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Risø-M-2173

ACCELERATOR DEPARTMENT

Annual Progress Report

1 January - 31 December 1978

Abstract. A description is given of work in the fields of irradiation technology, chemical dosimetry, radiation chemistry, physical dosimetry and radiation bacteriology research, as well as of the operation of various irradiation facilities.

INIS-descriptors: ACCELERATOR FACILITIES, BACTERIA, DOSEMETERS, DOSIMETRY, IRRADIATION DEVICES, RADIATION CHEMISTRY, RESEARCH PROGRAMS, RISØE NATIONAL LABORATORY.

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PREFACE

The objective of the Accelerator Department is to contribute to research, development, and the implementation of processes based on ionizing radiation; thus the following activities are carried out:

- Operation and maintenance of the irradiation facilities (three electron accelerators and three ^{60}Co -units).
- Customer irradiation services for laboratories within and outside Risø, for hospitals, and for industry.
- Irradiation technology studies, including the upgrading of present facilities, development of new irradiation equipment, and improvement of equipment and methods for customer irradiation services. Design and construction of equipment for radiation experiments.
- Radiation chemistry research in relation to chemical dosimetry and pulse radiolysis of aqueous solutions connected with fundamental problems in chemistry.
- Radiation physics research in relation to systems used in dose calibration and dose distribution calculations and measurements.
- Radiation bacteriological research mainly in relation to radiation sterilization problems and radiation-resistant microorganisms, and also to increase basic knowledge of the radiation resistance mechanism. Production and supply of bacteriological standard preparations for control of irradiation sterilization plants.

The principal activities in these fields are presented in this report which covers the period from 1 January to 31 December 1978. The contributions marked with \$ are abstracts of published articles. References to these articles are given in section 6.2.

1. OPERATION AND MAINTENANCE OF IRRADIATION FACILITIES^{x)}

(J. Fenger and B. Lynggård)

1.1 HRC electron linear accelerator

Operation of the accelerator was troublefree during the year. During routine shut-down periods minor new installations were carried out. Some vital components in the transmitter were replaced. The weekly schedule for the accelerator is four days for experimental irradiation, half a day for service irradiation and half a day for maintenance.

Microwave transmitter: A new high power microwave klystron was installed (replacement for the one destroyed in 1977). After some installation problems due to technical inaccuracies (solved by ITT), the klystron is now in normal operation.

A new klystron driver unit (spare part delivered in 1977) was repeatedly returned to the factory in USA for warranty repair. The problem was finally solved by the department itself, and the unit is now in normal operation.

The main thyratron failed after 4500 hours in operation (ITT warranty covers 500 hours, average lifetime is in the order of 10,000 hours). The spare thyratron was installed without problems. A new thyratron will be delivered in February 1979.

In connection with the thyratron problems the transmitter high voltage rectifier was destroyed. The type used was no longer in stock, and a complete new rectifier was ordered.

Vacuum system: For a period of 3 months a sufficient vacuum pressure could not be established due to a leak in the system and due to a defect vacuum pump, which was replaced. The pressure in the system is proportional to the pump current and is measured in that way. In case of a defect pump the

^{x)} Technical specifications of the facilities are listed in Appendix 4.

pressure information is misleading. To overcome this problem an independent pressure measuring system will be installed.

Water cooling system: In 1977 a double bed deionizer was installed in the accelerator cooling system in order to reduce the conductivity of the cooling water. Unfortunately the new system is not working satisfactorily, as the copper from the cooling channels is dissolved by means of the aggressive cooling water. The lifetime of the filter substance in the deionizer is heavily shortened by the handling of such an amount of copper. This problem is still under consideration.

Gangway and experimental platform: An experimental platform and a gangway were installed in connection with the radiation field. Design and construction was carried out at the Construction Department.

1.2 Febetron, field emission accelerator

The field emission accelerator was used for Raman spectroscopy experiments and for pulse radiolysis of liquids and gases.

It was shut down for a periodical maintenance, and also for replacement of a defect pulser module.

1.3 ICT, low energy accelerator

Apart from a case of high ozone concentration in the accelerator room operation did not cause troubles. Utilization has been low.

1.4 10,000 Ci ⁶⁰Co-facility

The 10,000 Ci ⁶⁰Co-facility was used for radiation research and for customer services. It further serves as a reference source for microbiological efficiency testing according to the IAEA's recommendations for the radiation sterilization of medical products.

1.5 5,000 Ci ⁶⁰Co-facility

The 5,000 Ci ⁶⁰Co-cell, presently located in the Control Department of "Statens Seruminstitut", Copenhagen, was used for bacteriological research.

1.6 3,000 Ci ⁶⁰Co-facility

The 3,000 Ci ⁶⁰Co-cell was used for research in radiation chemistry, radiation bacteriology and customer services.

2. EXPERIMENTAL EQUIPMENT

2.1 Fast photomultiplier

(J. Fenger)

To take advantage of the accelerator short pulse capability (10 ns pulse width for the pulse radiolysis work), a fast photomultiplier set-up with a rise time of the order of 1 ns has been developed. The photomultiplier will be used for detection of shortlife-reactions mainly in connection with the pressure cell experiments.

2.2 Light intensifier

(J. Fenger)

In order to improve the analyzing light capability, especially in the far ultraviolet region for the pulse radiolysis equipment, a pulse current generator for boosting the light source, a 150 W Xenon lamp, is under development. The design criteria is to increase the light intensity by a factor of 100-200 in a period of up to 10 ms.

2.3 Interface bus

(B. Lynggård)

A bus was built for interfacing the PDP-8 I computer with the Biomation Model 8100 Transient Recorder, to be used in connection with the pulse radiolysis equipment. The recorder has a selective address response capability, and it is planned

to use the bus also for other parts of the instrumentation.

2.4 Fast conductivity equipment

(Johnny W. Hansen)

A fast transient conductivity measuring equipment is being constructed for the pulsed radiolysis experiments. A fast signal amplifier operates in a feed-back loop automatically compensating for the background conductivity, thus performing high sensitivity. To avoid overloading of the preamplifier from the beam electrons a clamping circuit provides protection during the electron pulse time. The cell configuration is arranged in a coaxial geometry matching the transmission line and input impedance of the amplifier and providing a proper shield against electrically disturbances. The covered conductivity range is from 5×10^{-3} to 5×10^{-7} Siemens with a sensitivity better than 10^{-7} Siemens at all ranges, and with a time response of about 10 ns.

In the design of the equipment difficulties exist in keeping the compensating feed-back loop free from oscillations in the rather broad conductivity band.

2.5 Flash-photolysis equipment

(A. Miller)

A rather simple flash-photolysis equipment has been constructed. Its maximum flash energy is 600 J and pulse length is about 10 μ sec.

