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PROCEEDINGS

Swift ion beams for solid state and materials science

Andrea Denker,
Wolfgang Bohne,
Jürgen Heese,
Heinrich Homeyer,
Heinz Kluge,
Sven Lindner,
Jörg Opitz-Coutureau,
Jörg Röhrich,
Erik Strub

Abstract Ion beams are unique tools in modern science and technology. They are used for the analysis and modification of materials and are applied in medicine and technology. In cancer therapy fast protons allow a precise tailoring of the radiation field to the tumour, thus maximising the tumour control probability and simultaneously reducing the risk of side effects. Modification of the structure of solids by ion irradiation results in local structures on nano-scale, e.g. high-tech filters having defined pore numbers and pore sizes are produced by high-energy heavy ion irradiation with consecutive etching. Electronic devices utilised in areas with high radiation level have to be tested for their radiation hardness. The devices are irradiated with accelerated ions to receive the same dose by high-energy ions as expected during their lifetime. For materials sciences the analysis of composition and structure of solids is of uppermost importance. Complex layered structures are analysed by ERDA (Elastic Recoil Detection Analysis). Investigation of art and archaeological objects has to be non-destructive. PIXE (Proton Induced X-ray Emission) allows elemental analysis without sampling of the object. Different applications of high energy ions will be presented.

Key words tumour therapy • materials modification • materials analysis • single event effects • technical applications • cyclotrons

A. Denker✉, W. Bohne, J. Heese, H. Homeyer, H. Kluge,
S. Lindner, J. Opitz-Coutureau, J. Röhrich, E. Strub
Ionenstrahllabor,
Hahn-Meitner-Institut,
100 Glienicker Str., D 14109 Berlin, Germany,
Tel.: +49 30/ 8062 2498, Fax: +49 30/ 8062 2097,
e-mail: denker@hmi.de

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Introduction

In the past, accelerator laboratories were mainly dedicated to nuclear physics. But new, up-coming, and promising fields of work increasingly use ion beams from accelerators for their purposes. These fields cover all kinds of materials analysis and materials science as well as biology or archaeology. Practical applications have been developed, involving hospitals and industry in the use of accelerators. In the low energy regime more than 8000 dedicated small accelerators [1] have been installed world-wide for ion implantation, Rutherford back scattering analysis (RBS), Proton Induced X-ray Emission (PIXE), or Accelerator Mass Spectrometry (AMS) among other themes. In addition, there is an increasing demand of solid state and medical physics as well as materials science for high energy ions with energies above 1 MeV/u especially for heavy ions.

Many of the ion beam applications and technologies profit from the excellent beam quality and beam variety delivered by cyclotrons. For some applications like tumour therapy or materials modification the short pulses with high frequency of a cyclotron represent a quasi-continuous beam. Other applications deliberately use the time structure of the pulsed beam. The vast choice of ions available from cyclotrons – from hydrogen up to uranium – is of high interest for materials analysis and radiation effects.

Tumour therapy

Proton beam radiotherapy is worth considering when a healthy tissue with a high complication risk is very close to the target volume. It is a world-wide growing field.

In recent years, many therapy facilities have been designed and installed and 23 are active at the moment [24]. It is worth noticing that approx. 70% of them are using cyclotrons as accelerators. Among these facilities, 20 operate with protons, three use heavier ions. The dynamics of this field is reflected in the proposals for 20 new centres, and at least half of them will be equipped with cyclotrons. The development of compact 230–250 MeV proton cyclotrons for clinical proton radiotherapy reflects the technical advantage of cyclotrons [6, 16]. The advantages of proton therapy over conventional radiotherapy are illustrated in Fig. 1.

Regular news and semi-annually updated numbers of patients may be found on the web page of the particle therapy co-operative group PTCOG [15].

