

Technical University of Denmark



## Radiation Research Department annual report 2002

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**Radiation Research  
Department  
Annual Report 2002**

**Edited by B. Majborn, A. Damkjær and S.P. Nielsen**

**Abstract** This report presents a summary of the work of the Radiation Research Department in 2002. The department's research and development activities are organized in two research programmes: "Radiation Physics" and "Radioecology and Tracer Studies". In addition the department is responsible for the task "Dosimetry". Lists of publications, committee memberships and staff members are included.

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# 1 Introduction

The research of the department includes dosimetry, optically stimulated luminescence, nuclear emergency preparedness, contamination physics, reactor safety, radioecology, and tracer and isotope techniques.

In 2002 the department was organised in two research programmes: “Radiation Physics” and “Radioecology and Tracer Studies”, in addition to the task “Dosimetry”, which includes personnel dosimetry and industrial dosimetry. The former task “Irradiation and Isotope Services” has been included in the Radioecology and Tracer Studies programme.

2002 was the first year of the third performance management contract between Risø and the Ministry of Science, Technology and Innovation. Risø’s main areas of research are (with the approximate share given in parenthesis): energy (50%), industrial technology (25%), bioproduction (15%), and radiation research (10%).

The research and development work of the department is carried out in close co-operation with Danish and foreign universities and research institutes and also with the Danish nuclear and radiation protection authorities. The department participates in national and international research programmes including the European Commission research programmes and the Nordic Nuclear Safety Research Programme.

This report presents a summary of the work of the department in 2002 with an emphasis on the results of the research and development activities. Lists of publications, committee memberships and staff members are included.

## 2 Radiation physics

The radiation physics programme carries out research and development in the fields of radiation protection and radiation technology with special emphasis on dosimetry, emergency management and aspects of decommissioning. The aim is to contribute to the protection against the harmful effects of ionising radiation through co-operation with the Danish nuclear and radiation protection authorities and to develop new methods and new applications of nuclear methods in research and industry.

### 2.1 Luminescence dosimetry

#### **Optical fibre dosimeter system**

Initial work carried out at Risø on the development of optical fibre dosimeters for radiotherapy is described in a paper “Development of optical fibre luminescence techniques for real-time in-vivo dosimetry in radiotherapy” which was presented at the IAEA Symposium on standards and codes of practice in medical radiation dosimetry, Vienna, November 2002. This paper reflects the collaborative research between Risø, Malmö University Hospital (Sweden), Oklahoma State University (USA) and Landauer Inc. (USA) covering a work period of approximately one year up to end of October 2002. This work was based on a prototype fibre dosimeter system developed at Risø using a 1x1x2 mm rod of  $\text{Al}_2\text{O}_3:\text{C}$  which was cut from a standard single crystal chip (5 mm diameter x 1

mm thick) initially manufactured by Landauer Inc. The small  $\text{Al}_2\text{O}_3:\text{C}$  rod was coupled to the end of a 15-metre long 1 mm diameter plastic fibre. A 20 mW green (532 nm) laser served as stimulation light source thus providing a real-time combined radio luminescence (RL) / optically stimulated luminescence (OSL) in-vivo dosimeter system.

The same optical fibre dosimeter prototype has recently been used intensively in experiments carried out at the linear accelerator and mammography facilities at the Malmö University Hospital. Also recently we have begun modelling of the clinical beam interactions with the fibre probe in different measurement geometries using Monte Carlo calculations (MCNP code).

### Radiotherapy experiments

Our RL/OSL fibre dosimeter system was tested at a clinical linear accelerator at Malmö University Hospital. We used a typical water phantom calibration configuration and typical beam parameters: the fibre probe was positioned in a cubic tank made of Perspex and filled with water ( $50 \times 50 \times 70 \text{ cm}^3$ ), and irradiated with a 10 cm x 10 cm beam of 6 MV photons. Measurements were acquired with the probe positioned at different depths in water from 16 cm to 20 cm. The OSL results are plotted in Fig. 2.1 and compared with those obtained from similar measurements using conventional diodes, which is a commonly used dosimeter in radiation therapy. The two systems (OSL and diodes) agree within 2%. The results were used to benchmark a Monte Carlo simulation code, which can reliably model the beam produced by a linear accelerator. We anticipate this tool to be extremely useful for the optimisation of the design of future OSL fibre probes.

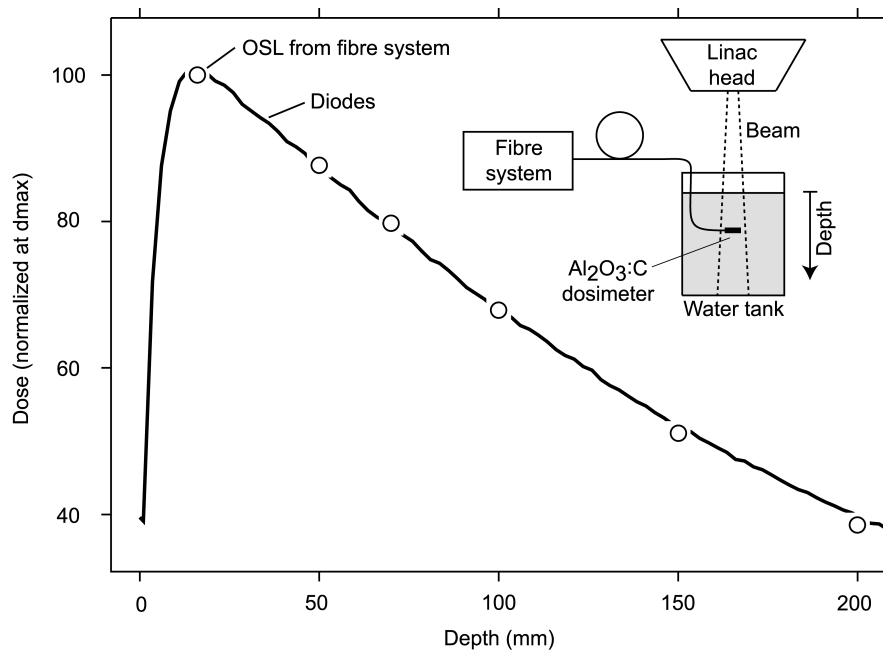


Figure 2.1. Dose from 6 MV photons versus depth into a water phantom as measured using conventional diodes (bold line) and the OSL signal from the Risø fibre dosimeter (open circles).

### Diagnostic radiology/mammography experiments

Diagnostic radiology presents a different challenge for the new fibre probe: the doses delivered are several orders of magnitude smaller than in radiotherapy, the energy of the radiation beam is much lower, and the exposure times much

shorter. We carried out experiments using a mammography unit at Malmö University Hospital. The “prompt luminescence” (RL) feature of our device was used to visualize every individual short pulse of radiation delivered by the mammography unit. Fig. 2.2 shows RL pulses measured with the real-time RL/OSL fibre probe as a result of four repeated short exposures to 25 kV X-rays. These results demonstrate that the sensitivity of the fibre probe is extremely high and that doses of the order of 100 micro Gy can easily be measured. This is of particular importance in the control of doses received by patients under mammography diagnostics, since both entrance and exit doses to the breast can be determined.

