Belgian users is of more importance than a temporary and marginal boost of the Benesync juste retour numbers.

8.2 Delivered shifts

The ESRF is contractually obliged to deliver a certain number of user shifts/year. However, the accelerator complex is operating for a larger number of hours in different machine modes. Apart from the ‘4 bunch mode’ all other machine modes are useable to the DUBBLE beamlines. On average DUBBLE should deliver around 650 shifts/year to the CRG users and around 280 shifts/year to the general ESRF users.

In general DUBBLE delivers more than is contractually required. It should be remarked here that this is favorable for the users but sometimes rather hard for the user support staff.

There is no data available on the reliability of the beamlines. Since the start of user operation only 1 – 2 user groups have received no useable data due to beam line failure (in 10 years time > 1000 experiments sessions have taken place). Occasionally there have been small issues with beam line vacuum or detectors.

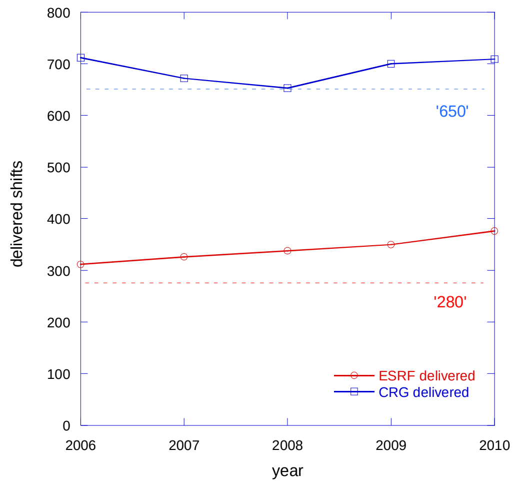


Figure 8.3 Number of shifts annually provided to users for BM26A and BM26B combined.

8.3 Publication output

The SAXS/WAXS beam line has been in use since 1999. Operation in the first years was hindered by the construction of the EXAFS and Protein Crystallography beamline. When the decision was made to abandon the idea of performing PX on DUBBLE both beamlines became fully operational. Fig @ shows the total output per technique. Included in this are the publications in powder diffraction which have been obtained under the collaboration contract with the Swiss-Norwegian beam line.

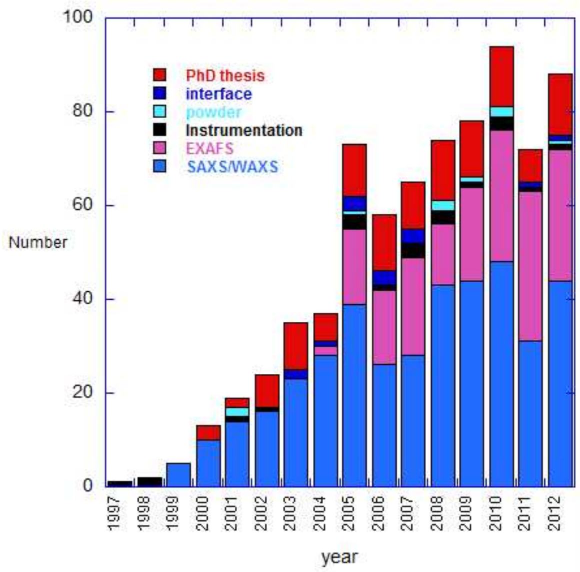


Fig 8.4
The annual publication and PhD thesis output of BM26A and BM26B.

Both beamlines are mature now and the average annual production is stabilizing at an average of around 75-80/year. In general the SAXS/WAXS beamline has a somewhat higher publication output than the EXAFS line. The contribution of the Interface Diffractometer is marginal.

The quality of the output is also a parameter that was investigated. In figure 8.5 one finds the journal impact factor versus the percentage of publications above a certain impact factor.

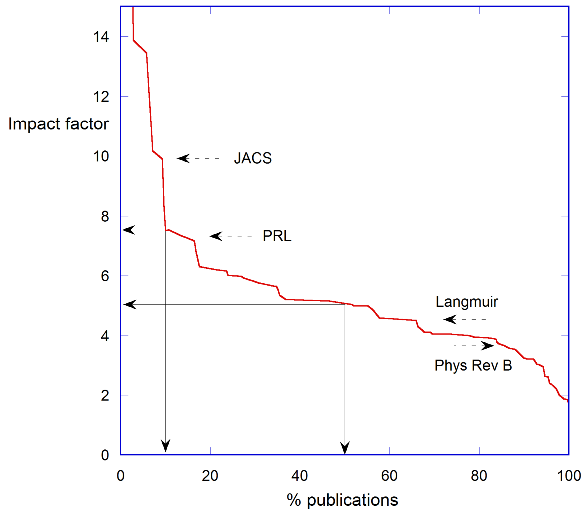


Fig 8.5
On the vertical axis is plotted a journal impact factor. On the horizontal the percentage of publications above a certain impact factor. This indicates that the median impact factor per publication is 5.04 and that 10% of the articles are published in journals with an impact factor of more than 7.54. Some well know chemistry and physics journals are indicated for comparison.

Figure 8.5 shows the combined output of both beamlines. The median point, 5.1, is within the error margin the same for both beamlines. It can be noted that the median impact factor for DUBBLE publications in the previous ESRF review in 2007 was around 4.2.

8.4 The Dutch-Belgian context

For the Dutch and Belgian funding agencies it is interesting to place these numbers in context with the total publication ESRF output with at least one Dutch-Belgian associated co-author.

In figure 8.6 the comparison is made, first in approximated output and second normalized on the budgets involved in the ESRF Benesync contribution and the DUBBLE budget. For the ESRF output the library data base is used. From the total Dutch-Belgian output 0.7 times the DUBBLE output is subtracted. This is in line with the 30% of the DUBBLE beamtime that has to be made available to the general ESRF users and therefore is less likely to have Dutch-Belgian co-authors.

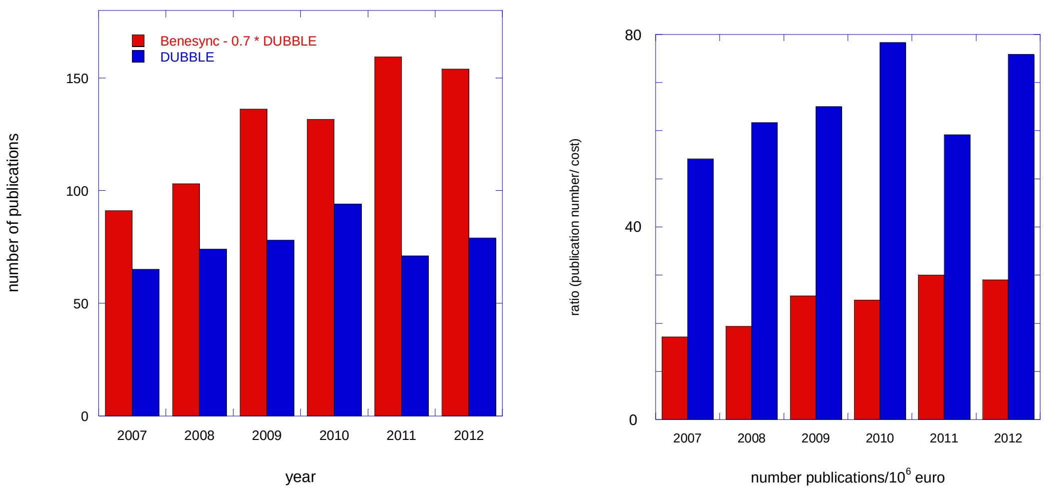


Figure 8.6
Approximate publication output of Benesync and DUBBLE. Left hand panel total numbers, right hand panel normalised on Benesync ESRF contribution and DUBBLE budget i.e. the number of publications

per 10^6 euro spend at the ESRF. This can be interpreted as an indication of the cost effectiveness of operating a CRG.