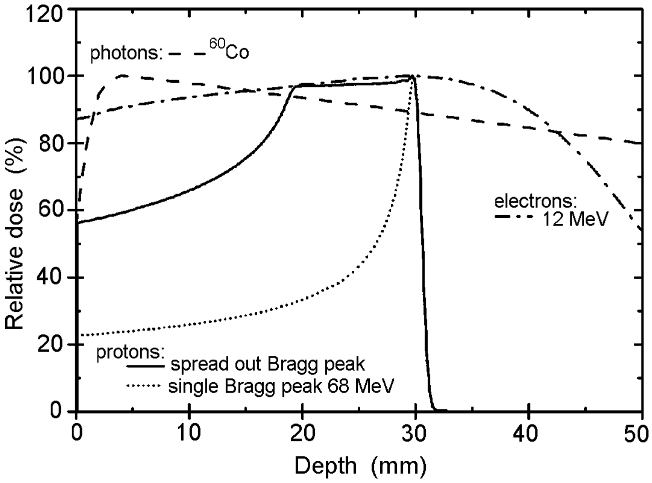

Fig. 1. Dose-depth distribution for different types of radiation. Electron (dashed-dotted line) and γ-radiation (dashed line) are widely dispersed into the tissue in front of and behind the tumour, while the dose of protons is restricted to a very narrow area, the so-called Bragg peak (dotted line). By superimposing many proton beams of different energy a homogeneous dose distribution over the tumour depth, the so-called spread-out Bragg peak (solid line) can be achieved.

Materials modification

Fast heavy ions deposit more energy locally in a shorter time than can the most powerful lasers currently available. The extremely high energy deposition along the flight path of an ion can destroy chemical bonds, so that permanent materials modifications are left in its track. These tracks may be conducting tracks in an isolating matrix. Thus the created changes of material on a nano-scale may lead to flat screens or nano-electronics. The achieved different optical properties of solids are already exploited in the production of picosecond laser diodes or used to increase the data transfer rate of optical fibres.

When ion tracks in polymer foils are etched, the etching rate along the track is up to three orders of magnitude higher than in the surrounding material [2], which enables the formation of pores. The number of the incoming ions settles the number of holes created, whereas the size of the pores can be determined precisely by the etching time, solution, and temperature. In addition, using fast heavy ions from an accelerator, one has a good control of allocation and direction of the ion tracks. This is an advantage compared to irradiations using nuclear fission products.

Materials modification is performed at many laboratories mainly for research purposes [2, 3, 18, 22, 23]. At some places, however, the irradiation of polymer foils in order to produce filters is already performed on an industrial base. These filters, for instance, are applied in the semiconductor industry for clean rooms, for medical purposes, and as cell collectors for microbiology as well as membranes for fuel cells [9, 27].

All materials modifications profit from the stability, intensity and homogeneity of the heavy ion beam available at a cyclotron.

Radiation effects

When a high-energy ion passes through an electronic device, the ionisation in the track can destroy the device or switch the status of it. Each measurable effect on an electronic device produced by an impinging ion is called Single Event Effect (SEE). High-energetic ions are part of cosmic radiation, solar wind, and radiation belts of the earth. Especially, the electronics in aircrafts and satellites is subject to this radiation [10]. Owing to cosmic showers, also electronic devices operated on ground level may suffer from these effects [19]. The radiation hardness of the devices can be tested by ion irradiation, which simulates the operation conditions. Thereby, the irradiated devices receive the same dose as they would receive during their operation lifetime. By these experiments one gets information if the device will survive the irradiation or which degradations are expected to occur.

Irradiation set-ups exist in large vacuum chambers as well as in air, the latter one being mainly used for protons [8, 11–14, 28]. On the one hand, there is the growing number of satellites used for telecommunication or weather forecasts, on the other hand, modern electronics becomes miniaturised and, hence, much more sensitive to radiation effects. These two facts lead to an increasing demand of radiation-hardness tests. In addition, the effects of the high levels of radiation to which astronauts will be exposed in manned space-flight, can be studied [17]. Under controlled conditions, the interaction of fast ions with living cells or tissues is investigated, giving an interdisciplinary link to medical applications.