2.6 Accelerator exit window assembly

(Johnny W. Hansen, P. Lundsager (Engineering Dept.), and B. Lynggård)

A report describing temperature and stress calculations on an output window assembly designed for the linear accelerator has been accepted for publication in Nuclear Instruments and Methods. The paper describes a technique developed in connection with the design of an output window suspended in a collimator flange for use in Rise's linear accelerator. When the

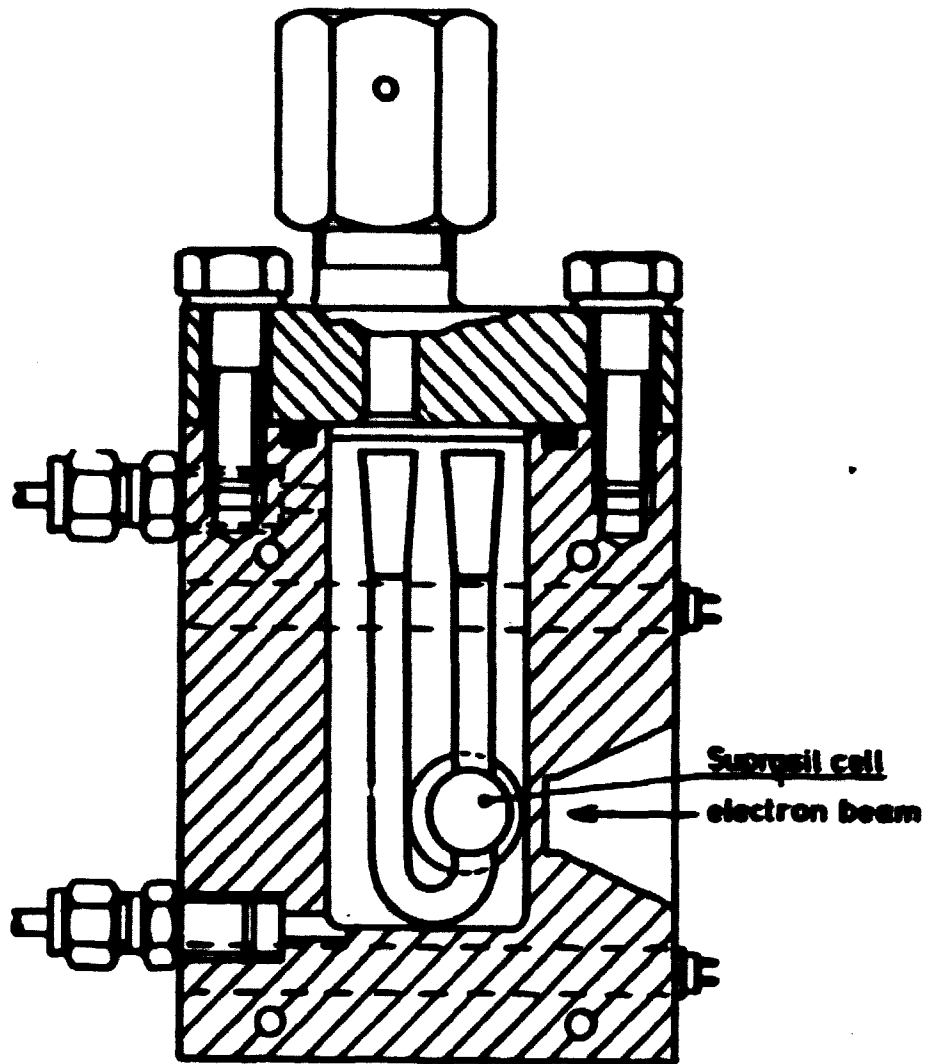


Fig. 1

High temperature pressure cell.

heat developed in the window due to electron absorption has been computed, the temperature distribution is determined by solving the equation for transient heat conduction by means of Hankel transformations. Finally the deflection and limit load of the window membrane are computed by means of simple modifications of small strain plasticity theory in order to cope with large strains. The paper includes an example showing the application of the technique on one aluminum and one titanium membrane.

2.7 High-temperature pressure cell

(E. Engholm Larsen)

A cell for pulse radiolytic measurements up to temperatures of 320°C and pressures of 14 MPa has been constructed (Fig. 1). It consists of a heavy-walled (thickness 2 cm) squareformed steel body with a lock mounted with 8 screws (M8 x 20). The lock is equipped with a top screw, which can easily be removed. Two pipes enter the steel cell, one is used for emptying of liquids, the other for filling with a suitable gas. In the steel body four channels are drilled for fast cooling with water. The body is also equipped with holes for heating elements (4 x 150 W), and with two Chromel-Alumel thermocouples for control and measurement of the temperature using a Deif Transal control.

The electron beam enters the cell through a window (10 x 25 mm) where the wall thickness has been reduced to 2.2 mm. Perpendicular to the electron beam the light passes the cell - through 20 mm thick entrance and exit windows made of synthetic quartz (Tetrafil A). Windows, lock and top are tightened by Vitone O-rings which are replaced when the temperature has been raised to 300°C.

An inner cell made of Suprasil is placed close to the beam window in such a way that it can be filled and emptied with solution from a syringe through the top screw and the "filling" capillary of the cell. The inside length of the Suprasil cell is 25 mm and a choice of cells with 10, 12 or 15 mm

inside diameters may be used. The Suprasil cell is not exposed to high pressure differences.

2.8 References

Johnny W. Hansen and Per Lundsager, A Technique for Temperature and Ultimate Load Calculations of Thin Targets in a Pulsed Electron Beam. Risø-M-2145 (1978). Nuclear Instruments and Methods 160 (1979) 203-210.

3. PHYSICAL DOSIMETRY AND TECHNOLOGICAL APPLICATION OF RADIATION

3.1 Radiochromic dye film dose meters

(A. Miller, Johnny W. Hansen, W. Batsberg Pedersen (Chemistry Dept.), and W.L. McLaughlin (National Bureau of Standards, Washington, D.C., USA))

Thin plastic dose meters containing different radiochromic dyes have been developed and produced in the department for some years(1). The work is performed in collaboration with W.L. McLaughlin. The films are produced with different plastics as base material thereby making it possible to simulate different materials commonly used in radiation processing. Dose measurements in such materials can then be more easily made.

A study of the characteristics of these plastic dose meters (in particular hexakis (hydroxy ethyl) pararosaniline cyanide in polyvinyl-butyril, P-15) is carried out under a research contract with IAEA (2051/RB). Two progress reports were submitted during this year(2,3) and some of these results were also presented at meetings in Vienna(4) and Miami(5). It was found that the sensitivity of the P-15 dose meter decreased during storage and that the optical density increased by 6-10% during the first 24 hours after irradiation. We have also observed this problem in similar commercially available dose meters.

The sensitivity to UV-light was also investigated. The highest sensitivity was found around 340 nm, but for wavelengths longer than 370 nm the sensitivity was drastically reduced. The possibility of producing a UV-dose meter is considered.

3.2 Dose measurements

(A. Miller, W. Batsberg Pedersen (Chemistry Dept.), and W.L. McLaughlin (National Bureau of Standards, Washington, D.C., USA))

In a paper presented at the 2nd International Meeting on Radiation Processing in Miami(6), the dose distribution measurements performed on the 400 keV ICT accelerator were summarized. More detailed information has been given in previous papers(7-9). Radiochromic dye films were used and one of the measurements is shown in Fig. 1. A 1 mm copper wire with 0.4 mm insulation has been irradiated from one side and the dose distribution is shown.