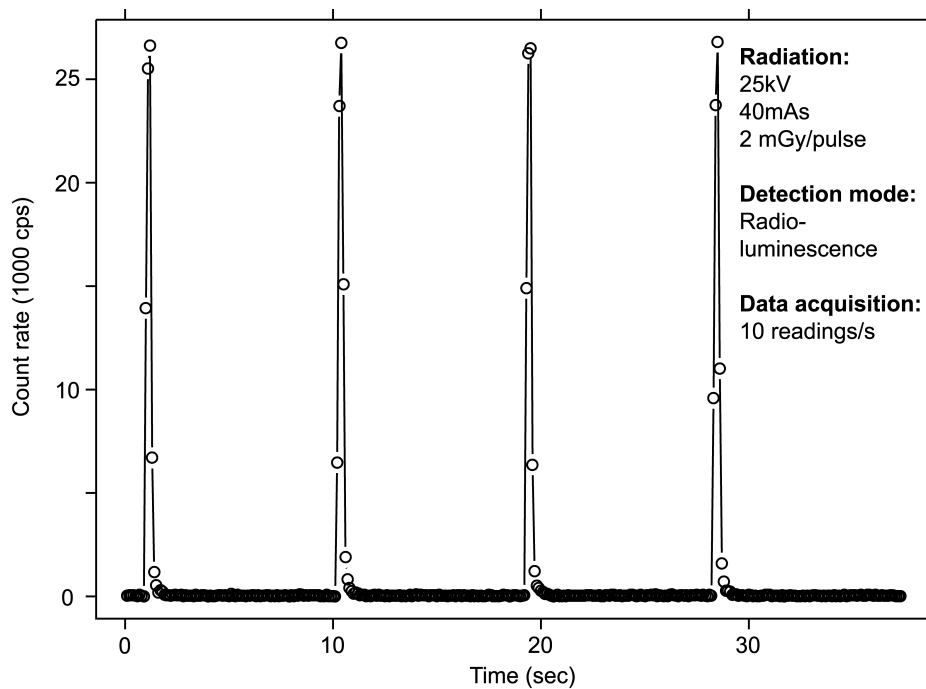


Figure 2.2. Dose RL measurements of 25 kV X-ray pulses generated by a diagnostic mammography unit. Each pulse represents a dose of approximately 2 mGy.

### Development of a new instrument platform

A new optical fibre instrument has been designed that allows a variety of fibre dosimeters to be attached. Also Labview software was developed for the control of the equipment and analysis of data using a dedicated laptop PC. The features of the new Risø Optical Fibre Probe System are illustrated in Fig. 2.3.

### Mini X-ray generators for luminescence research

We have carried out an investigation to test whether a mini X-ray generator is a suitable alternative to a radioactive source in luminescence dosimetry measurements. The study has mainly been motivated by the need for high dose rates (~1 Gy/s) e.g. to facilitate geological dating applications. Furthermore, X-ray generators can more easily be used in fieldwork and for work in countries where regulations restrict the use of radioactive sources.

In 2002, we conducted a study of the characteristics on a small low-voltage X-ray generator (50 kV, 1 mA). For example, we characterized its linearity, stability and dynamic dose-rate range. Because of the differences in radiation type and energy, we also compared single-aliquot regenerative-dose protocol (SAR) growth-curves obtained with a conventional  $^{90}\text{Sr}/^{90}\text{Y}$  beta source and growth

(a)



(b)

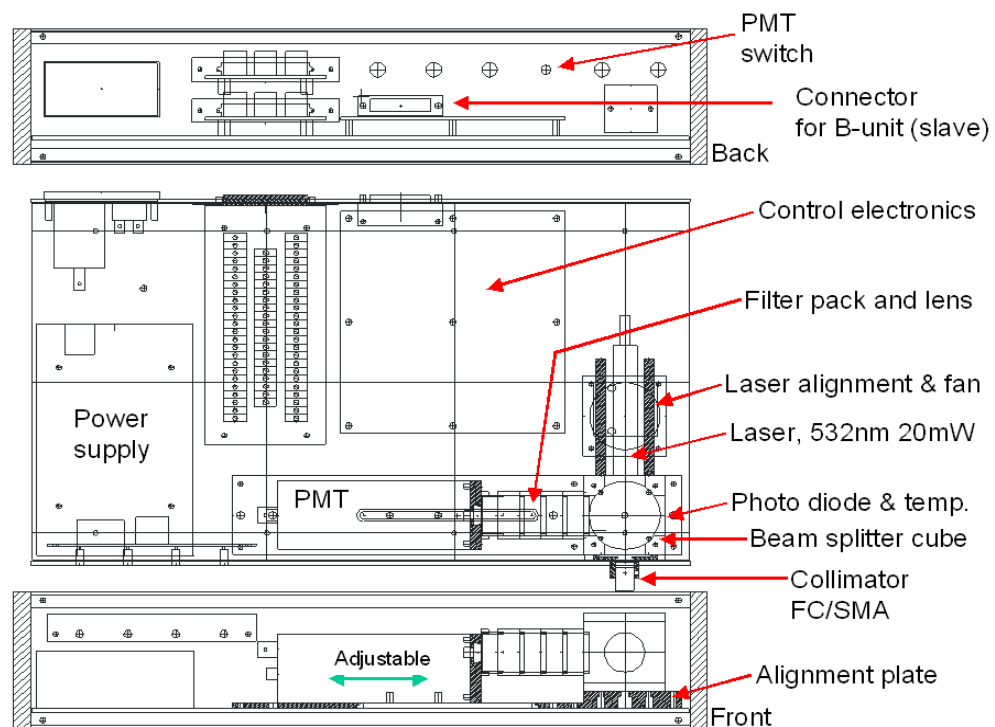


Figure 2.3. The Risø ME-03 OSL/RL fibre system contains electronics and optics for the read out of one fibre dosimeter. The main optical components are a 20mW green laser, a photon-counting module for the luminescence signals, and a photodiode for measurement of the laser-power. All optical components are housed in light-tight enclosures and aligned on a specially designed base plate. The ME-03 unit is controlled from a laptop PC with an E-series data acquisition card from National Instruments. Both soft- and hardware have been designed such that two ME-03 units can be read out simultaneously using only one single data acquisition card.



curves obtained using the X-ray generator (see Figure 2.4). Some tests were made with samples of thermally sensitised sedimentary quartz, which after X-ray irradiation were transferred to an automated Risø luminescence reader; others were made using an on-line dosimeter system based on the optical fibre  $\text{Al}_2\text{O}_3:\text{C}$  dosimeter described in the previous section.

The main conclusions from the study were that the tested system had (i) excellent linearity between tube current (as set by the user on the control computer) and dose rate, (ii) a wide dynamic range: 2–30 mGy/s at 10 kV and 10–2000 mGy/s at 50 kV, (iii) short-term stability better than 0.2%, and (iv) a highly uniform irradiation of the sample area. Tests carried out with a sample of thermally sensitised sedimentary quartz suggest that we can use beta and X-ray irradiations interchangeably to construct growth curves using the single-aliquot regenerative-dose (SAR) measurement protocol.

Based on the above results, it is intended to develop an X-ray irradiation unit that can be attached to the automatic Risø TL/OSL reader.