Depending on the operation conditions, light ions up to heavy ions over a broad energy range are required for these tests. The possibility of cocktail beams, a mixture of various ions with the same charge to mass ratio, available at many cyclotrons in the world, offers fast switching between different ions of less than one hour. Hence, the strongly varying effects in the devices under test, created by different ions having different energy losses, can be studied in a time-effective way.

Materials analysis

ERDA – Elastic Recoil Detection Analysis

In materials research, it is of crucial importance to determine the elemental composition and structure of solids. A well suited tool for this purpose is the Elastic Recoil Detection Analysis (ERDA). For the ERDA-method the samples are irradiated with high energetic heavy ions at grazing incidence. The energy as well as the number of the outscattered atoms (recoils) of the sample components are measured at a fixed angle relative to the beam direction. Since the scattering probability is given by the Rutherford cross section and all the experimental parameters are known, ERDA is a standard free absolute method. Owing to the computable element specific energy loss in material, it is possible to calculate the concentration depth profiles from the measured energy spectra, simultaneously for all components of the sample. The detection sensitivity is almost the same for all elements. For hydrogen the sensitivity is even enhanced by a factor of four. When using heavy projectiles no restrictions of the detectable mass exist.

ERDA can be found at numerous accelerator laboratories (e.g. Tandem-Labs in Munich, Canberra and Rossendorf). Two different techniques are applied for the identification of the recoils: i) the energy loss and the energy of the recoils are measured with two different detectors; ii) the energy and the time-of-flight (TOF) of each recoil on a fixed flight path is determined. The latter technique profits in particular from the time structure of pulsed ion beams from cyclotrons.

PIXE – Proton Induced X-ray Emission

For the Proton Induced X-ray Emission (PIXE) the interaction between the protons and the electrons of the sample atoms is exploited. The irradiation of any material with a proton beam induces the emission of characteristic X-rays, which identify the elements in the material, independently of their chemical bonding. Sometimes PIXE also stands for Particle Induced X-ray Emission, but today, mostly proton beams are used. For analytical purposes, only X-rays above about 2 keV, i. e., from elements heavier than phosphorous, can be used due to the absorption of the X-rays in the sample itself and in the entrance window of the detector, and in the air between sample and detector, if the measurements are performed in normal atmosphere. PIXE is performed at many places world-wide with protons of about 3 MeV. The use of higher proton energy increases the analytical depth due to the larger range of protons and larger cross sections for hard, penetrating X-rays. High-energy PIXE in vacuum for heavy element analysis was used in South Africa [21] and Canada [29] using high energy protons from the cyclotron.

Applications at ISL

Treatment of ocular melanomas

At the HMI, between 1995 and 1998 a therapy unit for proton beam treatments of ocular melanomas was built in collaboration with the university hospital Benjamin Franklin. The first patient was treated in June 1998. The HMI eye treatment facility is the only German proton therapy facility and schedules ten treatment weeks per year. A homogeneous proton dose over 40 mm diameter at the treatment position is guaranteed by a passive scattering system. In order to keep the beam position stable and to generate a homogeneous source, the beam is focussed onto a 50 μm thick tantalum scattering foil where a Gaussian angular distribution with approx. 12 mrad half-width is generated. The beam axis is defined by two narrow slit pairs and a circular collimator. A fast-acting beam shutter is located close to the scattering foil. The small beam diameter at this position guarantees fast beam switching times of less than 30 ms. The beam passes a 200 magnet and exits the accelerator vacuum 150 cm upstream from the treatment position through a 50 μm thick Kapton window. The beam range is continuously adjustable by a plexiglas wedge. Time-averaged uniform depth-dose distributions are generated by a rotating modulator wheel. Proton irradiation fields with distal 90–100% dose fall-offs of 1.0 mm in water and a lateral penumbra of approx. 2.5 mm are available at the treatment position.