A paper describing measurements of the beam spot of the 400 keV ICT accelerator has been accepted for publication in Radiat. Phys. Chem.(10).

3.3 Microdosimetry

(Johnny W. Hansen, Per Knudsen (Niels Bohr Institute), Robert Katz (University of Nebraska, USA), and W.L. McLaughlin (National Bureau of Standards, Washington, D.C., USA))

A collaboration program has been initiated with professor Robert Katz, Nebraska, USA, to study radiation spectral sensitivities of the thin-film radiochromic dye dose meter (hexahydroxyethyl pararosaniline cyanide in nylon). This 50 μ m thick dose meter is presently used in photon and electron beams, but may also be useful in registering beams of heavy charged particles. Moreover, earlier work has indicated that the dose meter may serve as a "one-hit" detector, i.e. the response appears to show no back reactions for a single hit even by a low specific-ionization particle. The

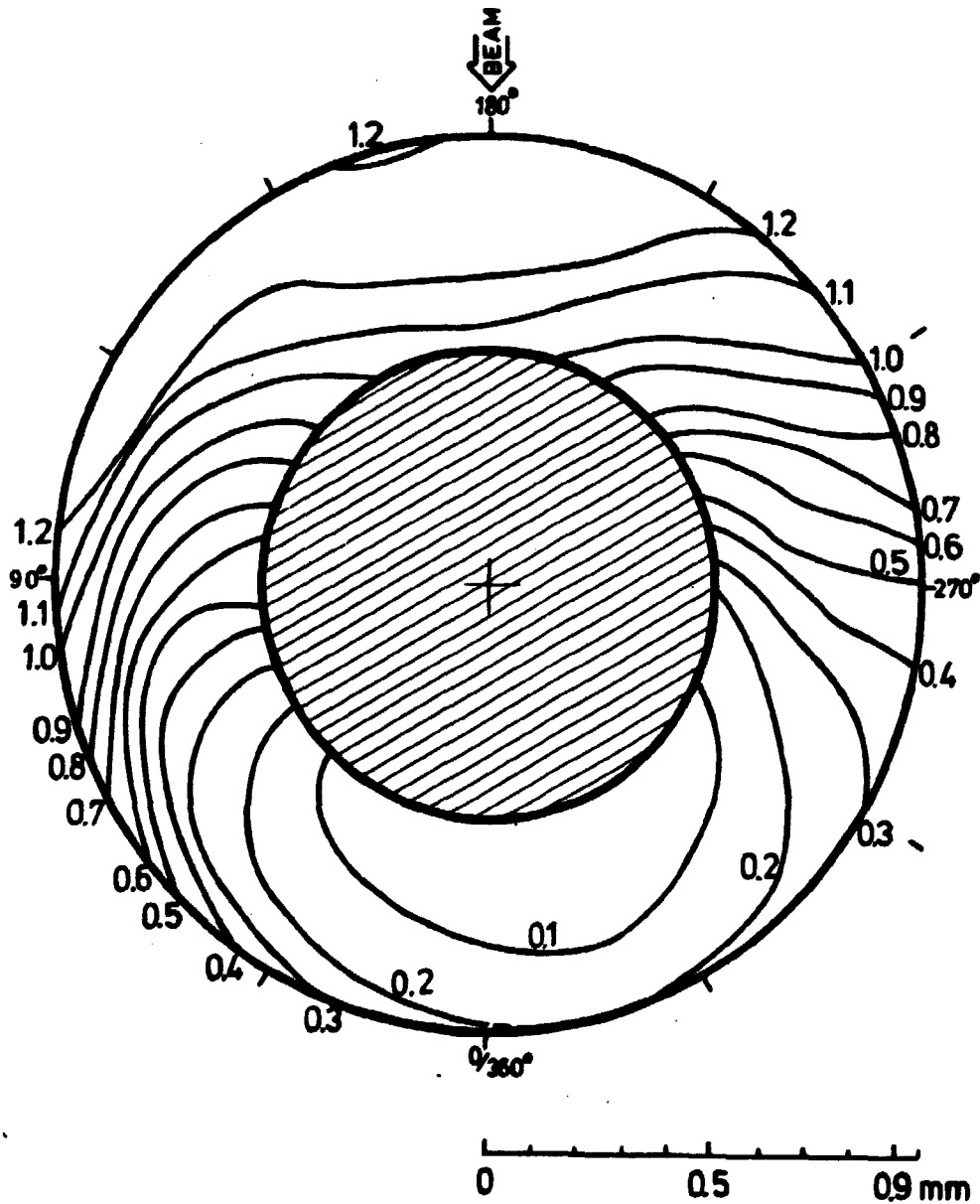


Fig. 2.

Dose distribution in the insulation of an irradiated wire. The wire was irradiated from one side with 400 keV electrons and there is an isodose curve for each 0.1 Mrad.

dose meter should have less variation of response with changes in the linear energy transfer (LET) of the radiation than many other film dose meters. Our purpose of the present study is to determine the response of the dose meter to radiation with an LET significantly greater than previously used, and further to provide useful information about details of radiation distribution along single particle trajectories in matter, including secondary radiations emitted radially from the particle track core. Such information is important in understanding biological and chemical processes initiated by ionizing radiation.

The dose meter film has been thoroughly investigated in electron and photon beams and more detailed experiments have been initiated on the response of the dose meter to higher LET radiation by using the tandem Van de Graaff accelerator of the Niels Bohr Institute, University of Copenhagen.

In the experimental situation with heavy particle irradiation the film has to be irradiated in vacuum which made it necessary to investigate the influence on the optical density versus dose response from the environmental gas atmosphere as well as from vacuum. This investigation was performed with a 10 MeV electron beam(11).

3.4 Dosimetry at low energy

(A. Karadjov (Institute of Roentgenology and Radiobiology, Sofia, Bulgaria) and Johnny W. Hansen)

A report(12) describing a dose meter for low energy, high dose electron irradiation has been accepted for publication in Radiat. Phys. Chem.. The paper describes the determination of a 0.4 MeV electron dose from a bremsstrahlung dose measurement using a converter-detector system. The detector used is the Fricke dosimeter, and the converters are aluminum, copper, and lead foils. Optimal converter thickness is ascertained experimentally for each material, namely: 0.13 g/cm² aluminum, 0.06 g/cm² copper, and 0.045 g/cm² lead. The corresponding peak bremsstrahlung doses at 20 Mrad electron dose

in water are 370, 460, and 940 rad, respectively. A relation is given enabling prediction of optimal converter thickness and peak bremsstrahlung dose for converter materials within a Z-range of 13 to 82. A linear relation is found between bremsstrahlung dose and electron dose ranging from 2 to 20 Mrad. Finally the effect of converter area on detector response has been studied.

3.5 International dose intercomparison

(A. Miller)

IAEA is organizing a study with the aim of establishing an international dose measurement service for commercial irradiation plants. The department has irradiated dose meters and participated in a meeting in Vienna on this subject.

3.6 Commercial activities

(A. Miller, W. Batsberg Pedersen (Chemistry Dept.))