Obviously these numbers should not be over-interpreted. It is for instance not taken into account that the protein crystallography beamlines have on average a higher number of publications. Since about 25% of the ESRF beamlines is dedicated to this technique and DUBBLE is not involved in this the numbers for material science/chemistry/physics related publications will be somehow skewed. It is also found that the library database does not contain all the DUBBLE publications. It is reasonable to assume that the average Benesync users is as forgetful as the average DUBBLE user and it cannot be expected from the ESRF librarian to follow each beamline as close as the DUBBLE staff can do with their user community. However, it is difficult to avoid the conclusion that operating a CRG, in addition to the contribution to the ESRF itself can lead to a rather effective output/cost ratio.

Both DUBBLE beamlines are multipurpose instruments mainly dedicated to the demands of the Dutch and Flemish user community. This is reflected in the availability of sample handling equipment and associated infrastructure specifically developed to the demands of this user group. However, there is still a fairly large variety of research topics addressed by the researchers on the beamlines. In figure 8.7 one can see a rough estimate of the partition of the number of publications over different research fields. This can only be treated as a rough indication since there are many overlapping and interdisciplinary fields.

A remark that should be made here is that the category 'colloids' is mainly hard condensed colloids like for instance colloidal crystals used for photonic applications. Food and other soft condensed matter colloidal systems are placed in the categories 'food', 'bio/pharma' or 'polymers'.

The category 'energy' contains subjects from Li-ion batteries to ceramic sieves for diesel particulates.

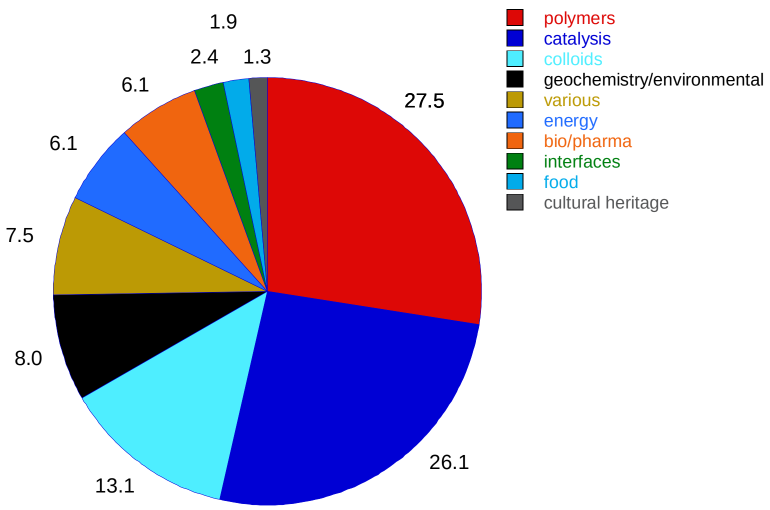


Figure 8.7

Approximate distribution of the publications over different research areas.

Due to the nature of the beamlines it is not surprising that catalysis and materials science are topics that are often addressed by the researchers using the beamlines. However, a substantial fraction of the output in colloids, interfaces and 'various' deals with area that fall within the remit of the more physics oriented research councils.

Figure 8.8
An example of the large scale industrial processing equipment that can be mounted on the SAXS beam line. Polymer structure and processing forms a large part of the DUBBLE research portfolio.

9 Future developments

The philosophy followed in the last 10 years with respect to developments has been to carry these out gradually in order not to disturb the user program and to remain within the allocated budget. The main developments have been in the implementation and development of sample environments and technique combinations. Also work has been done to extend the existing techniques with options which form a logical addition to the already existing infrastructure. The introduction of the electron yield EXAFS and the GISAXS are examples of this.

The main research subjects have remained unchanged. As predicted in 2007 the percentage of publications related to EXAFS has sharply increased. This is due to the fact that in the previous review period the EXAFS station had only become operational two years earlier whilst in the 2007-2011 review period this station was fully active. The relative decrease in polymer related publications should be seen in the light of the increased output. The absolute number is hardly changed (around 100 publications in both periods). Growth can be seen in geochemistry and energy materials. Also an increase in metals and alloys research is seen in the beam time proposals. However, this is a recent trend and does not show up in the publication output yet.

In figure 9.1 the percentage of publications in different research fields is shown and a comparison is made with the situation in 2007.

The degree of sophistication of the experiments carried out on both BM26A as well as BM26B has increased with more emphasis on technique combinations in the case of BM26A and more complicated sample environments on BM26B. Most of these complicated experiments are driven by the interaction of the DUBBLE staff with the Dutch-Flemish user community.

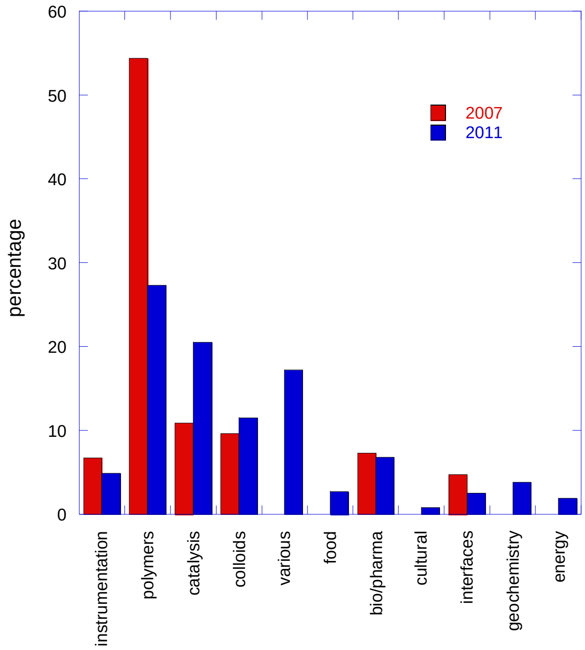


Figure 9.1

Development of the main research areas targeted by the DUBBLE user group

In the coming years the user community can benefit from the substantial improvements that already have been made to the two beamlines. At present the DUBBLE team is working at implementing microfocus and grazing incidence options on both beamlines. These will become available for the general user community in the coming 2 years.