Figure 2 shows an outline of the therapy unit. Special needs and techniques for a very precise patient positioning for proton beam treatments of the eye are evident. The positioning system consists of a treatment chair and two horizontal, orthogonal X-ray systems, one aligned with the proton beam axis and the other one perpendicular to the beam. Both systems use a fixed mounted X-ray tube and a retractable image intensifier. The patients are immobilised in sitting position on the treatment chair with individually fabricated bite blocks and face masks. Eyelid retractors are used to limit the irradiation of eyelids. To obtain an optimal eye orientation for the treatment, the patient has to look to a fixation light. Axial and lateral radiographs are taken simultaneously. Digital image processing allows corrections of image distortions and automatic detection of the tantalum clip coordinates with an accuracy better than ±0.2 mm. The reference positions calculated by the treatment planning program can be superposed with the current radiographs. The treatment chair allows precise movements of the patient with a position reproducibility <0.1 mm. The patient is moved with the chair until an optimum agreement between the actual clip positions on the radiographs and their reference positions is achieved. The tumour eye is observed in the treatment position by an infrared sensitive TV camera. A typical irradiation dose of 15 Gy is applied within

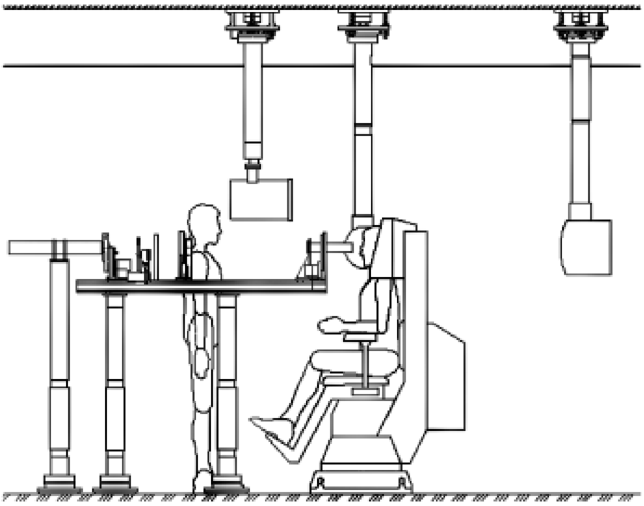

Fig. 2. Schematic set-up of the HMI eye treatment unit.

30 s. However, the patient alignment for therapy takes about 10 min.

In nearly five years of operation, more than 300 patients with ocular tumours have been treated at the HMI facility. The majority (75%) of the pathologies are uveal melanomas. All patients mostly show small and medium-sized tumours in a critical location to optical disc and/or macula. Other indications treated are uveal hemangiomas (12%), iris melanomas (10%) and conjunctival melanomas (3%).

After nearly five years of patient treatments, the HMI eye facility has shown a very good technical reliability. No problems were encountered with the realisation of the treatment unit. The follow-up results of patients treated during the first two years of operation look very promising.

Filter production by heavy ion irradiation

At the ISL, the interaction of fast ions with solids is one of the main research topics, ranging from the study of ultrafast dynamics to examinations of the applicability of non-equilibrium thermodynamics. One of the practical applications is the irradiation of polymer foils on an industrial level (see Fig. 3).

The requests for industrial production are far more challenging than for experimental purposes: first of all, strictly keeping to the beam schedule becomes an essential issue. Second, the beam stability is of crucial importance, as it determines the homogeneity of the pore density of the foils. Going from irradiation of some 100 cm^2 for research purposes to real production involves the irradiation of many square metres, meaning a change of several orders of magnitude in irradiated area. To be time-, manpower-, and cost-effective, the homogeneity of the foils has to be constant over these huge areas, involving excellent long and short term beam stability for fast, heavy ions. In addition, the beam intensity has to be high, to keep the irradiation times reasonable as needed fluences range from 10^6 to 10^8/cm^2. Cyclotrons are perfectly suited to fulfill all these demands, but beam tuning has to be done very carefully. At ISL, many efforts have been made to achieve high beam stability on short as well as on long terms (see Fig. 4).