In collaboration with the Chemistry Department an effort is made in order to promote ionizing radiation as an industrial process. Test irradiations have been performed for several firms, and an article was published in the Danish journal "Ingeniøren" (13).

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4. CHEMICAL DOSIMETRY AND RADIATION CHEMISTRY

4.1 Reactions of the radical cations of methylated benzene derivatives in aqueous solution^S

(K. Sehested and J. Holcman)

The radical cations of methylated benzene decompose in acid solution into the corresponding methylbenzyl radical and a proton. The rate constant for this reaction decreases by three orders of magnitude as the number of methyl groups increases from one to five. The rate constants can be correlated with the ionization potential of the parent compound. In neutral solution the reverse reaction to the acid-catalyzed OH adduct conversion occurs and the radical cations react with water to form the OH adduct. In slightly alkaline solution the radical cations of the higher methylated benzenes ($n \geq 3$) react with hydroxide ions forming the OH adduct.

4.2 Radical cations of ethyl-, isopropyl- and tert-butyl- benzene in aqueous solution^S

(K. Sehested and J. Holcman)

Formation of radical cations from the OH adducts of ethyl-, isopropyl- and tert-butylbenzene in strong acid solution is demonstrated. The radical cation forms as an intermediate species in the acid catalyzed water elimination reaction. The proton loss from the radical cation is predominantly from the α -position, but may also occur from β -positions with a rate constant two or three orders of magnitude lower as shown for tert-butylbenzene. The reactivity of the α -hydrogen atoms towards the OH radical increases from

$-\text{CH}_3, >\text{CH}_2$ to $\rightarrow \text{CH}$ as measured from the direct abstraction of hydrogen atoms. This increasing reactivity is also demonstrated in the proton loss reaction.

4.3 Radical cations of p-substituted benzene

(J. Holcman and K. Sehested)

The formation of the radical-cation in N_2O saturated aqueous solution of p-dimethoxybenzene at neutral pH's was demonstrated. This formation is simultaneous with the formation of the OH-adduct and amounts to about 10% of $g(\text{OH})$. In contradiction to other known systems the intermediacy of the OH-adduct in the formation of the radical cation seems to be doubtful, yet the OH radical is a precursor for the radical cation. The analogical formation of semiquinone radical was observed with hydroquinone. Only para substituted benzenes show this type of formation, and the reaction is tentatively ascribed to the quinoid structure of the compounds.

4.4 Pulse radiolysis study of aqueous p-toluonitrile^s

(J. Holcman and K. Sehested)

Hydrated electrons, hydrogen atoms and hydroxyl radicals react with p-toluonitrile with rate constants of 1.3×10^{10} , 7.0×10^9 and $1.2 \times 10^{10} \text{ M}^{-1}\text{s}^{-1}$, respectively. In alkaline solution, O^- radicals react with p-toluonitrile ($7 \times 10^9 \text{ M}^{-1}\text{s}^{-1}$) forming p-nitrilobenzyl radicals. About 6% of OH radicals from p-nitrilobenzyl radicals whereas the rest adds to the aromatic ring. The electron adduct undergoes protonation with $\text{pK} = 8.6$ forming a radical different from the H adduct.

4.5 Pulse radiolysis of aqueous naphthalene solutions^s

(N. Zevos (State University of New York, Potsdam, New York, USA) and K. Sehested)

The reaction between the OH radical generated by ionizing radiation and aqueous solutions of naphthalene have been studied by the technique of pulse radiolysis. The OH radical adds to the naphthalene molecule to form a hydroxyl radical

adduct. The rate constants for the formation and decay of this radical adduct in neutral solutions is $1.20 \pm 0.07 \times 10^{10} \text{ M}^{-1} \text{ s}^{-1}$ and $2k = 1.35 \pm 0.10 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$, respectively. In oxygen saturated acidic solutions the adduct is converted to the naphthalene radical cation by the reaction with H_3O^+ . The rate constants for the formation process and the disappearance of this cation species are 7.0×10^5 and $1.74 \times 10^4 \text{ s}^{-1}$ at pH 0.5. The cation has absorption peaks at 305, 315, and 380 nm and a broad peak between 500 and 700 nm which correspond to the naphthalene cation peaks reported in the literature. The cation is also formed in argon saturated acidic aqueous solution (pH 3) by direct reaction between the naphthalene molecule and the sulfate radical anion. The spectrum of the cation species generated under these conditions is identical with the spectrum in the oxygen saturated solutions. The rate constant for the disappearance of the cation species was found to be $3.46 \times 10^4 \text{ s}^{-1}$ under these conditions.

4.6 A pulse radiolysis study of aqueous cyanamide solutions⁵

(I.G. Draganić, Z.D. Draganić (Boris Kidric Institute, Beograd, Yugoslavia) and K. Sehested)

The radiolysis of oxygen-free, aqueous solutions of cyanamide was studied by fast kinetic spectrophotometry. Computer simulation of the reaction mechanism was used to evaluate the experimental data. Four different species are identified: (1) the radical anion $(\text{NH}_2\text{CN})^-$ absorbing light in the UV with $\lambda_{\text{max}} < 240 \text{ nm}$ and $\epsilon_{240} = 1500 \text{ M}^{-1} \text{ cm}^{-1}$; the disappearance is a second-order process with $2k = 1.3 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$; (2) the hydrogen adduct, $\text{NH}_2\text{C}(\text{H}) = \dot{\text{N}}$ (or $\text{NH}_2\dot{\text{C}} = \text{NH}$), with $\lambda_{\text{max}} 300 \text{ nm}$ and $\epsilon_{300} = 150 \text{ M}^{-1} \text{ cm}^{-1}$ decaying by second-order kinetics with $2k = 3.1 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$; (3) the hydroxyl radical preferentially adds to the cyano group, $\text{NH}_2\text{C}(\text{OH}) = \dot{\text{N}}$ (or $\text{NH}_2\dot{\text{C}} = \text{NOH}$). This species rearranges in the submicrosecond scale to $\text{NH}_2\text{C}(=\text{O})\dot{\text{N}}\text{H}$ ($\lambda_{\text{max}} 325 \text{ nm}$ and $\epsilon_{325} = 1900 \text{ M}^{-1} \text{ cm}^{-1}$) and disappears by a second-order process with $2k = 6.3 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$. (4) It is estimated that about 10% of OH radicals attack the substituent group

and by H abstraction produce the $\dot{\text{N}}\text{HCN}$ radical ($\lambda_{\text{max}} 370 \text{ nm}$ and $\epsilon_{370} = 1800 \text{ M}^{-1} \text{ cm}^{-1}$); it disappears by a pseudo-first-order process attributed to a hydrolysis reaction. At increasing acidities, protonation of this radical takes place, $\dot{\text{N}}\text{HCN} + \text{H}^+ \rightarrow \text{}^+\dot{\text{N}}\text{H}_2\text{CN}$; the protonated form decays faster and absorbs more strongly. In a cyanamide solution containing $\text{S}_2\text{O}_8^{2-}$, the $\text{SO}_4^{\cdot-}$ radicals react with cyanamide, $k = 1 \times 10^8 \text{ M}^{-1} \text{ s}^{-1}$, producing $\text{}^+\dot{\text{N}}\text{H}_2\text{CN}$ radicals. The dependence of the optical density at 325 nm on the dose rate and solute concentration are quantitatively consistent with the assumption that the OH radicals react with the $\text{NH}_2\text{C}(=\text{O})\text{NH}$ species with $k = 4 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$. It is concluded that the cyano group in cyanamide, a N-cyano compound, is the main point of attack by e_{aq}^- , H, and OH as was the case with previously studied nitriles with a C-cyano group and various cyanides.