The funding from the Dutch and Flemish research councils is secured till end 2017. From a financing perspective it is unpredictable what will happen after this period ends. From e technical point of view the beamlines are capable of operating beyond this point at an internationally competitive and leading level.

A major point of uncertainty is the ESRF upgrade plans. If these bring substantial changes in the source quality or require the beam lines to be moved, which will have financial implications, then this could bring the DUBBLE project in a less comfortable position with respect to the willingness of the funding agencies to continue.

10 Capita Selecta

In this section the attention is focussed on some aspects of the DUBBLE beamlines and the associated research. This is not intended as an all encompassing oversight.

10.1 The collaboration with the Swiss-Norwegian CRG (SNBL)

This collaboration was initiated several years ago with the idea of broadening the number of techniques available for the DUBBLE and SNBL user communities and to make more efficient use of resources such as sample environments, workshops, sample preparation laboratories and detectors. Under this agreement, user groups can apply for beamtime in order to carry out experiments that cannot be done on the 'home' beamline. It also allows for staff to have access to in-house research beam time on any of the beamlines operated by either CRGs. This can be used to develop new techniques or alternatively, it is often used for extra characterization of samples. However, the exchange of beam time and services is also accompanied by a considerable exchange of expertise. For example, DUBBLE has recently benefited greatly from the experience gathered at SNBL with constructing a high pressure gas handling system.

The main technique used by Dutch-Flemish users on SNBL is powder diffraction and to some degree XAS in combination with on-line Raman spectroscopy. In exchange the SNBL users have benefitted from access to the DUBBLE SAXS/WAXS facilities. So far around 20 publications using data obtained under this exchange have appeared, and a few examples are listed below. In some cases, the research leading to a publication has been done on both DUBBLE and SNBL beamlines, and a recent publication on zeolites which appeared in *Nature Materials* is a good example of this synergy.

Some examples of publication arising from the DUBBLE/SNBL collaboration agreement:

Stability and reactivity of ε–χ–τ iron carbide catalyst phases in Fischer-Tropsch synthesis: Controlling μ(c), E. de Smit, F. Cinquini, A. M. Beale, O. V. Safonova, W. van Beek, P. Sautet and B. M. Weckhuysen, Journal of the American Chemical Society 132 (2010), no. 42, 14928-14941.

Design of zeolite by inverse sigma transformation, Verheyen, Elke, Joos, Lennart, Van Havenbergh, Kristof, Breynaert, Eric, Kasian, Nataliia, Gobechiya, Elena, Houthoofd, Kristof, Martineau, Charlotte, Hinterstein, Manuel, Taulelle, Francis, Van Speybroeck, Veronique, Waroquier, Michel, Bals, Sara, Van Tendeloo, Gustaaf, Kirschhock, Christine E. A., Martens, Johan A., Nature Materials, 11,1059–1064(2012)

P. Mandaliev, R. Dahn, J. Tits, B. Wehrli and E. Wieland, EXAFS study of Nd(III) uptake by amorphous calcium silicate hydrates (C-S-H), Journal of Colloid and Interface Science 342 (2010), no. 1, 1-7.

Nematic textures in colloidal dispersions of Na-fluorohectorite synthetic clay
N. I. Ringdal, D. M. Fonseca, E. L. Hansen, H. Hemmen, and J. O. Fossum*
Phys. Rev. E 81, 041702 (2010)

Mesoscopic structure of dry-pressed clay samples from small-angle X-ray scattering measurements
Yves Méheust, *Simon Dagois-Bohy*, *Kenneth D. Knudsen* and *Jon O. Fossum*
J Appl Crystallography Volume 40 Part s1 Pages s286-s291 2007

Note: this section is co-authored between Dr. Phil Pattison (SNBL) and WB.

10.2 The gas mixing system

In order to perform in-situ X-ray Absorption Spectroscopy (XAS) and X-ray Diffraction (XRD) experiments on materials that are subjected to heating, gaseous environments and pressures, a system has developed that allows experiments under high pressure and with controlled flow. Also a steam system is available[8].

Figure 10.1

The gas handling system on the XAS station which is used to simulate real operating flow and pressure conditions during catalysis experiments.

The schematic drawing of the gas rig showing gas handling components is presented in Figure 1. The rig consists of manual valves (MVs), mass flow controllers (MFCs), mixing cylinders, pneumatic valves (PVs), 4-way fast switching valve (SVs) and back pressure controllers (BPCs).

Desirable gas flow rates with high accuracy and repeatability (low (1-10 Nml/min), medium (1-30 Nml/min) and high (1-100 Nml/min)) are measured by six Brooks MFCs. Each MFC is pre-set with several pre-programmed calibration curves for pure gases and gas mixtures. Each MFC has an option for manual configuration of calibration curves, i.e. a gas conversion factor for a specific gas can be entered. MFCs are divided into two groups on the assembly panel. Each group of three mass flow controllers, covering accurately the low, medium and high flows, is fed into one of the two mixing cylinders. The mixing cylinders have a volume of ca. 4 ml and are filled with SiC. They are separated by a manual locked valve. Switching between different gas mixtures is rapid and can be done with remotely controlled 4-way fast switching valves.

To visually monitor flows, the gas flow transmitters are fixed before and after an experimental cell. The pressure in the cell and tubing is controlled by two 5866 Brooks pressure controllers (BPCs). The BPC can control pressure of dry gases up to 50 bars.

A steam generation system (e.g. vaporizer, saturator) can be connected to the system via 4-way fast switching valve, housed in the heating box (see Figure 1). The benefit of using the second SV in the heating box is that the steam can be produced in a closed loop without interfering with the rest of the gas rig. These switching valves can be operated up to 240^{o}C.

The rig is controlled remotely and the dead volume is minimised in order to allow for rapid changes. Also a mass spectrometer can be attached to the exhaust for detailed analysis of the reaction products.

[8] This system has been designed in close collaboration between SNBL, Weckhuijsen group (Utrecht), Hensen group (Eindhoven) and the Dutch Energy Research Centre. Around 50% of the costs have been externally financed. The development was part of Vladimir Martis PhD thesis work.