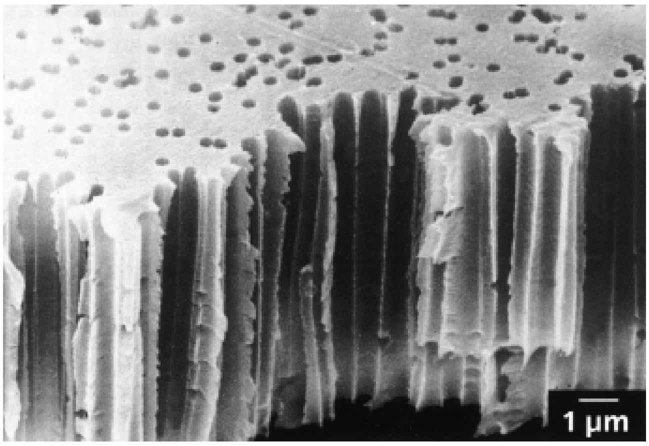

Fig. 3. Photograph of a foil, taken with a scanning-electron microscope after the etching procedure, showing the entrance holes of the ions in the surface. At the edge the channels are visible.

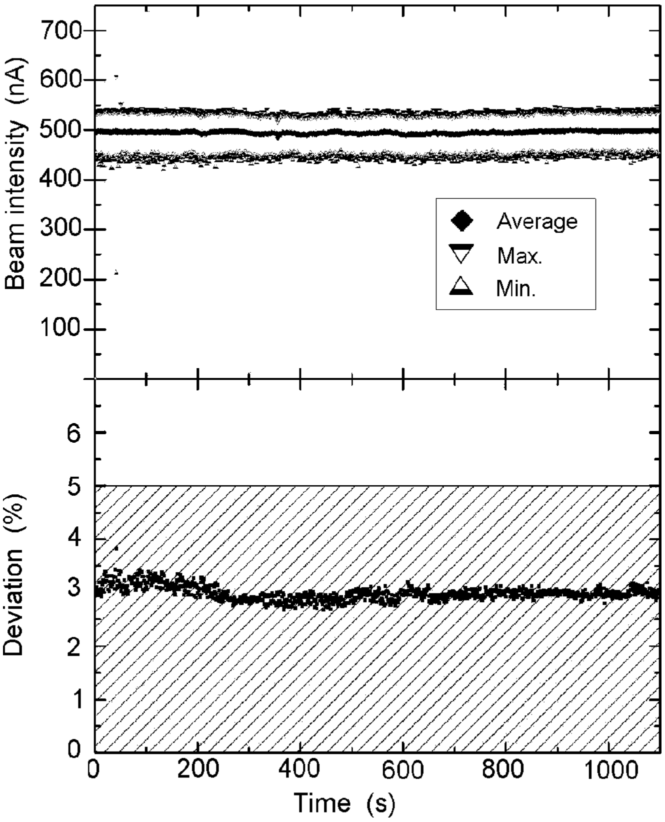

Fig. 4. Beam stability measured on the last Faraday cup in front of the target station of the ISL cyclotron of a high-energy heavy ion beam. The squares in the upper part represent the average beam current at a time interval of 1 s, whereas the triangles display the worst minimum and worst maximum values for each interval. The sampling rate was 1 kHz. The lower part shows that the standard deviation (one sigma) for each point is less than 5%.

Single event effects

At the moment, ISL provides two high-energy target stations in normal atmosphere, where satellite components or electronic devices can be tested for their radiation hardness. The following three examples demonstrate the broad range of these tests: complete modules like the ABRIXAS star camera [20] were irradiated, yielding the information if they can be accepted for their mission. Detectors, like the Dosimetry Telescopes (DOSTEL), used for manned space-flight [7] were calibrated. Single electronic devices like high-power diodes [25] were tested with the aim of performance improvement.

A challenge of these experiments is their required low beam intensity of about 100 particles per second with precise on-line dosimetry. Several low transmission grids in front of the cyclotron attenuate the beam, already reduced to 10^9 particles/s by slits, to the required values.