4.7 The radiation chemistry of aqueous solutions of dicyandiamide⁵

(Z.D. Draganić, I.G. Draganić (Boris Kidrić Institute, Beograd, Yugoslavia) and K. Sehested)

Oxygen-free aqueous solutions of dicyandiamide, DCDA, were exposed to ^{60}Co γ -rays or a pulsed 10 MeV electron beam. Fast kinetic spectrophotometry was used for the study of the free-radical intermediates, and the computer simulation of reaction mechanism for the evaluation of experimental data. The hydrated electron reacts by addition, $k(\text{e}_{\text{aq}}^- + \text{DCDA}) = 1.1 \times 10^{10} \text{ M}^{-1} \text{ s}^{-1}$; the anion-radical $[\text{NH}_2\text{C}(=\text{NH})\text{NHCN}]^{\cdot-}$ absorbs in UV region with $\lambda_{\text{max}} < 255 \text{ nm}$ and $\epsilon_{255} = 1150 \text{ M}^{-1} \text{ cm}^{-1}$. It disappears with $2k = 1 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$ by forming a product which absorbs with $\lambda_{\text{max}} < 255 \text{ nm}$ and $\epsilon_{255} = 2400 \text{ M}^{-1} \text{ cm}^{-1}$. The hydrogen atom also reacts by addition, $k(\text{H} + \text{DCDA}) = 2.7 \times 10^6 \text{ M}^{-1} \text{ s}^{-1}$, and the radical intermediate has $\lambda_{\text{max}} 350 \text{ nm}$ and $\epsilon_{350} = 1250 \text{ M}^{-1} \text{ cm}^{-1}$; it disappears with $2k \geq 10^9 \text{ M}^{-1} \text{ s}^{-1}$. Hydroxyl radicals react both by addition and by abstraction, $k(\text{OH} + \text{DCDA}) = 5.6 \times 10^6 \text{ M}^{-1} \text{ s}^{-1}$, and two transients with $\lambda_{\text{max}} 450 \text{ nm}$ and 355 nm appear. It is estimated that about

90% of OH radicals react by abstraction. It has also been found that the radical-anion $\text{SO}_4^{\cdot-}$ reacts efficiently with DCDA and the product of this reaction, $\text{NH}_2^+\text{C}(=\text{O})\text{NHCN}$, absorbs the light with λ_{max} 355 nm and $\epsilon_{355} = 1150 \text{ M}^{-1}\text{cm}^{-1}$. The OH adduct of DCDA has λ_{max} 450 nm and $\epsilon_{450} = 1700 \text{ M}^{-1}\text{cm}^{-1}$; it disappears by a second order process with $2k \geq 4 \times 10^9 \text{ M}^{-1}\text{s}^{-1}$. The search for stable radiolytic products in the kilorad-megarad dose range has shown the presence of only few smaller molecules.

4.8 Different isomers of OH-adducts

(K. Sehested and J. Holcman)

The spectra of OH-adducts of hydroxy- and methoxy-substituted benzenes has been studied, and the spectral differences attributed to different isomers of the same OH-adducts have been found. It has been found that absorption of the ipso isomers is generally shifted 30-40 nm towards the blue. These differences have been supported by kinetic studies.

4.9 Water elimination from the benzyl alcohol OH-adduct

(J. Holcman)

Preliminary experiments with pulse radiolysis of aqueous benzyl alcohol have been carried out. The O^- radical abstracts methylic hydrogen forming a radical identical with the e_{aq}^- adduct to benzaldehyde. The OH adduct undergoes acid catalyzed water elimination reaction. In contrast to phenolic compounds the water elimination has been observed in alkaline solution.

4.10 The relation between the acid-base behaviour of the OH adducts of substituted benzenes and their ionization potential⁵

(J. Holcman and K. Sehested)

The OH adduct of aromatic compounds is considered as a pseudo-base of the corresponding radical cation and a correlation

between dissociation constants and the ionization potential of the parent compounds is presented. Several well known uncatalyzed water elimination reactions from OH adducts of hydroxy and anilino compounds are included because this reaction is believed to proceed via radical cations.

4.11 Photolysis of periodate and periodic acid in aqueous solution⁵

(U. Kläning (Aarhus University, Denmark) and K. Sehested)

The photochemistry of periodate and periodic acid in aqueous solution was studied by quantum yield measurements at low light intensity by flash photolysis, and by photolysis of glassy samples at 77 K. The photochemical studies were supplemented with pulse radiolysis studies of aqueous periodate solutions and with kinetic studies using stopped-flow technique. In strongly alkaline solution the photodecomposition of periodate proceeds via a formation of O^- and I^{VI} . At pH < 12 an additional primary process is the formation of I^V and O^1D . In neutral solution O^3P is formed in a small yield. The energetics of the subsequent reaction of O^1D with H_2O under formation of H_2O_2 is discussed. Mechanisms for the secondary processes involving I^{VIII} and I^{VI} are given. I^{VIII} and its relatively stable complex with I^{VII} both form I^V , I^{VII} , and O_2 . Depending on pH and concentration, I^{VI} either disproportionates to I^V and I^{VII} , reacts with I^{VII} under formation of I^V and I^{VIII} or dissociates into $O^- (OH)$ and I^V .

4.12 Pulse radiolysis of xenate and perxenate

(U. Kläning (Aarhus University, Denmark), E.H. Appelman (Argonne National Laboratory, USA), and K. Sehested)

The reaction of e^-_{aq} , OH and O^- with xenate and perxenate in alkaline solution is studied. It is found that Xe^{VIII} is oxidized by OH and O^- to Xe^{IX} , which has an absorption in the visible region (~625 nm). It is reduced to Xe^{VII} by the electron and this species absorbs at 310-360 nm. The Xe^{VII}

reacts with or form a complex with Xe^{VIII} , which eventually gives Xe^{IX} . The Xe^{VII} from the oxidation of Xe^{VI} is able to release an $\text{OH}(\text{O}^-)$, which suggests an equilibrium reaction. Some rate constants and extinction coefficients are determined. The radiation behavior of the Xenon compounds are rather similar to the radiolysis of iodate and periodate.

4.13 Pulse radiolysis at high temperatures and high pressures

(H. Christensen (Studsvik Energiteknik AB, Nyköping, Sweden) and K. Sehested)

A cell for pulse radiolytic measurements up to temperatures of 320°C and pressures of 14 MPa is constructed. The activation energy of the reaction $\text{OH} + \text{Cu}^{++}$ is determined to $13.3 \text{ kJ} \times \text{Mol}^{-1}$ ($3.2 \text{ kcal} \times \text{Mol}^{-1}$). A preliminary study of the reaction $e_{\text{aq}}^- + e_{\text{aq}}^-$ yields an activation energy of $22 \text{ kJ} \times \text{Mol}^{-1}$ ($5.3 \text{ kcal} \times \text{Mol}^{-1}$).