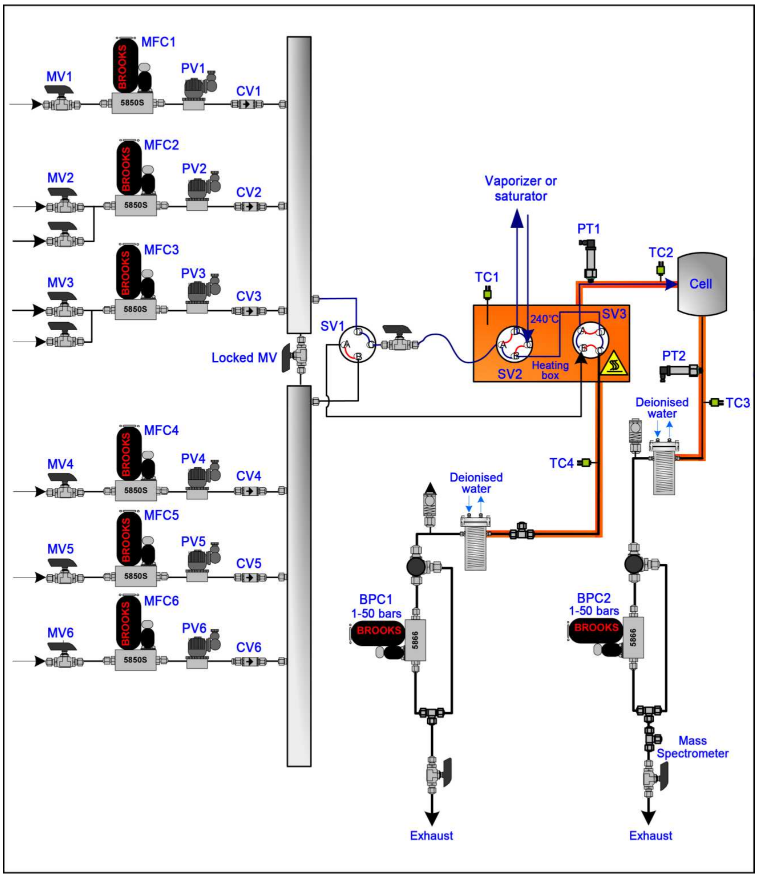


Figure 10.2
Scheme of a high pressure gas rig. The rig contains six MFCs that are fed into two mixing cylinders. Output of mixing cylinders is fed into 4-way fast switching valve (SV1). The latter is used to switch over between required gas mixtures. The pressure in tubing and reaction cells is controlled by two BPCs operating in the range of 1-50 bars. For experiments requiring steam, a steam generation system i.e. vaporizer or saturator can be connected via SV2 placed inside the heating box. The benefit of using two SVs is that mixture of steam and gases can be produced in a closed loop without interfering with the rest of the setup. To avoid cold spots, tubing is thermally insulated and the temperature is controlled at several places using thermocouples (TC) connected to Eurotherm. To avoid condensation in the BPCs, wet gas is condensed in condensers fitted before the BPCs. The maximum pressure using wet gas up to 20 bars in reaction cells can be obtained.

10.3 Chemically selective X-ray micro-imaging at the ESRF DUBBLE XAS station

In a series of pilot experiments in the period of 2007-2011, the Ghent University XMI research group in collaboration with DUBBLE staff successfully demonstrated chemically selective X-ray micro-imaging at DUBBLE by scanning micro-XRF/XAS methods [LV1, LV2]. The test set-up was based on glass polycapillary optics, specifically optimized for DUBBLE as a focusing element, providing a beam size down to 10 µm. These experiments have shown that the use of polycapillary allowed to obtain micro-XANES and micro-EXAFS at DUBBLE [LV3]. The focusing properties of the polycapillary optics result in a stable position of the focused beam in the focal plane, moreover, the EXAFS spectra show no significant distortions or amplitude losses compared to the standard non-focused detection mode [LV3]. In the framework of these pilot experiments, both microfocus and conventional XAS measurements were performed to solve problems in the field of 1) environmental science [LV4, LV5] and 2) cultural heritage [LV6]. It is expected that the micro-XAS capabilities of DUBBLE will be further developed for routine use and will be made available for the entire DUBBLE user community.

Future development work

Next to the above described microbeam based XRF/XAS imaging, we aim for the development and applications of a novel full-field X-ray fluorescence (XRF) and X-ray absorption spectroscopic (XAS) imaging approach based on a *unique two-dimensional (2D) energy-dispersive detection system for non-destructive chemical microanalysis* [LV7]. The XMI group has acquired the only existing prototype of this detector in 2012, which has been tested successfully at the BAM-line of BESSY-II (Berlin) and at beamline P06 PETRA-III (Hamburg). This detector is a development of the Institute for Scientific Instruments GmbH (IFG, Berlin, Germany) and PNSensor GmbH (Munich, Germany), currently being tested and optimised for XRF/XAS applications by the UGent XMI-group in the framework of an UGent-IFG collaboration agreement (UGent A11/TT-0609).

The above described extension of DUBBLE towards XRF/XAS microimaging will enable and/or complement X-ray micro-spectroscopic studies on unique materials including (i) inclusions in rare natural superdeep diamonds (ii) stellar condensates trapped in presolar carbon micro spheres and (iii) biological model organisms subjected to controlled exposure to toxic metals. The methodological development work and applications are associated with IWT doctoral research programmes.

Note: text by L. Vincze and WB

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10.4 Microradian diffraction set-up

X-rays possess a number of advantages in studies of colloidal self-assembly with colloidal particles in the micron size range. The intrinsically low contrast of x-rays allows one to collect clear structural information in suspensions exhibiting high turbidity and absorption as well as photonic materials with very high refractive index contrast in the visible [1]. The access to a broad q-range makes x-ray scattering a unique tool to study the structure of various lyotropic liquid crystals formed by anisometric colloidal particles with high aspect ratio [2,3]. The application of x-rays is, however, challenging due to the difference between the x-ray wavelength and the particle size. This challenge can be readily met using the microradian x-ray diffraction (µradXRD) setup [1,4] at the DUBBLE beamline, which is developed by Utrecht group in collaboration with the beamline personnel. This setup exploits x-ray refractive optics and is not routinely available elsewhere. It permits studying self-assembled structures with periods over a micron and detecting long-range periodic order on distances up to tens of microns from the intrinsic width of the diffraction peaks.

Figure 10.3 presents a few recent examples of the diffraction data obtained with the µradXRD setup. Panel (A) presents a typical scanning electron microscopy (SEM) image of the surface of a photonic colloidal crystal deposited on a glass substrate overlaid by a µradXRD pattern. The diffuse x-ray scattering along the so-called Bragg rods unambiguously shows that the defects seen in the SEM image, which were believed to be line defects, are in fact the openings of the stacking faults running into the bulk of the colloidal crystal [5]. Panel (B) displays µradXRD patterns measured in suspensions of core-shell magnetic colloids at different concentrations and applied magnetic field strengths. The data allows one to follow the self-assembly of particles into 1D chains through 2D hexagonal sheets into 3D structures [6]. Panel (C) shows an x-ray scattering pattern from a biaxial nematic phase of board-like goethite particles [7], which was discovered at DUBBLE.