ERDA

At ISL, the element respectively to mass identification is done by means of the TOF method, i.e. the coincident measurement of energy and flight time for each recoil [4]. With this set-up a mass resolution of one unit up to about A = 50 is achieved. Since the detection efficiency of the used channelplate detectors is less than 100% for protons and alpha particles, also the RF signal from the cyclotron is used for the TOF measurement. With a pulse width of about 0.5 ns it is possible to resolve light elements with an efficiency of 100% even for hydrogen. The detection limit is of the order of 0.001 at.% for all elements. The applied energy of the heavy projectiles of about 1.5 MeV/u enables depth profiling up to a maximum layer thickness of several micrometers with a resolution of less than 20 nm at the surface. In the last years, hundreds of samples from various fields, like semiconductors, metals, and in a few cases polymers and teeth, were analysed routinely. The main focus is the study of photovoltaic materials, mostly produced at the photovoltaic department of the HMI. Figure 5 shows the TOF *vs.* energy graph of all detected recoils, the so-called scatterplot, of such a sample. The measurement was performed with a 350 MeV gold beam. All components are clearly resolved down to a depth of about 3 µm. The good mass resolution enables the determination of the stoichiometry of all layers. The iodine contamination originates from the deposition technique of the ZnO window layer.

High-energy PIXE

At the ISL, a 68 MeV proton beam in air is used [5] for the PIXE measurements. The range of these protons in the irradiated object is up to a few centimetres, offering the fairly unique possibility of a non-destructive analysis to this depth. 68 MeV protons also induce for the heaviest elements the emission of K X-rays with large cross sections. These high energy X-rays are weakly absorbed and, therefore, can be also detected if they are produced deep inside the sample. The absorption of the X-rays on their way out from deep inside of the studied object, depending on the composition and any layer structure of the sample, makes a quantitative analysis of the elements difficult if a priori no information about the sample is available. Nevertheless, some information on the depth, where an element is present, can be gained from the measured intensity ratios of the various K and L lines of this element. Their absorption in the sample differs markedly because of their different energy and depends, therefore, on the depth of their production. Regarding the chemical composition, the sensitivity limit of the concentration is 0.1% down to a few ppm, depending on the element and sample composition. Due to the high cross sections, only low beam intensity is needed, so that the analysis is non-destructive, an important feature when analysing art or archaeological objects.

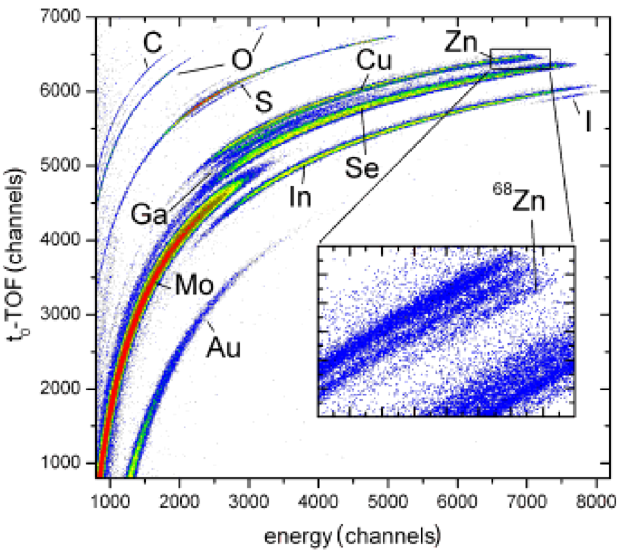

Fig. 5. Scatterplot for an ERDA measurement of a complicated multilayer structure [glass/Mo/($Cu_xIn_yGa_z$)(Se,S)$_2$/ZnO] at a detection angle of 40° obtained by using 350 MeV gold ions.

Fig. 6. Chinese bowl after the analysis. The age of this bowl was not certain, as two contradictory art historian reports are existing.