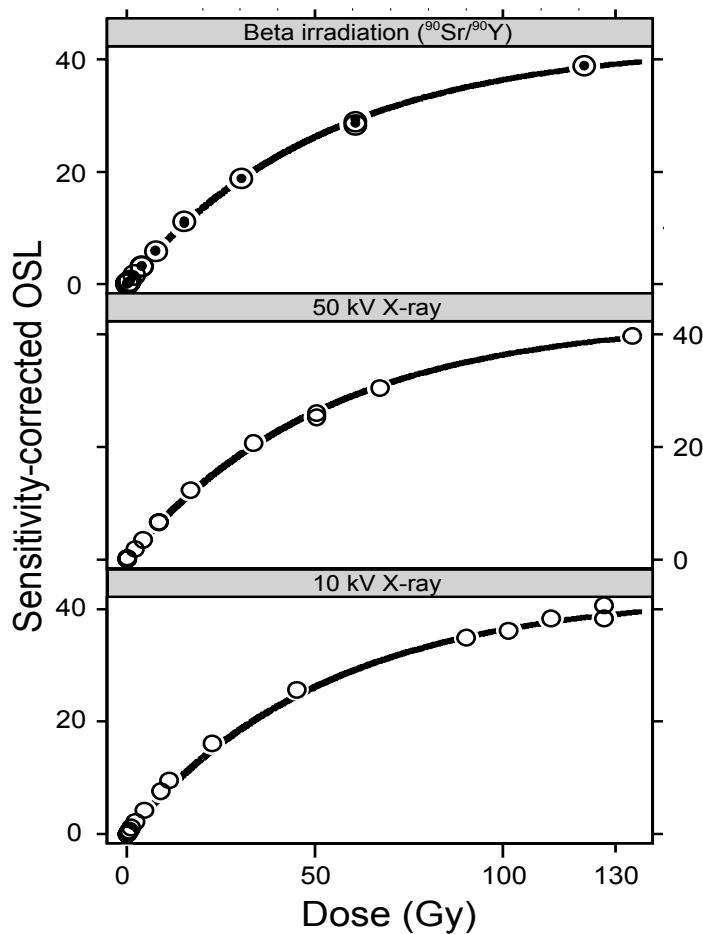


Figure 2.4. SAR growth curves with regeneration doses provided by: (top) beta irradiation, (middle) 50 kV X-rays, and (bottom) 10 kV X-rays. We performed the beta growth curves twice (shown in the top panel with dots and circles), and the two others only once. The solid line (shown in all three panels) is a first-order exponential model fitted to the pooled beta data. The good agreement between the three growth curves shows that we can use beta and X-ray irradiation interchangeably.

## 2.2 Emergency preparedness

### **ECCOMAGS project**

As part of the European ECCOMAGS project (European Calibration and Coordination of Mobile and Airborne Gamma Spectrometry) the RESUME 2002 exercise was held in SW Scotland. At the exercise, large areas were surveyed for anthropogenic and natural radioactivity with Airborne Gamma Spectrometry (AGS), Car-borne Gamma Spectrometry (CGS) and in-situ measurements. The primary aim of the exercise was to evaluate the ability of AGS teams in Europe to produce consistent dose rate and radionuclide deposition data using predefined protocols and to demonstrate mapping capabilities following nuclear fallout. Ten AGS teams, three CGS teams, and seven teams performing in-situ and dose rate measurements took part in the exercise. In addition, soil cores were collected from 39 locations within the exercise area for laboratory measurements of the radionuclide depth profiles.

Risø has participated in the exercise design and evaluation including post-exercise data processing and analysis. Exercise survey tasks included measurements at calibration sites and surveys of common areas to enable direct inter-comparisons between airborne systems and comparison with ground-based measurements. The work also included a composite mapping task where each team was asked to measure part of a very large area. The combination of the results from many teams demonstrated speed of data capture, and the abilities of teams from diverse countries to cooperate effectively in a nuclear emergency. An important objective of the RESUME 2002 exercise has been to validate measurement protocols for the use of airborne gamma spectrometry to estimate ground level environmental gamma dose rates and deposited activity, in order to develop them as European standards for AGS following a nuclear emergency.

### **Data assimilation of atmospheric dispersion**

The results of an atmospheric dispersion experiment carried out in Mol, Belgium, in which  $^{41}\text{Ar}$  and a visible tracer were released from a nuclear reactor stack have been analysed in terms of the Gaussian plume model and the RIMPUFF model developed by Risø. In both cases the models were found to agree well with the measurements of the dispersion of the tracer gas and the gamma radiation field from  $^{41}\text{Ar}$  decay, cf. Fig. 2.5.

A data assimilation model is being developed for on-line source term prediction in the early phase of a nuclear emergency. The model input is radiation data from automatic off-site gamma monitoring stations. The model is developed over the Gaussian plume model and employs an Extended Kalman Filter procedure for filtering radiation measurements. Model parameters such as measurement and process covariance are estimated with a maximum likelihood method. Data from the  $^{41}\text{Ar}$  atmospheric dispersion experiment is used to validate the model. The study is part of an on-going Ph.D. project in data assimilation in atmospheric dispersion of radioactive materials.

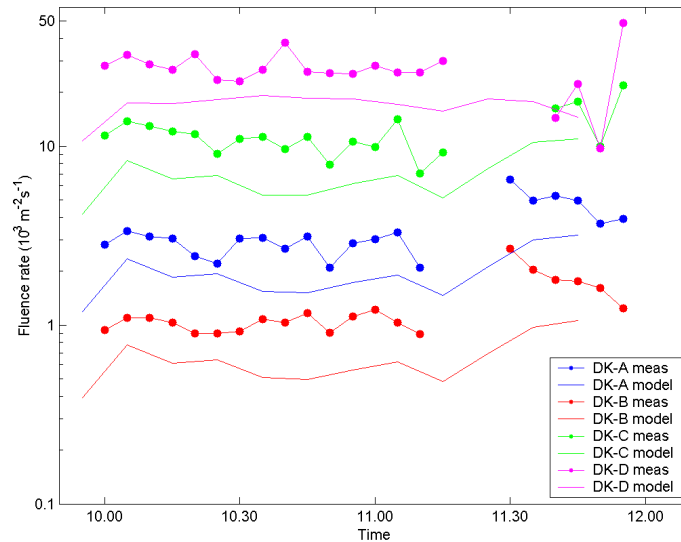


Figure 2.5. Primary photon fluence rates from  $^{41}\text{Ar}$  decay recorded by NaI detectors (dots) and compared to the Gaussian plume model. The four detectors are placed approx. 200 m from the stack and at 0 m to 300 m from the plume centreline.

### The Danish early warning stations

Risø operates the Danish early warning system in parallel with the Danish Emergency Management Agency. In January 2002 the old GK-network was closed down as the last three stations using the GK-network to collect data, Herning, Haderslev and Bornholm, were changed to use normal telephone lines. During February and March 2002 all Danish early warning stations had a major upgrade of hardware and software. The old microcomputer, using an EPROM to collect data from the instruments, store the data and send them to the central computers, was replaced with a standard PC. The spectra are now stored every 10 minutes instead of once every hour and the number of channels in the spectra have been increased from 256 to 512.

Since 1995 Ispra in Italy has hosted the EU Radioactivity Data Exchange Platform (EURDEP), in which 23 countries including Denmark exchange data. In 2002 81 new measuring stations were started while 67 old stations were removed, increasing the number of early warning stations in the European system from 1623 to 1637.

## 2.3 Contamination physics

Research at Risø National Laboratory within the area of contamination physics is focused at improving the understanding of mechanisms governing the consequences of airborne contamination in terrestrial environments. The work includes aspects of, e.g., health physics, aerosol physics, and soil physics, as well as forced decontamination of living areas and agricultural and natural ecosystems.