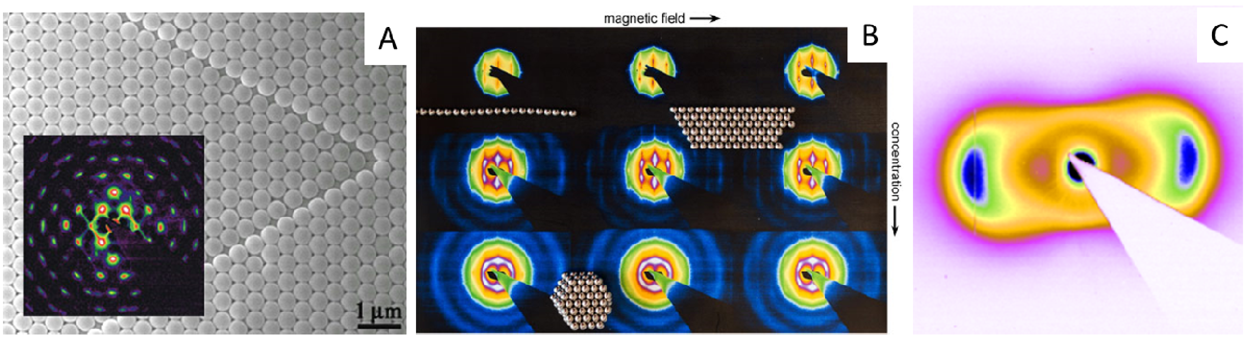

Figure 10.3.
Examples of microradian x-ray diffraction patterns measured from photonic colloidal crystals (A), suspensions of magnetic core-shell particles (B) and biaxial nematic phase of goethite boards-like particles.

Although several beamlines at the ESRF would be suitable for this development it would have been difficult to develop this set-up there due to the specialised nature of the experiments and the amount of beamtime required. This set-up has provided data for around 20 publications in the last 5 years.

Note: text and figure provided by A. Petukhov with some additions by WB

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10.5 Skin research

The research on the structure of human skin using SAXS has been carried out, initially on the Dutch Daresbury beamlines, and subsequently on the DUBBLE beamlines, for the last 25 years. This program driven by the group of Prof J. A. Bouwstra (Leiden University) has been very successful and encompasses several aspects of skin research. This started with the unraveling of the structure of the lipid layers in healthy stratum corneum (the upper layers of the skin, composed of dead material interleaved with lipids). The structure of these lipids is relevant in the framework of transdermal drug delivery. Subsequent work on the lipid structure of diseased skin has followed and is still continuing. One of the main interests is to obtain a better understanding of the structure of skin afflicted with atopic eczema. Also the development of creams on the basis of the vernix caseosa structure, which covers the skin of embryos in the womb, has been subject of research and development. In the light of European legislation regarding the elimination of the use of test animals it was opportune to develop a method to simulate the penetration profiles of substances through the skin. On the basis of the knowledge derived from the structure of the lipids it has been possible to create a test method in which synthetic ceramides can be sprayed with a linomat in such a fashion that a layered structure is created which accurately mimics the transport properties of real human skin. The group also studies the lipid organization in *in vitro* cultured skin. This *in vitro* skin can also be used to test diffusion of compounds through the skin and to mimic diseased skin.

We consider this work to be one of the prime examples of an interdisciplinary (biology, physics, chemistry and medicine) combination of academic and applied research with a high relevance for society in which the valorisation is obvious due to the patents granted and in which synchrotron radiation beamlines play an important role. It is also interesting to see that the amount of material required for these experiments has been reduced by nearly a factor of 50 since the first experiments 25 years ago.

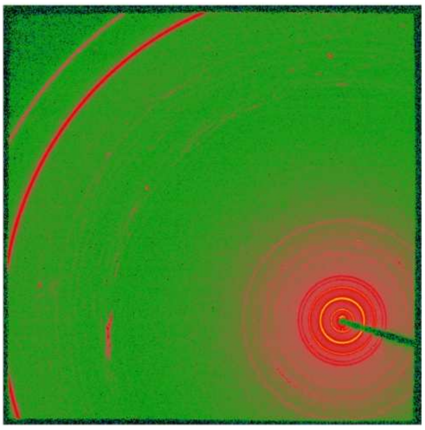

Figure 10.4
Typical human skin diffraction pattern obtained using microfocus technology. The use of the microfocus reduces the amount of material required which is of prime importance in for instance the study of diseased skin in which the samples have to surgically obtained from patients.

Note: Text by G. Gooris, M. Janssens and WB

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PhD Theses in period 2007-2013

2006 M.W. de Jager: Development of a stratum corneum substitute for in vitro percutaneous penetration studies
2009 R. Rissmann: Development of a vernix caseosa substitute
2009 J. Caussin: Stratum corneum hydration. Mode of action of moisturizers on a molecular level
2011 D. Groen: Stratum corneum model membranes: molecular organization in relation to skin barrier function
2012 V.S. Thakoersing: Barrier properties of human skin equivalents
2013 (May) M. Janssens: Atopic eczema: The role of stratum corneum lipids in the skin barrier

10.6 Polymer processing

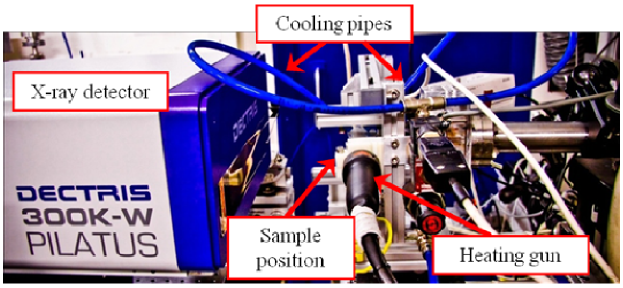
Despite their relatively young age, polymer products, thanks to their outstanding range of physical properties, had and still have a profound impact on our lifestyle. Obviously, their presence is certainly not going to decrease, since new applications are currently envisaged; both in completely new fields, e.g. biomedical, and as substitute for more energy-costly materials, such as metals. However, new uses implicitly mean more demanding requirements. This challenge to material science can be successful only when a detailed understanding of the relationships between the polymer, its processing and the end-use properties of the product is available.

The largest part of polymer products used in everyday life, from the fibers of our clothes to the shoppers at the supermarket, is made of semi-crystalline polymers. Shaping of polymeric materials to produce any kind of goods, from films to complex objects, involves application of flow fields accompanied or followed by rapid cooling. For semi-crystalline materials, the final morphology dependents strongly on the whole thermo-mechanical history experienced during processing. The imposed processing conditions determine orientation, size and structure of the crystals which, in turn, determine the final properties of the product.

Extensive characterization of polymer crystallization behaviour has been carried out in the last decades in several laboratories, but rarely the adopted experimental conditions mimic the actually industrial ones. Questions arise about the applicability of data acquired in this way to real life problems of crystallization during processing.

The BM26/DUBBLE beamline currently provides the optimal combination of techniques and expertises to pursue leading research in the field of polymer crystallization under processing conditions.