Figure 6 shows a large Chinese bowl having a diameter of 38 cm and a weight of 4.8 kg. One art historical report dates the bowl back to the Jihan era, Ming dynasty (1522–1566). Another report says that shape and decoration were copied from the Wanli time, Ming dynasty (1573–1620), but that the bowl itself has a maximum age of about 100 years. The age of the bowl is of fundamental importance for its value. Thermoluminescence does not work with porcelain, hence, only indirect dating is possible. One possibility is the analysis of the characteristic elements in the colours used for the decorative painting, because their recipes vary over time [26]. For instance, green colours were based on copper before the middle of the 19th century. Only later chromium containing colours became available.

Porcelain is extremely radiation sensitive, the low beam intensity needed for high energy PIXE due to the high cross sections insures a non-destructive analysis. Figure 7 shows the results from the yellow colour compared to the white porcelain. The yellow colour contains zinc and lead, but no antimony. The missing of antimony is a clear indication for the manufacturing time: all recipes up to about 1850 name Zn combined with Fe and Sb, the amount of Sb_2O_3 being two times higher than ZnO. Zinc and iron only were applied as a yellow pigment not until modern times. The report stating a maximum age of the bowl of about 100 years could, therefore, be confirmed.

Conclusions

The presented overview can be only a brief outline, as there are tremendous possibilities of applications with fast ions.

Starting new applications offer challenging and interesting fields for the use of cyclotrons. The user community becomes more and more manifold, ranging from art

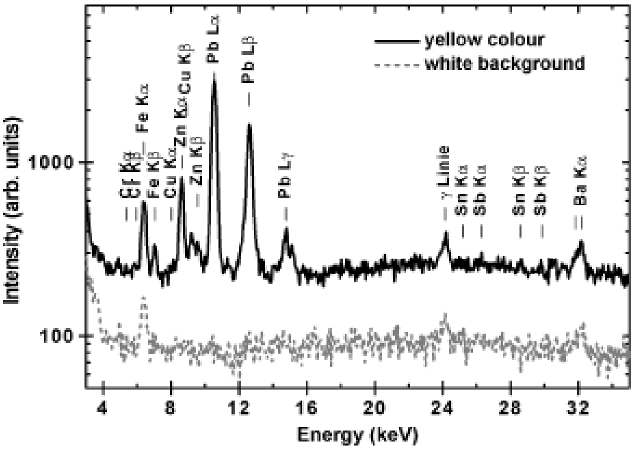

Fig. 7. High-energy PIXE spectra of the yellow colour (solid line) and the white porcelain (dashed line) of the Chinese bowl. For better visibility the spectra are scaled to arbitrary units. In the yellow colour, Zn and Pb are visible beside the elements already visible in the white porcelain.

historians over physicists and semiconductor specialists to physicians. The various experiments require a dedicated beam preparation covering strong variations concerning ion type, ion energy, and beam intensity. Industrial collaborations and tumour therapy have high demands on the reliability and stability of the machine, whereas, the other applications and experiments using the accelerator will profit from the efforts undertaken to improve beam stability and intensity.

Many of the practical applications request usually only part of the beam time of a large accelerator, but beside the proposals for dedicated proton accelerators for cancer therapy there exists already a proposal of a specific accelerator only for filter production, demonstrating the increasing requests in this field. Ion beam applications are becoming more and more important for manufacturing and technology of every day products.