### Sustainable restoration of contaminated living areas

With the financial support of the Commission of the European Communities, Risø participates in the STRATEGY project to identify and describe potential elements of countermeasure strategies to sustain acceptable living and working conditions in the various types of terrestrial environments, which may be af-

ected by a major nuclear accident. In this context, a need was identified for a comprehensive investigation of each countermeasure that might be considered for implementation in a contaminated living environment. Risø has for many years played a leading role in the field and also led this investigation. It was decided to report the findings of the investigation in a special datasheet format, which would clarify the various factors that would determine the feasibility of applying each countermeasure in a restoration strategy.

The datasheets represent a further development of previously developed databases including new and updated technical data and a greater level of detail. One of the novel features of the STRATEGY database is the inclusion of social, psychological, ethical, legal, and communication aspects, which have previously only been given limited consideration in reports on countermeasure options. A total of 27 countermeasures were found to be of possible relevance to urban contamination situations in European Member States, and these were described. The countermeasures are designed for treatment of different types of contaminated surface in the inhabited environment (streets, pavements, walkways, areas of soil of varying size, vegetation, snow-covered areas, walls, roofs and indoor surfaces of dwellings).

One of the cost elements that will arise after decontamination has been carried out is that associated with the waste generated by the countermeasures. These costs must be regarded as an inherent part of a countermeasure strategy, and descriptions of recommendable waste management options are therefore also reported.

The work has been peer reviewed and also reviewed by groups of potential users and 'stakeholders' (representatives of organisations or individuals that would in some way be involved in parts of the implementation of a countermeasure strategy).



*Figure 2.6. Local workers attempting to decontaminate a wooden wall in the strongly contaminated village of Savichi (Belarus) situated within the 30 km zone around the Chernobyl nuclear power plant. Decontamination of walls is generally only worthwhile in strongly contaminated areas. High-pressure water hosing requires that walls are waterproof. Safety precautions are often neglected if they are inconvenient for the work process.*

## 2.4 Reactor safety

### The Nuclear Knowledge Preparedness Group

The group is composed of members from Risø National Laboratory, the Nuclear Office of the Emergency Management Agency and the Technical University of Denmark. The group has published its annual report on the international status of nuclear power and has held two seminars where current nuclear topics were discussed.

### The DR 1 Project

The DR 1 reactor is being prepared for decommissioning. In that connection its 16 l core solution of 20 % enriched  $\text{UO}_2(\text{SO}_4)\cdot\text{H}_2\text{O}$  has been drained to a safe geometry. Reactivity calculations have been performed to assure a sub-critical storage. These have led to a design where the core solution has been distributed equally in four stainless steel cylinders with an inner diameter of 8 cm and a height of 120 cm. Each steel cylinder is placed in a 12 cm thick Pb-cylinder to reduce radiation from the fission products in the core solution. In Figure 2.7 is shown the k-infinity calculations for the two worst-case geometries. With the four steel cylinders close together without Pb-shield, k-inf is equal to 0.95; the Pb-shield reduces k-inf to 0.78.

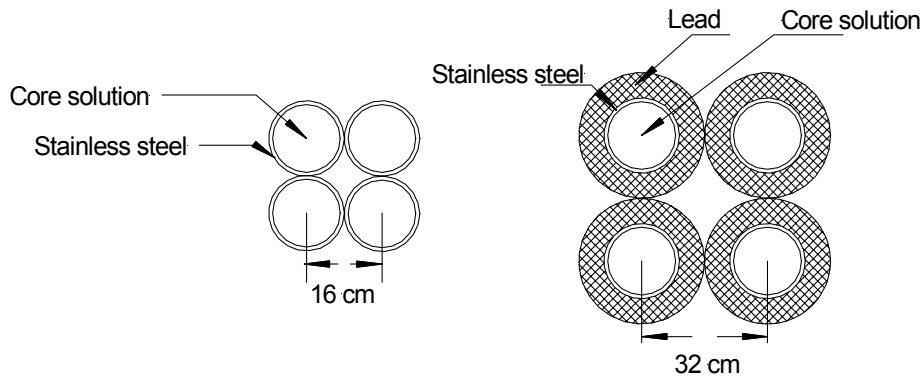


Figure 2.7. Horizontal view of the two worst cases of geometry with respect to criticality. To the left the four stainless steel cylinders without lead shield,  $k_{\text{inf}}=0.95$ , and to the right the four cylinders with 12 cm lead shield,  $k_{\text{inf}}=0.78$ .

### The DR 2 Project

The project, characterizing the status of the reactor, in particular the remaining radioactivity, is almost finished and a report on the results of the investigations performed is under preparation. Measurements of the radioactivity of the cores from cored boreholes through the concrete shield of the reactor have been performed. Monte Carlo flux/activity calculations of the concrete bore samples, based on an evaluated power history, have also been made. The calculations, show fairly good agreement with the measurements, cf. Figure 2.8.

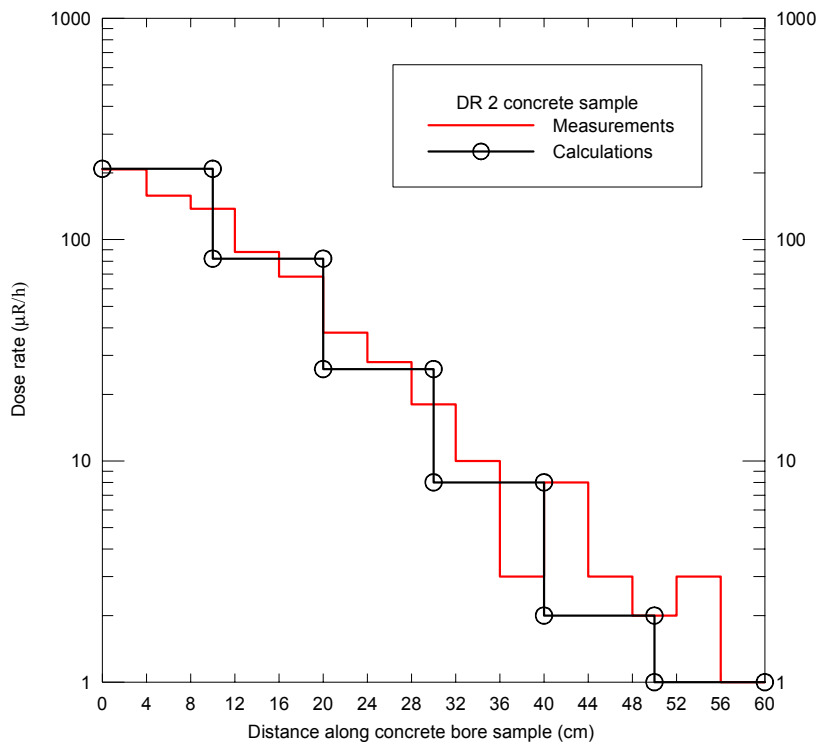


Figure 2.8. Comparison of calculated and measured dose rate in a concrete bore sample of the DR 2 reactor.

#### IAEA Decommissioning Publication

In March a staff member was made available to the IAEA in Vienna where a group of foreign experts on decommissioning of nuclear facilities reviewed, edited and contributed to a publication on “The Transition from Operation to Decommissioning of Nuclear Installations” which the IAEA will publish in its Technical Report Series.

#### Nordic Project on Source Terms for Accidents with Nuclear Submarines

A Nordic project investigating source terms for various nuclear submarine accidents is under preparation. Papers on the problems of submarine decommissioning were published at a Russian-NATO workshop in Moscow and at the American Nuclear Society 2002 winter meeting in Washington.