Representative examples are given in Figures 1 and 2. By combining a specially designed quenching device with a state-of-the-art Pilatus detector, the crystallization behavior of industrially relevant materials, such as i-PP homo- and copolymers and polyamide-6, was measured while cooling the sample up to 200°C/s. The use of the high frame rate Pilatus detector enable to collect 20 frames per second, with a remarkably good signal-to-noise ratio notwithstanding the small thickness of the sample and the short acquisition time of 47 ms. Detection of the onset of crystallization, as well as monitoring of the full crystallization process, is straightforward, even if the solidification occurs in tens of seconds. (see Figure 1).



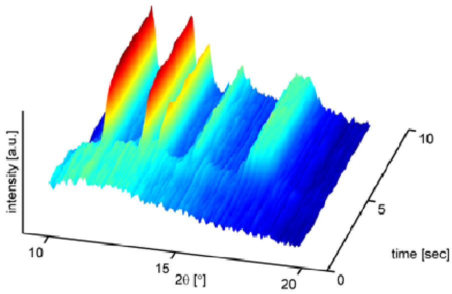
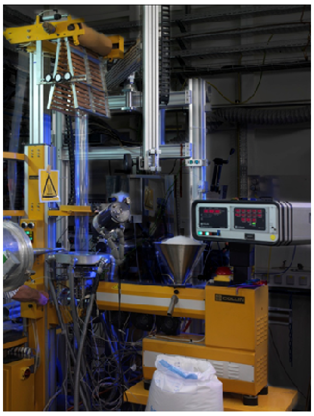


Figure 10.5

Real-time ultrafast WAXD during rapid quenching of thin polymer films. Experimental setup (left) and a typical outcome (right).

A second example of application of synchrotron X-ray to in-situ studies on industrial polymer transformation process is given in Figure 2. The blown film extrusion process is the main process to manufacture polymer packaging products. Complex biaxial stretching of the molten polymer occurs, imparting enhanced properties to the final film. X-Ray characterization of the structure development in low density polyethylene materials submitted to film-blowing was done at BM26/DUBBLE, by vertically moving the coupled system of extruder and die with respect to the beam. It was shown that processing conditions strongly impact the crystallization process moving from the die along the bubble (see Figure 2). These important results on crystallization during polymer processing will allow a new knowledge- based approach to materials and processes optimization, rather than the present trial-and-error method.



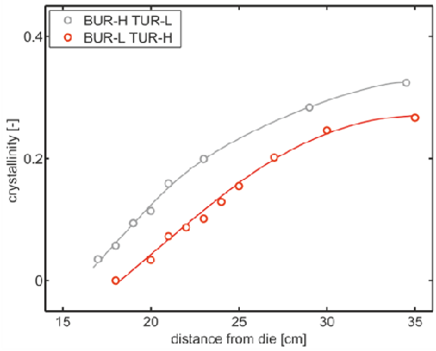


Figure 10.6. Pilot film-blowing plant at BM26/DUBBLE (left). Crystallinity of LDPE at different distances from the die for distinct stretch conditions (right).BUR = Blow up ratio, TUR = Take up ratio.

Other examples of in-situ X-ray measurements on experiments where industrial processing conditions were applied can be found in the references given below.

Note: section authored by G. Peters, D. Cavallo, H. Goossens and WB

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11 Recommendations ESRF review 2007

Recommendations made by the review committee of 2007.

1. *The recommendation to hire, beside the project leader, at least one scientific staff member that can enhance the in-house research program.*

In the last period two positions have been converted from time limited to permanent posts. With the recruitment of Giuseppe Portale and Dipanjan Banerjee the scientific profile of the in-house program is raised. The project leader has also managed to obtain funding for three postdoc positions and three student positions. Obviously this does not relieve the shortage of manpower for user support since these postdocs do not have any user support role apart from the DPI postdoc whom is involved in the support of DPI specific proposals.

2. *The implementation of micro focus options on the SAXS beamline*

There have been two microfocus options installed on the SAXS beamline. Both have been tested in improvised settings and recently a more permanent solutions has been installed that allows the changeover between options to be reduced to about 2 hours. In addition to this also on the EXAFS line we have implemented micro focus using capillary options. This will also be modified so that the installation time can be reduced.

3. *The replacement of the detectors for the BM26B beam line*

Both the SAXS as well as the WAXS detectors have been replaced with photon counting Pilatus detectors

4. *An associate membership of DUBBLE in the ESRF soft condensed matter group*

This has not materialized. We have seen no efforts from the ESRF management to implement this. A participation in the Soft Condensed Matter Initiative between the ILL and the ESRF could be feasible but in the present situation it is absolutely not clear if this would be value for money and in view of the limitation on budgets the participation in this initiative without any integration in the ESRF soft condensed matter group appears to be futile.

5. *Efforts to further develop the Dutch-Flemish XAFS community*

The Dutch EXAFS using community is already fairly highly developed. Two EXAFS courses have been initiated by Ghent University. The Flemish use of this technique is increasing not only in number of shifts used but also in the number of publications. This specifically in geochemistry and catalysis

6. *The collaborative development of data analysis software for time-resolved SAXS/WAXS data*

This has unfortunately not progressed much. The limitation is to find and retain capable manpower.

7. *The status of the interface diffraction station*

This technique has not been used in the last two years. The equipment might be used for reflectivity measurements in combination with GISAXS experiments. There will be no effort to revive a stand-alone program on this instrument.

Acknowledgements

Our colleagues Han Goossens and Guy Luijckx are thanked for providing information and advice. Sophie Rio from the ESRF library has provided the data on publications, Marion Glueckert poured these in a useable form and Joanne McCarthy and her team were very helpful in providing the beam time statistics. Obviously the DUBBLE team has helped in a great way with this report.

The requested contributions for the Capita Selecta from G. Peters, D. Cavallo, H. Goossens, G. Gooris, M. Janssens, A. Petukhov, L. Vincze are thankfully acknowledged

This report is written by the project leader W. Bras whom bears responsibility for the contents.

Appendix 1 Beamlines technical

The optical system of the beamlines has not fundamentally changed since the previous reviews. Beam position monitors and feedback loops from these monitors have been installed or are going to be installed. This improves the beam stability and beamline reliability but is less relevant for the overall picture. In this section only a very brief description of the optics is given.

Bending magnet BM26 is the radiation source for the DUBBLE beamlines. As usual at the ESRF this magnet doesn't have a uniform field but has a 0.4 Tesla and 0.8 Tesla section. BM26A uses the 0.4 T radiation and BM26B the 0.8 T. There are marginal differences. Both beamlines receive significant flux in the 4 – 60 keV photon energy range.

The experimental hutch wall thickness of BM26A is cleared for white beam operation, i.e. also suitable for higher energies. However, extending the range from BM26A to higher energies would require extra Brems-strahlung shielding and photon shutters. BM26B is limited to 30 keV photons.