4.14 Radical yields in alkaline water

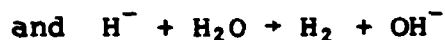
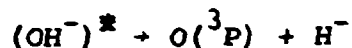
(E. Bjergbakke and E.J. Hart (Argonne National Laboratory, USA))

The work on alkaline water radiolysis was continued. Computer simulation was applied in order to establish the reaction mechanism in the pulse radiolysis experiments; the work is not yet completed.

4.15 $\text{O}(\text{}^3\text{P})$ yields in aqueous solution

(E. Bjergbakke and E.J. Hart (Argonne National Laboratory, USA))

A study on the $\text{O}(\text{}^3\text{P})$ atom was initiated by E.J. Hart. Some experiments performed at Argonne indicates an increase in $\text{O}(\text{}^3\text{P})$ yield at pH 12. The study covers three different systems in order to prove this increase in yield. The mechanism causing the increase in $\text{O}(\text{}^3\text{P})$ yield is assumed to be direct excitation of OH^- :



If H_2O_2 is added to the water, H_2 will be protected and $g(\text{H}_2)$ can be measured. We found an increase in the H_2 yield from $g(\text{H}_2) \approx 0.40$ at pH 10 to $g(\text{H}_2) \approx 0.42$ at pH 13. The second possible system is ClO^- , ClO_2^- where $\text{O}(^3\text{P})$ forms oxygen via $\text{O}(^3\text{P}) + \text{ClO}^- \rightarrow \text{Cl}^- + \text{O}_2$, and the oxygen is protected in this system against further attack by radicals. The fact that also the molecular H_2O_2 forms O_2 makes the measurements rather difficult. An increase in $G(\text{O}_2)$ is expected from 0.70 to 0.72. The results in this system is not yet conclusive, apparently there is an unknown chain reaction yielding much more oxygen than expected. The third system: formate, N_2O and O_2 will protect the H_2 , but for unknown reasons an increase in H_2 yield with pH was not found in this system.

4.16 References

- K. Sehested and J. Holcman, Radical cations of ethyl-, isopropyl- and tert-butylbenzene in aqueous solution. Submitted to Nukleonika, Warsaw, Poland.
- J. Holcman and K. Sehested, Pulse radiolysis study of aqueous p-toluonitrile. Submitted to Nukleonika, Warsaw, Poland.
- J. Holcman and K. Sehested, The relation between the acid-base behaviour of the OH adducts of substituted benzenes and their ionization potential. Submitted to Nukleonika, Warsaw, Poland.
- Z.D. Draganić, I.G. Draganić, and K. Sehested, The radiation chemistry of aqueous solutions of dicyandiamide. Submitted to J. Phys. Chem.
- H. Christensen and K. Sehested, Pulse radiolysis at high temperatures and high pressures. Submitted to Radiat. Phys. Chem.

5. RADIATION BACTERIOLOGY RESEARCH

Bacteriological research concerns the development and testing of radiation sterilization processes, as well as advice and assistance on specific projects to prospective users of radiation sterilization. Research interests are concentrated on the mechanisms of radiation resistance.

5.1 DNA damage induced proteins in *M. radiodurans*

(M. Trier Hansen)

Mechanisms responsible for the repair of DNA damages in the radiation resistant strain *M. radiodurans* were investigated. Total protein synthesis decreased in response to agents causing DNA damage. In contrast, for a few specific proteins a considerably increased rate of synthesis was discovered. These damage induced proteins are presumed to be involved in DNA-repair functions. UV-treatment of the wild type strain induced 3 major proteins which were characterized by isoelectric focusing and SDS-acrylamide gel electrophoresis (Fig. 3).

5.2 Repair of DNA crosslinks in *E. coli*

(M. Trier Hansen and A. Zaritsky (Ben-Gurion University of the Negev, Beer-Sheva, Israel))

The importance of growth rate and chromosome configurations for the repair of DNA interstrand crosslinks in *E. coli* was studied. Psoralen derivatives were used to photochemically introduce crosslinks in vivo. Fast growing cells with greater possibilities for genetic recombination were found to be more resistant to this type of damage than slow growing cells.

A mutant of *E. coli* showing a greatly increased sensitivity to psoralen treatment was isolated. Genetic and physiological experiments were started to establish the map position of the mutation and to determine the mutationally altered function.

5.3 W-reeactivation in *Acinetobacter calcoaceticus*

(D. Berenstein)