#### Safeguards Consultant

During the second half of 2002 the Department made a consultant available to the Danish Delegation at the European Union in Brussels in connection with the negotiations under the Danish Chairmanship of the new Commission Regulation on the application of the Euratom safeguards provision.

## 3 Radioecology and tracer studies

The research programme on radioecology and tracer studies focuses on the occurrence and transport of anthropogenic and naturally occurring radionuclides in the environment and the radiological impact on man. The research programme participates in national and international projects concerning studies of radionuclides in terrestrial and marine environments. Furthermore, investiga-

tions on environmental radioactivity are carried out in Denmark, the Faroe Islands and Greenland and the results reported to national authorities and international organisations.

### 3.1 Uranium uptake by plants with and without mycorrhiza

In the EC MYRRH project an experiment was set up to determine the transfer of uranium from two types of soil available to the *Medicago truncatula* plants used. Sixteen plants of *Medicago truncatula* were placed in soil containing various amounts (8 levels from 0.5 to 25%) of rock-phosphate having a uranium concentration of about 4000 Bq kg<sup>-1</sup>. Eight of the plants were inoculated with *Glomus intraradices* mycorrhiza while the remaining eight were not. After 4 weeks the plants were harvested with plant parts and root parts separated and every visible sign of soil contamination removed from the samples.

The samples were dissolved by microwave digestion using a mixture of HNO<sub>3</sub> and HF. The dissolved samples were analysed for uranium (<sup>238</sup>U) by HR-ICP-MS without any radiochemical treatment. The uranium isotope <sup>233</sup>U was used as an internal standard.

The uranium signal in the plant material was well above background and typical procedure blank levels were two to three orders of magnitude lower than what was found in the plant material, yet the levels in the plants were far too low to be analysed by radiometric techniques.

The results for the uranium in plants with and without mycorrhiza are shown in Figure 3.1. Notice the logarithmic concentration scale.

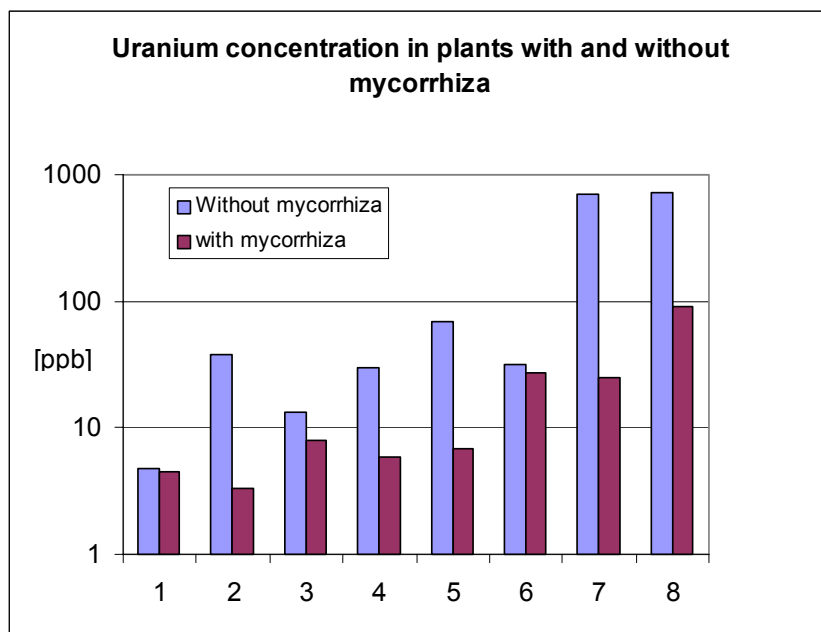


Figure 3.1. Uranium in plant material.

The mycorrhiza does not only promote plant growth significantly but it also seems to help the plant to discriminate against uptake of some substances, like uranium studied in this case. Similar, but not yet quantified, results were obtained for a range of heavy metals in the same study.

### 3.2 Levels and trends of radioactive contaminants in the Greenland and the Faroese environment

Under the international Arctic Monitoring and Assessment Programme (AMAP) levels of radioactive contaminants in various Greenland and Faroese environments have been assessed.

The source of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{239,240}\text{Pu}$  in Greenland terrestrial and fresh water environments is global fallout mainly. In addition, the Chernobyl accident gave a small contribution of  $^{137}\text{Cs}$  in Greenland. Reindeer and lamb hold the largest observed  $^{137}\text{Cs}$  concentrations in the terrestrial environment – up to  $80 \text{ Bq kg}^{-1}$  have been observed. Due to special environmental conditions,  $^{137}\text{Cs}$  is transferred to landlocked arctic char with extremely high efficiency in south Greenland leading to concentrations up to  $100 \text{ Bq kg}^{-1}$ . In these cases very long ecological half-lives are seen.

Concentrations of  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in seawater are decreasing in the order North East Greenland and the coastal East Greenland Current > south west Greenland > central west Greenland and north west Greenland > Irmiger Sea ~ Faroe Islands (cf. Figure 3.2). The general large-scale oceanic circulation combined with European coastal discharges and previous contamination of the Arctic Ocean causes this. The same tendency is seen in marine biota. The peak  $^{99}\text{Tc}$  discharge from Sellafield 1994-1995 has only been slightly visible in the survey year 2001. The concentrations are expected to increase in subsequent years, especially in east Greenland.

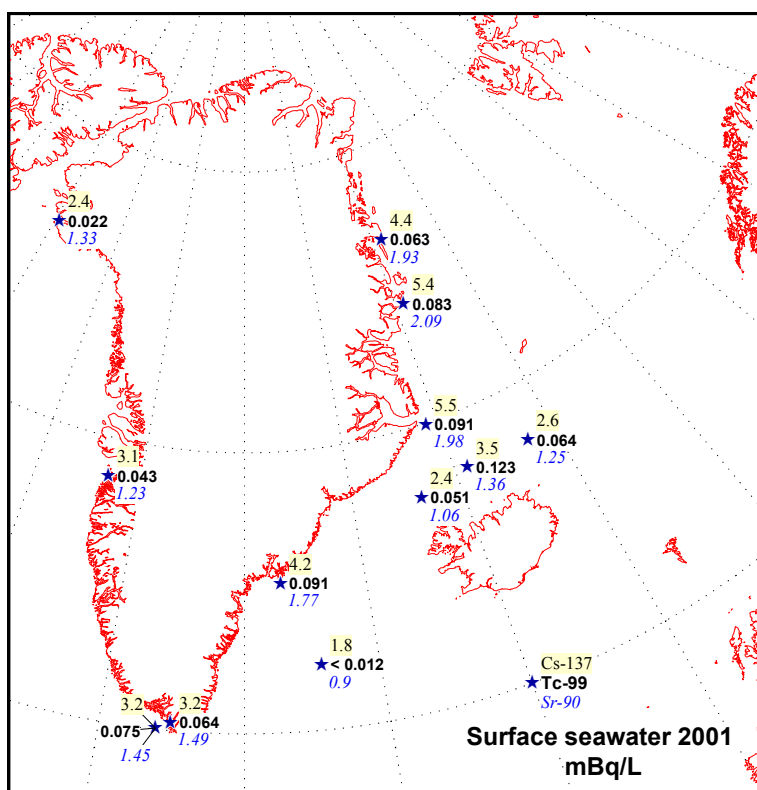


Figure 3.2.  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$  and  $^{90}\text{Sr}$ ,  $\text{mBq L}^{-1}$ , in surface seawater observed in 2001.