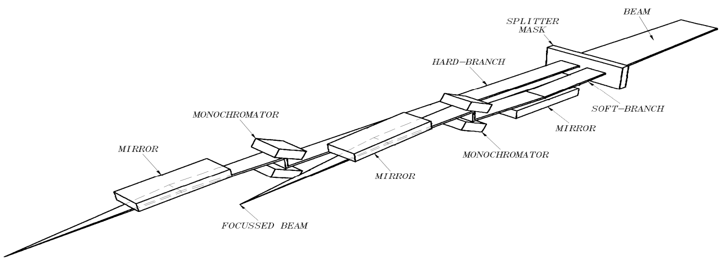

Figure A1
The schematic optical lay-out from the beamlines. BM26A (soft branch) and BM26B (hard branch)

The collimating mirror on the soft branch is installed early 2012. On both beamlines the focusing mirrors and monochromators are installed.

	BM26A	BM26B	unit
Energy range	4 – 42	5 -30	keV
Maximum Flux	> 10^{12}	> 10^{12}	ph/sec
Spot size			
Normal	0.3 x 1	0.3 x 0.3	mm^2
Microfocus 1	8	12 (KB system)	μm^2
Microfocus 2	-	10 (CRL)	μm^2
$\Delta E/E$	1.8 x 10^{-4} (with collimating mirror)	2.4 x 10^{-4}	

Appendix 2 Industrial relevance of DUBBLE

The group of Prof. Joke Bouwstra (Simon Stevin meester 2005) has received over 3.5 M€ to perform studies on the structure of skin and eczema. For five PhD students funded by either STW or industrial funds crucial parts of their thesis depended on data collected at the DUBBLE beamlines and patents have been obtained. Industrial partners that can be named are Unilever, River Diagnostics and Leo-Pharma (Denemark).

The group of H.E.H. Meijer/ G.Peters has received STW and DPI funding for 7 postdocs/PhD whose work hinged on DUBBLE data. Projects ranged from flow induced polymer crystallisation to materials behaviour under mechanical deformations. Major industrial partners in these projects included BASF, Borealis, Braskem, Dow Benelux, DSM, Exxon Mobil, LyondellBasell, Sabic Europe, Sabic Innovative Plastics, Teijin Wavin Technology & Innovation, Moldflow (*Victoria, Australië),* Kiwa Gas Technology, Intertek Polychemlab (Geleen, The Netherlands), IME Technologies, Purac Biochem. So far the Dutch Polymer Institute has recognized that and funded in total 12 postdoc and 4 student years worth of funding to assist in the further development of the polymer processing activities on DUBBLE

In the group of B. Weckhuysen (Utrecht) around 40% of the postdocs/PhD positions is funded private-public or private. The private-public funding route consists of M2i, ACTS en CatchBio initiatives whilst the industrial partners, interested in processing catalyst include Shell, ExxonMobil, Dow Chemicals, BASF, SK Energy, Albemarle Catalysts, Total, Sumitomo and Rhodia. Besides this there are companies specialized in biomass conversion like Avantium en Croda and the automotive industries like BASF, DAF en Toyota. A crucial part of the research here depends on the unique possibilities at DUBBLE to combine different techniques on-line. Around 10 students of this group have collected important parts of their thesis on DUBBLE.

The group of Kelder (Delft) has been instrumental to obtain funding for a postdoc to be placed at DUBBLE via the European private-public Alistore network (http://www.alistore.eu). The main activity will be the development of Li-ion battery for traction purposes. Industrial partners include Volvo, Renault and Spijkstaal (car manufacturers) and GAIA AkkumulatorenWerke (battery manufacturer).

The Energy Centre Netherlands (ECN) has used DUBBLE in the framework of an EU FP7 project in which Air Products and BP are major industrial partners for applications and process scale-up.

Several other groups have also acknowledged industrial funding for DUBBLE related research. Companies involved in this are among else Tata Steel Europe, Ocas - Arcelor Mittal, SKF, Nedschroef, Ovako wire, Kodak, OCE, Jansen Pharmaceuticals, Loreal.

Only in exceptional cases does one contemplate to directly sell beam time to industries. The minimal income, the complicated scheduling and the loss of publishable output are the main reasons. The former is the main parameter upon which the DUBBLE project is assessed by the funding agencies.

This text is a brief overview and certainly not complete. Only some representative user groups have been asked for information.

Appendix 3 SWOT analysis

The SWOT[9] analysis is a standard part of the review procedures of the Dutch research council. This section is taken from the 2011 NWO/FWO review and it might be informative to obtain some insights into the funding framework from NWO/FWO as well. It might overlap to some degree with previous chapters.

The strengths of the DUBBLE project

- The limited size and stability of the user community allows investment in experimental infrastructure tailored for this Dutch-Flemish user community. This is in contrast to comparable ESRF beamlines which have to satisfy a much larger and more fluctuating user group.
- Worldwide there is a shortage of XAFS and SAXS/WAXS beamlines. Due to the rather large dependence of the Dutch and Flemish economy on food and chemical industries and the increasing importance from nanotechnology in general these are techniques that are in high demand from many groups within Belgium and the Netherlands associated with the academic research relevant for these industries. DUBBLE is well poised to address this demand.
- The existing infrastructure allows for the rapid interfacing of complicated sample environments and additional experimental techniques which makes experiments feasible which cannot be carried out anywhere else unless with great difficulty. The qualitatively very high level catalysis experiments are a good example of this as well as the pioneering role in the introduction in pulsed high magnetic fields.
- Both beamlines have been upgraded in the period 2007-2011 to a level that will allow a high level of experiments for the coming 7 – 10 year. Both SAXS/WAXS as well as XAFS are very well matched to the ESRF bending magnet spectrum.

The weaknesses of the DUBBLE project

- The small amount of user support staff does not allow a high level of ‘user aftercare’ for a broad user group and only limited time for training. This sometimes results in collected data sets not being used since users lack the experience to handle these. In time, with user groups gaining more experience, this problem is slowly reducing but at times it proves a barrier for new groups.
- The fact that DUBBLE is not embedded in a ‘home institute’ has as a consequence that it is difficult to find and retain staff. There are no promotion possibilities and it is more difficult to set up larger scale collaborations and have access to sample preparation facilities. The latter is an obstacle for the in-house research of younger (non-permanent) staff.
- Both beamlines are used heavily and successfully for the two major techniques SAXS/WAXS and XAFS. Interface Diffraction has hardly been used in the period 2007-2011 (although the diffractometer itself might play a role in newly developed GISAXS experiments). A rejuvenation of this technique will be at the expense of the SAXS user program and it is

[9] SWOT analysis (alternatively SWOT Matrix) is a structured planning method used to evaluate the Strengths, Weaknesses, Opportunities, and Threats involved in a project or in a business venture.

doubtful if this is useful in the light of the availability of ID on other ESRF beamlines. It will also require additional staff with sufficient expertise in this field. (This in addition to the extra staff that will be required to operate the two main techniques)

- Most of the expertise within DUBBLE on the operation of the beamlines, the research landscape suitable for the DUBBLE beamlines and insight in the feasibility for further development is vested in two persons. One of these, after 15 years service, still does not have a permanent position. This is a very fragile basis.