Studies on W-reeactivation in *Acinetobacter calcoaceticus* were

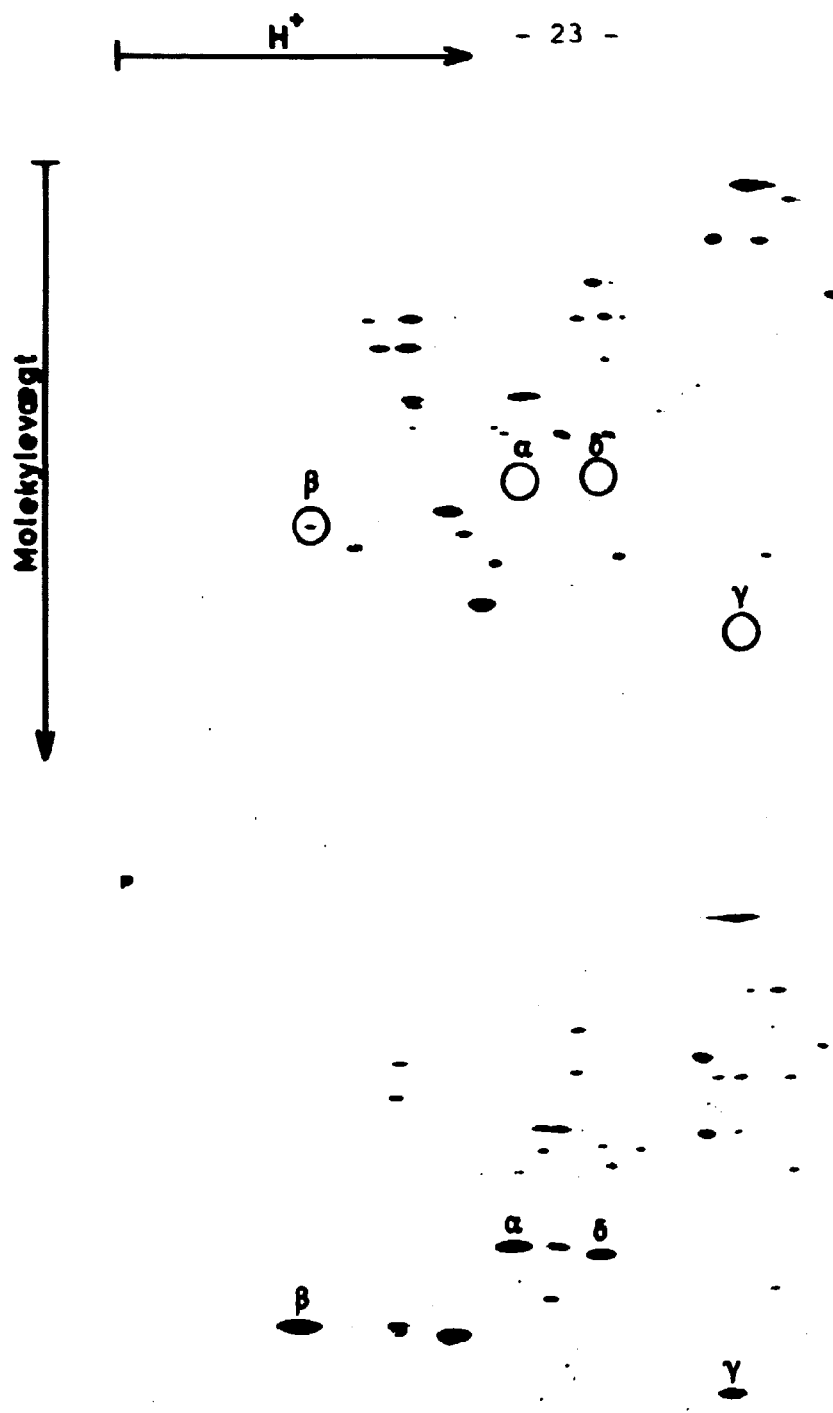


Fig. 3.

Specific proteins synthesized in response to radiation

The proteins of M. radiodurans labelled with ^{35}S -methionine were separated according to charge by isoelectric focussing in the first dimension and according to molecular weight by SDS-PAGE in the second dimension. The lower autoradiogram of extracts from cells exposed to UV irradiation demonstrates that these cells contain four major proteins (α , β , γ , δ) not present in unirradiated control cells (upper autoradiogram).

continued. W-(Weigle) reactivation is defined as the increased survival of an irradiated bacterial virus (phage) upon infection of irradiated bacteria as compared to infection of unirradiated bacteria. The surviving fraction of an Acinetobacter phage is about 10 times greater when assayed by infection of irradiated rather than unirradiated bacteria. The results are compatible with the hypothesis that irradiation of bacteria induces repair enzymes, that are responsible for W-reactivations. Proteins synthesized by unirradiated and irradiated Acinetobacter strains were compared by isoelectric focusing and SDS-acrylamide gel electrophoresis. Preliminary results revealed that probably one protein is induced by UV-irradiation of the cells.

5.4 Production and supply of microbiological standard preparations and biological indicators

(D. Berenstein and M. Trier Hansen)

The laboratory produced, supplied and assayed standard preparations of the spore former Bacillus cereus, strain C 1/1, as well as of the vegetative Acinetobacter calcoaceticus, strain OA4. Tests of the microbiological efficiency of Danish radiation facilities were performed.

5.5 Customer service for hospitals, research laboratories and industry

(D. Berenstein and M. Trier Hansen)

The following services were maintained:

- General consultation, irradiation of test specimens, evaluation of materials and packagings in relation to the introduction of new hospital equipment.
- irradiation of pharmaceutical materials and fodders in order to reduce the initial number of bacteria.

6. EDUCATIONAL ACTIVITIES AND PUBLICATIONS

6.1 Lectures and poster session

Dvora Berenstein, DNA-Inducible Repair. 6th Conference of the Nordic Society for Radiation Research and Radiation Technology. Hirtshals, August 28-31.

E.A. Christensen and K. Sehested, Strålekonservering (Radiation Preservation). DIEU, Copenhagen, May 8.

E.J. Hart, Effect of pH and Bromate Ion Concentration on O(³P) Atom Formations in Gamma-Ray Irradiation Aqueous Solution. Risø, March 8.

Mogens Trier Hansen, Multiplicity of Genome Equivalents and Resistance. Ben Gurion University of the Negev, Beer-Sheva, Israel, February 28.

Mogens Trier Hansen, Regulation of Ribosome Synthesis During Shift-Down in E. coli. Ben Gurion University of the Negev, Beer-Sheva, Israel, March 8.

Jerzy Holcman, The Relation Between the Acid-Base Behaviour of the OH Adducts of Substituted Benzenes and Their Ionization Potential. 6th Conference of the Nordic Society for Radiation Research and Radiation Technology. Hirtshals, August 28-31.

Arne Miller, Industriel udnyttelse af stråling (Industrial application of radiation). Elektroteknisk Forening, Århus, February 16.

Arne Miller, Dose Distribution in Irradiated Wire Insulation. 6th Conference of the Nordic Society for Radiation Research and Radiation Technology. Hirtshals, August 28-31.

Arne Miller and W.L. McLaughlin, Absorbed Dose Distributions in Irradiated Plastic Tubing and Wire Insulation. 2nd International Meeting on Radiation Processing, Miami, October 22-26.

Arne Miller and W.L. McLaughlin, Evaluation of radiochromic dye films and other plastic dose meters under radiation process conditions. Presented at: Advisory group meeting on

Standardization and High-dose Intercomparison for Industrial Radiation Processing, IAEA, Vienna, September 25-29.

Arne Miller, Absorbed Dose Distributions in Electron-Beam Irradiated Plastic Tubing and Wire Insulation. Nuclear and Radiological Physics Seminar. National Bureau of Standards, Washington, D.C., USA, November 6.

A. Zaritsky (Ben Gurion University of the Negev, Israel), Chromosome Replication in Bacteria. Risø, October 4.

Poster Session

Dvora Berenstein, Reactivation of Radiation Damaged Bacteriophage in *Acinetobacter Calcoaceticus*. 6th Conference of the Nordic Society for Radiation Research and Radiation Technology. Hirtshals, August 28-31.

6.2 Publications

Accelerator Department Annual Progress Report (1978), 1 January-31 December 1977. Risø-M-1981.

I.G. Draganić, Z.D. Draganić, and K. Sehested (1978), A Pulse Radiolysis Study of Aqueous Cyanamide Solutions. *J. Phys. Chem.* 82 (7), 757-761.

J.W. Hansen and Per Lundsager (1978), A Technique for Temperature and Ultimate Load Calculations of Thin Targets in a Pulsed Electron Beam. Risø-M-2145.

Mogens T. Hansen (1978), Multiplicity of Genome Equivalents in the Radiation Resistant Bacterium *Micrococcus radiodurans*. *J. Bacteriol.* 134, 71-75.

Ulrik Kläning and Knud Sehested (1978), Photolysis of Periodate and Periodic Acid in Aqueous Solution. *J. Chem. Soc., Faraday Trans. I*, 74, 2818-2838.

W.L. McLaughlin, A. Miller, K. Pejtersen and W. Batsberg Pedersen (1978), Distribution of Energy Deposited in Plastic Tubing and Copper Wire Insulation by Electron Beam Irradiation. *Radiat. Phys. Chem.* 11, 39-52.

Arne Miller (1978), Investigation of the radiochromic dye film dosimeter under process conditions, including stability, precision, accuracy, the influence of dose rate, and the influence of the environment. Progress Report (1 August 1977-31 April 1978), IAEA Research Contract 2o51/RB.