### 3.3 Determination of C-14 and H-3 in graphite and concrete from Risø's research reactors

In connection with the decommissioning of Risø's nuclear facilities, the radioactivity of various materials needs to be assessed. Due to large volumes, graphite and concrete comprise considerable amounts of low to medium levels of radioactive waste. The radioactivity of the irradiated reactor graphite and concrete comes from many nuclides such as  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{63}\text{Ni}$ ,  $^{60}\text{Co}$ ,  $^{36}\text{Cl}$ ,  $^{55}\text{Fe}$ ,  $^{41}\text{Ca}$ ,  $^{154}\text{Eu}$ ,  $^{90}\text{Sr}$ ,  $^{133}\text{Ba}$ ,  $^{137}\text{Cs}$  and some transuranics. But, most of the activity is contributed from  $^{14}\text{C}$  and  $^3\text{H}$ . Because  $^{14}\text{C}$  and  $^3\text{H}$  are pure beta emitters, and the energy of their beta particles are low, the samples need to be decomposed to separate  $^{14}\text{C}$  and  $^3\text{H}$  from other interfering nuclides and matrix elements before measurement of their radioactivity by liquid scintillation counting. In graphite,  $^{14}\text{C}$  exists as carbon and  $^3\text{H}$  as HT or HTO. The most common method for decomposition of graphite is combustion, in this case  $^{14}\text{C}$  is released as  $\text{CO}_2$  and absorbed by an alkali solution, while  $^3\text{H}$  is released as HTO and collected by condensing or absorbed in a diluted acid solution. But high temperatures ( $>800\text{ }^\circ\text{C}$ ), a long combustion time (2-3 hours per sample) and special catalysts such as  $\text{V}_2\text{O}_5$  and  $\text{CuO}$  are needed to completely decompose graphite. In concrete,  $^{14}\text{C}$  exists as carbonate or carbon, and  $^3\text{H}$  as HT or HTO in pores and gaps of the concrete. Acid digestion is normally used for decomposing samples in order to release  $^{14}\text{C}$  and  $^3\text{H}$ . However, special apparatus and chemicals (such as HF) are needed to completely decompose concrete and release tritium and  $^{14}\text{C}$ . In addition, this procedure is also time consuming ( $>3$  hours per sample).

In this work, an oxidizing combustion method using a commercial oxidizer was investigated to decompose graphite and concrete for the determination of  $^{14}\text{C}$  and tritium. The graphite and concrete samples mixed with cellulose powder and combustion aids (an organic compound) are put in a platinum basket, and combusted under oxygen flow at temperatures of  $1100\text{-}1200\text{ }^\circ\text{C}$ . The tritium is released as HTO vapour and collected in a glass vial, and  $^{14}\text{C}$  released as  $\text{CO}_2$  is absorbed in Carbon-sorb solution (Figure 3.3). After mixing with scintillation cocktail, the contents of  $^{14}\text{C}$  and tritium are measured by liquid scintillation counting. By this method the time for sample preparation is reduced to 2-3 minutes. The detection limits with this method for  $^{14}\text{C}$  and tritium are 20 and 35 Bq/kg. The cross contamination of  $^{14}\text{C}$  and tritium in the preparation of samples is less than 0.1%. The interference of other radionuclides in samples to the determination of  $^{14}\text{C}$  and tritium in graphite is insignificant. The analytical accuracy investigated by standard addition method is better than 5%. In addition, an acid digestion method was also developed for decomposition of graphite. The results by two decomposition methods showed good agreement. The developed method has been used successfully for the analysis of tritium and  $^{14}\text{C}$  in graphite from the research reactors DR 2 and DR 3.

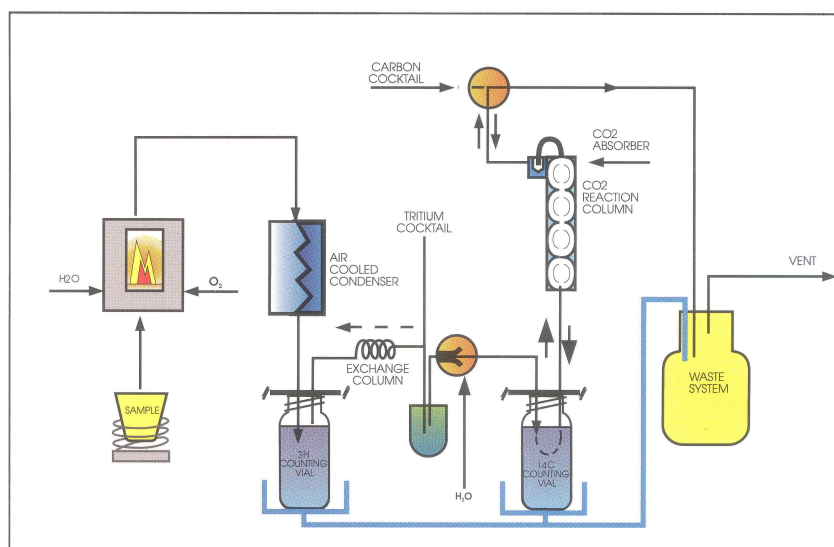


Figure 3.3. Schematic view of oxidiser.

### 3.4 Stable elements analyses in relation to the decommissioning of the reactors at Risø

In the process of the decommissioning of the reactors at Risø it is important to know or make well-educated estimates of the radionuclide inventories in the construction materials.

Development of high-quality analytical procedures of stable trace elements in the various construction materials, such as steels, graphite, concrete, aluminium and lead, is therefore essential. The knowledge of the concentrations of the stable trace elements is used to calculate neutron activation of the construction materials during the operation of the reactors.

We have analytical instrumentation that is suitable for multi-element analyses of major to trace components in gases, liquids and solids. The analytical equipment relies on inductively coupled plasmas (ICP) coupled to detectors based on optical emission spectrometry (OES) and mass spectrometry (MS). The combination of ICP with OES and MS detectors offers rapid multi-element analyses with parts per trillion (or lower) detection limits, good precision and a wide dynamic range.

The preliminary work for the decommissioning process was begun in 2002 and the initial process has involved development of accurate methods for the analyses of Ag, Ba, Co, Eu, Ni and U in aluminium, lead, graphite and concrete. <sup>107</sup>Ag, <sup>132</sup>Ba, <sup>59</sup>Co, <sup>151/153</sup>Eu and <sup>62</sup>Ni may be neutron activated with sufficiently high efficiency to form radioactive isotopes that have relevant half-lives for the decommissioning process (years or longer). Other trace elements of interest for the work of the Risø Decommissioning include Li, Cl, Nb, Sm and Pu.

The method development involves sample digestion and chemical analysis and the accuracy of the methods are verified by the use of certified reference materials in combination with standard addition. Typical values for the concentrations of some of the abovementioned trace elements in relevant samples of aluminium and lead are shown in Table 3.1. The measured concentrations of Ba and Eu in both metals are too low to be of any importance and this is probably also the case for Ag in aluminium and Co in lead.

The sample digestion for lead and aluminium is straightforward and involves acid digestion with half-concentrated HNO<sub>3</sub>. Analysis of graphite involves leaching with HNO<sub>3</sub>/HF/H<sub>2</sub>O<sub>2</sub> mixtures at elevated temperatures and pressure (in a closed vessel heated in a microwave oven) or pyrolysis at 800° followed by dissolution of the residue by HNO<sub>3</sub>. Total dissolution of concrete is more difficult; but rather efficient dissolution as well as leaching of the target elements is achieved with HNO<sub>3</sub>/HF mixtures in combination with prolonged heating using microwave digestion methodology. Alkali fusion with Na<sub>2</sub>CO<sub>3</sub> is necessary for the heavier concretes containing large amounts of BaSO<sub>4</sub>.