Threats

- The financial situation in the last years has been satisfactory apart from the fact that there is too little budget for extra staff available. This poses a threat to initially the user service but ultimately to the durability of the project. Also it can be foreseen that budget adjustments for inflation, which have not been made in 10 years, should be implemented.
- The changes within the ESRF, due to the ‘Upgrade Program’ can possibly start to pose a problem when the plans for the new machine, if implemented, would reduce the horizontal acceptance angle of the beamline. At present we are using a 9 mrad front end in a 2+5+2 configuration, in which the 5 mrad represents the ‘dead angle’ between the two beamlines. It is impossible to reduce this. Similar problems would occur if the whole beamline, including hutches, would have to move.

Strategy for the next period

At present DUBBLE is well placed to serve the demands of the existing user group and can provide excellent data quality and, when compared internationally, at a very high level of experimental sophistication. The output of the beamlines is also at a very high level and still increasing. Since around 50% of the Dutch-Belgian SR based material science publications are being generated at the DUBBLE beamlines one can conclude that nationally the existence of DUBBLE is quite relevant.
The strategy for the coming years:

- Keep gradually improving the infrastructure with respect to microfocus, detectors, beam stability without endangering the existing user program.
- Sample environments are the strong point of many of the successful user projects. We should keep investing and continue developing these.
- Stimulate data analysis workshops in order to improve the expertise in the existing user community. Further develop data analysis methods in collaboration with other SR laboratories.
- Try gradually to bring in new user groups and provide sufficient support to initiate and train these users in the use of the experimental techniques by either collaborations with staff members or to arrange that these groups are being supported by existing groups via collaborative projects.
- Try to interest researchers from areas so far not or hardly present at DUBBLE. Growth area here could be for instance biophysics.

With regard to the desirability of the introduction of new techniques on DUBBLE this is not a decision that could or should be taken at the level of DUBBLE. DUBBLE is part of the wider Dutch-Flemish X-ray infrastructure. At the national level an inventory should be made what is present and what is required. This not only with the existing user community in mind but also in the perspective of international developments and interests of user groups traditionally not using SR. This is too broad a task for the DUBBLE staff on its own and should be organized by the policy departments of the research councils.

Appendix 4 Journals

In the table below the journals in which data from DUBBLE has appeared in the period 2007-2012 are given , as well as the journal impact factor and the number of publications in that journal (counts). For the review journals only those that contain a very substantial amount of DUBBLE material are given. The number weighted average impact factor, excluding the three review journals, is 5.76[10]. The complete information regarding titles and authors can be found on: http://www.esrf.fr/UsersAndScience/Experiments/CRG/BM26/Publications

Journal	I. Factor	Counts	BM26A	BM26B
			(EXAFS)	(SAXS)
NAT MATER	36.732	1	1	
Chemical Society Reviews	28.76	3	3	
Accounts of Chemical Research	22.5	1		1
Advanced Materials	13.87	3		3
Angewandte Chemie-I. E.	13.45	8	8	
Advanced Functional Materials	10.17	4		4
J American Chemical Society (JACS)	9.9	6	3	3
Small	8.34	1	1	
Journal of Controlled Release	7.52	1		1
CHEM SCI	7.545	2	1	1
Physical Review Letters	7.37	7	2	5
Chemistry of Materials	7.2	9	2	7
ChemSusChem	7.17	1		1
Green Chemistry	6.76	1		1
Journal of Investigative Dermatology	6.31	2		2
Journal Catalysis	6.2	11	10	1
Chemical Communications	6.16	6	3	3
Carbon	6.008	1		1
Journal of Materials Chemistry	5.99	6	3	3
Analytical Chemistry	5.98	2	2	
Chemistry - A European Journal	5.92	2	1	1
Env Science and Technology	5.76	9	9	
Journal of applied Crystallography	5.65	10		10
BIOMACROMOLECULES	5.646	1		1
J LIPID RES	5.559	1		1
BIORESOURCE TECHNOL	5.352	1		1
CHEMCATCHEM	5.207	4	3	1
Macromolecules	5.16	29		29
Electrochemistry Communications	5.15	1		1
Journal of Physical Chemistry C	5.04	14	5	9
Faraday Discussions	5	1		1
Soft Matter	4.99	9		9
Crystal Growth & Design	4.87	3	1	2
Inorganic Chemistry	4.6	4	3	1

[10] Marion Glueckert (CRG office) is thanked for maintaining the data base and providing the information in this table.

Geochimica Et Cosmochimica Acta	4.58	3	2	1
Langmuir	4.51	20	2	18
Journal of Membrane Science	4.31	1		1
Acta Materialia	4.22	2	1	1
Biophysical Journal	4.13	2		2
Biochimica et Biophysica Acta-Biomembranes	4.11	4		4
CHEM GEOL	4.063	1		1
Journal of Physical Chemistry B	4.06	12	4	8
ELECTROCHIM ACTA	4.039	1		1
Polymer	4.009	9		9
Journal of Power Sources	4.006	1	1	
CARBOHYD POLYM	3.987	1	1	
Applied Catalysis A-General	3.96	2	2	
Physical Chemistry Chemical Physics	3.93	9	6	3
Journal of Polymer Science part a-Polymer Chemistry	3.91	3		3
Dalton Trans.	3.88	2	1	1
Journal of Structural Biology	3.8	0		
ATMOS ENVIRON	3.742	1		1
Physical Review B	3.69	3	1	2
Catalysis Today	3.58	4	2	2
ChemPhysChem	3.55	3	2	1
EUR J PHARM SCI	3.536	1		1
J COLLOID INTERF SCI	3.263	5	4	1
MICROPOR MESOPOR MAT	3.23	2	1	1
J of Analytical Atomic Spectrometry	3.22	4	3	
Journal of Chemical Physics	3.149	1		1
EUR POLYM J	3.058	1	1	
EUR J INORG CHEM	3.049	1	1	
TOP CATAL	2.973	3	3	
CHEM PHYS LIPIDS	2.647	1		1
APPL GEOCHEM	2.626	2	2	
J SOLID STATE CHEM	2.412	1		1
MATER CHEM PHYS	2.385	1	1	
J SYNCHROTRON RADIAT	2.33	1	1	
MACROMOL MATER ENG	2.222	2		2
Eur J for Lipid Science and Technology	2	2		2
J ELECTRON SPECTROSC	1.958	1	1	
Thin Solid Films	1.88	2		2
LIQ CRYST	1.858	2		2
ADV ENG MATER	1.688	1	1	
total		279	105	173

The median impact factor, excluding the three high impact review journals, is 5.1