References

1. Amaldi U (2000) The importance of particle accelerators. Europhysics News 31;6:5–9
2. Apel P (1996) Polymeric materials research with cyclotrons. In: Cornell JC (ed.) Proc of the 14th Int Conf on Cyclotrons and their Applications, 8–13 October 1995, Cape Town, South Africa. World Scientific, Singapore, pp 136–143
3. Balanzat E, Betz N, Bouffard S (1995) Swift heavy ion modification of polymers. Nucl Instrum Meth Phys Res B 105:46–49
4. Bohne W, Röhrich J, Röschert G (1998) The Berlin time-of-flight ERDA setup. Nucl Instrum Meth Phys Res B 136/138:633–637
5. Denker A, Maier KH (2000) Investigation of objects d'art by PIXE with 68 MeV protons. Nucl Instrum Meth Phys Res B 161/163:704–708
6. Flanz JB, Bradley SG, Goitein M *et al.* (1999) Initial equipment commissioning of the northeast proton therapy center. In: Baron E, Lieuvin M (eds) Proc of the 15th Int Conf on Cyclotrons and their Applications, 14–19 June 1998, Caen, France. IOP, Bristol, p 319
7. Gericke C, Kopp J, Beaujean R, Küchler S (2000) Calibration of Si-detectors for dosimetry in space. Annual Report HMI B 567:101
8. Hajdas W, Adams L, Nickson B, Zehnder A (1996) The proton irradiation facility at the Paul Scherrer Institute. Nucl Instrum Meth Phys Res B 113:54–58
9. Hayashi AM (1998) Taking on the energizer bunny. Scientific American 4:286–292
10. http://RADHOME.gsfc.nasa.gov/radhome/papers/seecai.htm
11. http://www.cyc.ucl.ac.be/HIF/HIF.html
12. http://www.cyc.ucl.ac.be/PIF/PIF.html
13. http://www.cyclotron.tamu.edu/ref/
14. http://www.ganil.fr/research/interdisciplinarity/industry/index.html
15. http://www.neurosurgery.mgh.harvard.edu/hcl/ptles.htm
16. Jongen Y, Beeckman W, Cohilis P (1996) The proton therapy system for MGH's NPTC: equipment description and progress report. Bull Cancer Radiother 83S:219s–222s
17. Kiefer J (2001) Space radiation research in the new millenium – from where we come and where we go. Phys Medica 14;S1:1–4
18. Krauser J, Hoffmann V, Harneit W *et al.* (2001) Graphitic nanowires embedded in diamondlike carbon films. Proc AIP 591:507–510
19. Maier KH, Denker A, Voss P, Becker HW (1998) Single event burnout of high-power diodes. Nucl Instrum Meth Phys Res B 146:596–600
20. Meinert D, Fritzke K, Bischof M, Popow E, Luebke-Ossenbeck B, Maier KH (1999) Strahlungstest der ABRIXAS Sternkamera. Annual Report HMI B 563:74
21. Pineda CA, Peisach M (1990) Prompt analysis of heavy elements by high-energy-induced (p,X) reactions. Nucl Instrum Meth Phys Res A 299:618–623
22. Schiwietz G, Luderer E, Grande PL (2001) Ion tracks – quasi one dimensional nanostructures. Appl Surf Sci 182:286–292
23. Schwartz K, Trautmann C (2001) Heavy ion-induced damage and modifications of insulating materials. Proc of SPIE 4415:248–259
24. Sisterson JM (2001) Ion beam therapy: overview of the world experience. In: Duggan JL, Morgan IL (eds) Proc of the 16th Int Conf Application of Accelerators in Research and Industry, 1–4 November 2000, Denton, TX, USA. AIP, Denton, CP576:865–868
25. Soelkner G, Voss GP, Kaindl W, Wachutka G, Maier KH, Becker HW (2000) Charge carrier avalanche multiplication in high-voltage diodes triggered by ionizing radiation. IEEE Trans Nucl Sci 47;6:2365–2372
26. Speel E, Bronk H (2001) Enamel painting: materials and recipes in Europe from c. 1500 to c. 1920. Berliner Beiträge zur Archäometrie 18:43–100
27. Spohr R (1990) Ion tracks and microtechnology. Vieweg Verlag, Braunschweig
28. Virtanen A, Hyvönen J, Ranttila K, Rekikoski I, Tuppurainen J (1999) Heavy ion and proton test site at JYFL accelerator laboratory. Nucl Instrum Meth Phys Res A 426:68–71
29. Yeo YH, McKee JSC, Smith GR *et al.* (1990) Commissioning and characterization of the Manitoba automated high energy proton microprobe. Nucl Instrum Meth Phys Res B 49:430–434