*Table 3.1. Typical concentrations (ppm) of trace elements in aluminium and lead.*

	<b>Aluminium</b>	<b>Lead</b>
<b>Ag</b>	0.04	21.5
<b>Ba</b>	0.07	0.8
<b>Co</b>	2.4	0.04
<b>Ni</b>	42.4	3.4
<b>Eu</b>	0.00	0.00
<b>U</b>	1.4	0.25

### **3.5 Baltic-Danish co-operation on radiology and radiation protection 2001-2003**

In 1999 the Danish Ministry of the Interior agreed with ministries in the Baltic States and Poland on co-operation and technical assistance in the field of nuclear safety, radiation protection and nuclear emergency management preparedness and response. In addition, the Danish Emergency Management Agency and Risø National Laboratory agreed in 2000 on conditions for projects under Sector Programmes for Central and East Europe on improving the nuclear safety, radiation protection and nuclear emergency preparedness in the Baltic States, Poland and Russia. On this background and after consultations with laboratories in the Baltic States and Poland, an agreement was made in 2001 between the Danish Emergency Management Agency and Risø National Laboratory on a project during 2001-2003 on Baltic-Danish co-operation on radiation protection with emphasis on environmental monitoring and radioecological studies.

Two training courses were organised at Risø National Laboratory in 2002. A one-week course for a single participant dealt with a rapid analysis of low-level amounts of <sup>228</sup>Ra by beta counting of the daughter <sup>228</sup>Ac. A two-week course for four participants dealt with analysis of transuranic elements in environmental samples.

Four seminars of 3 to 5 days each were held at Risø National Laboratory during 2002. The seminar topics and contents are listed in Table 3.2.

About 70 participants from Poland, Estonia, Latvia and Lithuania attended the seminars. An intercomparison exercise on laboratory analyses of radionuclides in seawater, lake water, soil, dry milk and seaweed has been organised. Samples have been distributed to the laboratories.

Table 3.2. Seminars on radioecology and radiation protection organised in 2002.

Topic	Contents
Monitoring strategies and programmes	<ul style="list-style-type: none"> <li>– International agreements</li> <li>– Environmental monitoring in Denmark, the Faroe Islands and Greenland</li> <li>– Environmental monitoring in the Baltic countries</li> </ul>
Sampling techniques, radiochemical methods, detectors and measurements	<ul style="list-style-type: none"> <li>– Sampling techniques</li> <li>– Radiochemistry incl. demonstration in laboratory</li> <li>– QA aspects</li> <li>– Liquid scintillation counting</li> <li>– Alpha spectrometry</li> <li>– Gamma spectrometry</li> <li>– Radionuclide analysis using mass spectrometry</li> </ul>
Dosimetry	<ul style="list-style-type: none"> <li>– Neutron and radon dosimetry</li> <li>– Beta dosimetry</li> <li>– Mobile measurements of radioactive fallout</li> <li>– Atmospheric dispersion experiment</li> <li>– QA in dosimetry</li> <li>– Optically stimulated luminescence for dosimetry</li> </ul>
Radioactive waste, modelling and dose assessment	<ul style="list-style-type: none"> <li>– ARGOS decision support system</li> <li>– Radioecological modelling and dose assessment</li> <li>– Case study of low-level repository in Sweden</li> <li>– Radioactive waste management at Ringhals NPP</li> <li>– Decommissioning of Risø's nuclear facilities</li> <li>– Environmental Impact Assessment on decommissioning</li> <li>– Radioactive waste management at Risø</li> <li>– Analytical issues of waste-related radionuclides</li> <li>– Decommissioning of research reactor DR2</li> </ul>



Figure 3.5. Participants at the Risø seminar on sampling techniques, radiochemical methods, detectors and measurements, 4-8 November 2002.

## 4 Dosimetry

The work of the accredited Risø High Dose Reference Laboratory continued at approximately the same level as recently, although the focus was shifted from dose map reports to calibrations and issue of calibrated dosimeters. The latter include calorimeters for dose measurement at electron accelerators, alanine dosimeters for use in both gamma- and electron-fields, and dichromate dosimeters for use in gamma-fields. Consulting work in connection with industrial radiation processing also increased.

The international standards for radiation sterilization (ISO 11137 and EN 552) are being revised, and we participate in this work as convener of the CEN working group.

Three training courses on “Validation and Process Control for Electron Beam Sterilization” were organized with a total of 24 participants from 12 countries.

### **Low energy beta dosimetry**

A calorimetric dose standard for low energy radiation (80-100 keV) is being developed in collaboration with National Physical Laboratory, UK, National Research Council, Canada, and two private firms. The calorimeter is designed for dose measurement and calibration of other dosimeters, which –almost regardless of their thickness – will exhibit dose gradients within the dosimeter material. Figure 4.1 shows - as an example - measured and calculated depth dose distributions for totally absorbing Mylar. Model calculations (Monte Carlo as well as thermal) have shown that there are considerable influences from external heat sources on the calorimeter response. A design involving a vacuum chamber is expected to solve these problems.

### **Personnel dosimetry**

The personnel dosimetry laboratory has issued 4000 dosimetry reports for dosimeters used by personnel at Risø. The workload has decreased compared to recent years due to the closure of the Risø DR3 research reactor, but the coming decommissioning of the facility is expected to lead to an increased workload.

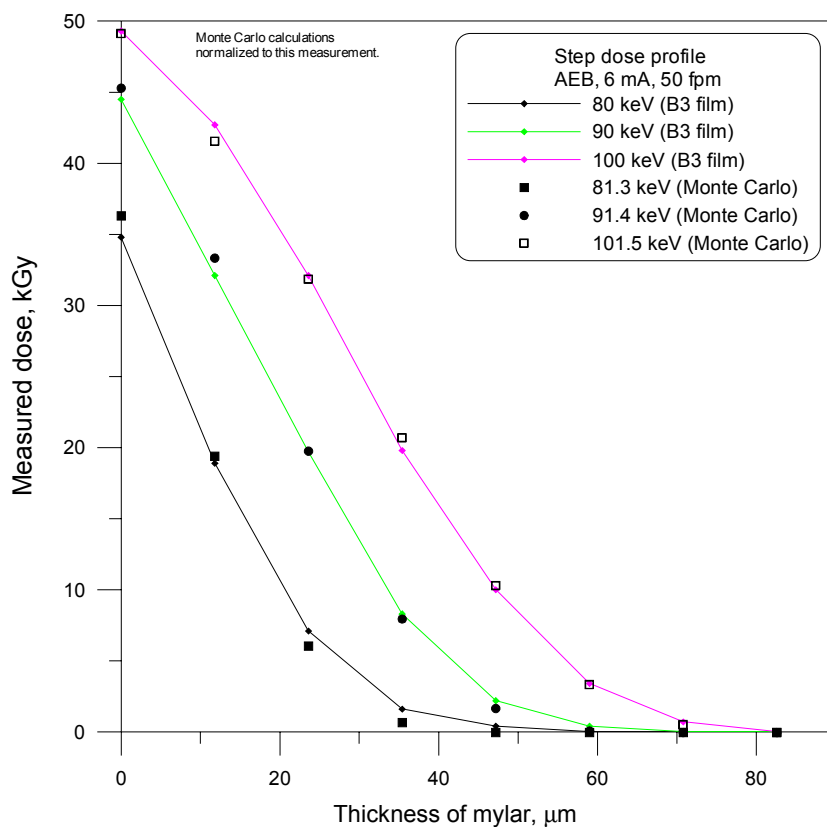


Figure 4.1. Depth dose calculation and measurements in Mylar irradiated with 80, 90, and 100 keV electrons.

## 5 Isotope and irradiation services

The Isotope Laboratory has continued its supply of radioactive materials to customers. Irradiation services at Swedish and Norwegian research reactors have been used to provide radioisotopes to customers and to maintain a production of radiochemicals such as  $^{203}\text{Hg}$ ,  $^{75}\text{Se}$ ,  $^{110\text{m}}\text{Ag}$  and  $^{35}\text{S}$ .

A total of 262 deliveries of  $^{82}\text{Br}$  were supplied to 13 Danish leak detection companies and 15 irradiations were carried out for production of radiochemicals for use at Risø and commercially. Altogether 63 shipments of other radioactive products were sent to domestic and foreign institutes, industry and hospitals. For educational purposes 453 solid radioactive sources were supplied to the Nordic countries. These solid sources are now classified and tested according to ISO 2919. For research applications at Risø 15 deliveries of specially prepared radioisotopes were made.

The first production of  $^{64}\text{Cu}$  radiochemicals for the J.F. Kennedy Institute based on irradiation of  $^{64}\text{Ni}$  in the cyclotron at the University Hospital in Copenhagen was made in the beginning of the year.

## 6 Education

The department is involved in educational activities at Risø, and staff from the department also contributes to education at the universities.

In 2002 one Ph.D. project was finalized in co-operation with Lund University and the degree was awarded at the university. At the end of the year four Ph.D. students were carrying out projects in the department in co-operation with the University of Copenhagen, the Technical University of Denmark, and Lund University.

Staff from the department contributes to a course on isotope techniques at the University of Copenhagen, a course on nuclear instrumentation and health physics at the Technical University of Denmark, and a course on health physics at Risø conducted by the Applied Health Physics section of Risø Decommissioning.

In 2002 three international courses on “Validation and Process Control for Electron Beam Sterilization” were arranged in the department, and one national course was arranged on “Introduction to Radiation Sterilization”.

A staff member from the department has been external examiner in physics at the University of Copenhagen and at the Technical University of Denmark.

A number of internal seminars were arranged in 2002:

- Luminescence dating: Does it work? Andrew Murray
- Retrospective dosimetry: Dose evaluation using unheated quartz Mayank Jain
- OSL developments Lars Bøtter-Jensen
- OSL in retrospective dosimetry Kristina Thomsen
- Dosimetry for Radiation Processing Arne Miller
- Development of imaging software for reading radiation-sensitive film Jakob Helt-Hansen
- Experience from a communication project in the former Soviet Union Jørn Roed
- Early warning system and data exchange Flemming Nielsen
- Restoration of contaminated living areas Kasper Andersson
- Luminescence study of incompletely bleached sediment from a Swedish archaeological site Joanna Baran
- Application of small X-ray tubes in luminescence research Claus Andersen
- Optically stimulated luminescence dosimetry in medical applications Marianne Aznar
- Kalman filtration of radiation data: preliminary studies of the Mol atmospheric dispersion experiment Martin Drews
- OSL “Glow Curves” – A New Paradigm Mayank Jain
- The DR-2 project P.L. Ølgaard
- DR-2 activity calculations Erik Nonbøl
- Luminescence dating of Japanese loess Comparison with an independent Chronology Takuya Watanuki
- Homo sapiens – Anatomical and behavioural origins. Who are we? Zenobia Jacobs
- OSL in retrospective dosimetri using unheated materials Kristina Thomsen

- OSL applications in radiation therapy and diagnostic radiology Marianne Aznar
- On Weapons Plutonium in an Arctic Environment (Thule, Greenland) Mats Eriksson
- MARINA II - Update of the MARINA project on the radiological exposure of the European Community from radioactivity in North European marine waters Sven P. Nielsen
- Plutonium in Skagerrak water and suspended matter Per Roos
- Iodine-129 in Chernobyl Contaminated Soil and Its Chemical Fractionation Xiaolin Hou
- Chemical Analyses of Stable Elements Lars Frøsig Østergaard

## 7 Committee memberships

### 7.1 National

The advisory committee on protection measures in the case of accidents in nuclear facilities (§ 9 stk 2)

B. Majborn and E. Nonbøl

The coordination committee of the Emergency Management Agency and Risø National Laboratory

B. Majborn and A. Damkjær

The coordination committee for nuclear safety in Central and Eastern Europe (Ministry of Foreign Affairs)

B. Majborn

The advisory coordination committee for research in environmental medicine (Ministry of Health)

B. Majborn

The Board of the Danish Nuclear Society

B. Majborn (chairman)

Danish National Council for Oceanology

H. Dahlgaard

Danish Medical Device Industry sterilization committee

A. Miller

Danish Standard, Sterilization committee

A. Miller (chairman )



## **7.2 International**

### **European Union**

Consultative Committee Euratom - Fission  
B. Majborn

Articles 35 and 36 of the European Treaty (Environmental Monitoring)  
S.P. Nielsen

Article 37 Group of Experts  
S.P. Nielsen

National Correspondents on Assistance and Emergency Planning in the Event  
of a Nuclear Accident or Radiological Emergency  
F. Nielsen

Group for Nuclear Safety Research Index, NSRI  
E. Nonbøl

### **OECD/NEA**

Nuclear Science Committee  
Erik Nonbøl

NEA Data Bank Executive Group  
Erik Nonbøl

### **Editorial Advisory Boards**

Radiation Measurements  
L. Bøtter-Jensen

Radiation Physics and Chemistry  
A. Miller (Editor-in-Chief)

Radiation Protection Dosimetry  
L. Bøtter-Jensen

### **Other Committees**

The Board of the Nordic Nuclear Safety Research Programme, NKS  
B. Majborn

Baltic Marine Environment Protection Commission Helsinki Commission  
(HELCOM), Group of Experts on Monitoring of Radioactive Substances in the  
Baltic Sea (MORS)  
S. P. Nielsen

International Solid State Dosimetry Organization  
L. Bøtter-Jensen

Standing Committee for the International Solid State Dosimetry Conferences  
L. Bøtter-Jensen

ISO TC198/WG2, Radiation sterilization working group  
A. Miller

CEN TC204/WG2, Radiation sterilization working group  
A. Miller (convener)

ICRU report committee, Dosimetry for Radiation Processing  
A. Miller

## 8 Publications

### 8.1 Publications in international journals, books and reports

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# 9 Personnel

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Benny Majborn

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Erik Nonbøl

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Povl L. Ølgaard (consultant)

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Mats Eriksson (post doc, from 1 August)

Elis Holm

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Mikael Jensen (from 1 October)

Jesper C. Jørgensen

Miranda Keith-Roach (post doc, until 31 March)

Per Roos  
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**Dosimetry**  
Arne Miller (head of task)

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Jytte Clausen  
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Lis Vinther Kristensen  
Kirsten Madsen  
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Kristina Thomsen

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Richard Bojanowski, Institute of Oceanology, Poland  
Sergej Bormotov, Ignalina Nuclear Power Plant, Lithuania  
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