Arne Miller (1978), Investigation of the radiochromic dye film dosimeter under process conditions, including stability, precision, accuracy, the influence of dose rate, and the influence of the environment. Progress Report (1 May - 31 October), IAEA Research Contract 2o51/RB.

W. Batsberg Pedersen and A. Miller (1978), Plastmaterialers egenskaber ændres ved stråling. Ingeniøren 4, nr. 47 tematillæg, 32-33.

K. Sehested and J. Holcman (1978), Reactions of the Radical Cations of Methylated Benzenes Derivatives in Aqueous Solution. J. Phys. Chem. 82 (6), 651.

A. Zaritsky, E. Ben-Hur and M.T. Hansen (1978), The Effect of Growth Rate on the Sensitivity of E. Coli K12 to DNA Crosslinks Induced by Psoralen plus Near Ultraviolet Light. Trans. Nucl. Soc. Israel 6, 17-2o.

N. Zevos and K. Sehested (1978), Pulse Radiolysis of Aqueous Naphthalene Solutions. J. Phys. Chem. 82 (2), 138-41.

W.H. Eriksen and C. Emborg (1978), Increase of Radiation Resistance of a Soil Microflora Exposed to Long Term Gamma Irradiation. Applied and Environmental Microbiology 36 (4), 618-619.

6.3 Test-irradiations

Test-irradiations were carried out for:

Biocoating Aps, Charlottenlund
Danmarks farmaceutiske Højskole, København
Den kgl. Veterinær- og Landbohøjskole, København
Fibiger-Laboratoriet, København
Finseninstitutet, København
Københavns Amts Sygehus, Glostrup

Københavns Universitet: Hygiejnisk Institut & Proteinlaboratoriet

Laboratoriet for teknisk Hygiejne, Dth, Lyngby

Mölnlycke Steritex A/S, Espergårde

Nordiske Kabel- og Trædfabriker, København

Novo Industri A/S, Bagsværd

A/S Nunc, Kamstrup, Roskilde

Planteavlslæder V. Schelbeck, Agedrup

Radiflex Aps, Hedehusene

6.4 Visiting scientists

D.A. Youngs, Travenol Laboratories, Inc., Morton Grove, Ill., USA.

O.H. Larsen, De forenede Papirfabrikker, København.

B. Vestergård, De forenede Papirfabrikker, København.

E. Lengfelder, Strahlenbiologische Institut der Universität München, Germany.

A. Zaritsky, Dept. of Biology, Ben-Gurion University of the Negev, Beer-Sheva, Israel.

D. Ražem, Ruder Bosković Institute, Zagreb, Yugoslavia.

R. Bøg-Hansen, Proteinlaboratoriet, KU, København.

J. Maffei, Proteinlaboratoriet, KU, København.

I. Lorenc-Kerlis, Institute of Biochemistry, University of Wrocław, Poland.

M. Soszynski, Institute of Biochemistry & Biophysics, Dept. of Biophysics, Łódź, Poland.

Z. Zunić, Institute of Nuclear Sciences, Boris Kidrić Institute, Vinča, Yugoslavia.

N. Hilmy, Pasar Jumat Research Center, National Atomic Energy Agency, Jakarta, Indonesia.

A delegation from the People's Republic of China visited the department on a study mission on biological products (sterilization procedures).

E.J. Hart, Argonne National Laboratory, Argonne, Ill., USA.
H. Christensen, Studsvik Energiteknik AB, Studsvik, Sweden.
W.L. McLaughlin, National Bureau of Standards, Washington,
D.C., USA.

7. IRRADIATION FACILITIES AT THE ACCELERATOR DEPARTMENT

Electron Accelerators

1. Linear Electron Accelerator, Haimson Research Corporation, Model HRC-712

Specifications:

Electron energy 10 MeV
Average electron current 1 mA
Peak electron current at 10 MeV 1100 mA
Pulse length, normal mode 1 - 4 μ s
Pulse length, short pulse mode 10 - 1000 ns
Pulse repetition rates single pulses and
12.5, 25, 37.5, 50, 100, 150 and 200 pps
Energy spread 78% of the beam
current within a spread of \pm 2.5%

Pulse-to-pulse dose variation:

- a) within a pulse train, less
than 1.8%
- b) for single pulses separated at
10 min. intervals, less than ... 3%

Electron pulse flatness over a 2 μ s
interval, better than \pm 1%

Accelerator room beam facilities:

- 1. A bent beam with scan width of 40 cm
providing a process irradiation ca-
pacity of 1000 - 1500 Mrad kg/hour.
- 2. A horizontal beam, full average beam peak
power, for electron and X-ray ir-
radiation.

3. A horizontal beam, reduced average beam power (12.5 pps) in connection with a $\pm 0.5\%$ beam slit.

Target room beam facilities:

1. Three horizontal beam ports, reduced average beam power (12.5 pps).

2. Field Emission Electron Accelerator, Febetron Model 705B

Specifications:

Electron energy 1.5 - 2.0 MeV
Peak electron current 4000 A
Pulse length (electron mode) 20 ns

3. Low-Energy Electron Accelerator, High Voltage Eng. Corp. Model EPS 400-IND

Specifications:

Electron energy 400 keV
Electron current 50 mA
Scan width 120 cm

The accelerator is provided with conveyors to permit pilot-plant irradiation.

⁶⁰Co-Facilities

10,000 Ci ⁶⁰Co-facility (built at Risø 1957)

Designed for very homogeneous irradiation of samples with a maximum length of 1,000 mm and diameters of maximum 180, 100, or 60 mm. The corresponding maximum dose rates (5,300 Ci, 1 January 1979) are 2.9×10^5 rads/h, 7.7×10^5 rads/h, and 1.9×10^6 rads/h, respectively.

5,000 Ci ⁶⁰Co-facility (built at Risø 1971)

Designed for laboratory use and fitted with a 123 mm^Ø x 150 mm irradiation chamber. The dose rate in the centre

of the chamber (3,600 Ci, 1 January 1979) is 3.1×10^5 rads/h. The cell is located at the Control Department, Statens Serum Institut, Copenhagen.

3,000 Ci ^{60}Co -cell (built at Risø 1968)

Designed for laboratory use and fitted with a 120 mm ϕ x 200 mm irradiation chamber. The dose rate in the centre of the chamber (1,700 Ci, 1 January 1979) is 1.7×10^5 rads/h.

8. STAFF OF THE ACCELERATOR DEPARTMENT

Head: Knud Sehested

Office staff: Ebba Haugaard and Ruth Madsen

Scientific staff

D. Berenstein
E. Bjergbakke
J. Fenger
M. Trier Hansen
J.W. Hansen
J. Holcman
B. Lynggård
A. Miller
K. Sehested

Technical staff

S. Bøjlund Andersen
K. Boysen
H. Corfitzen
I. Hansen
I. Høegh
T. Johansen
E. Engholm Larsen
F. Larsen
I.M. Larsen

L. Nielsen
W. Nielsen (until 31 October)
P. Broen Pedersen
K. Pejtersen
B. Thomsen (until 31 August)

Consultants

Dr. E.A. Christensen, Chief Physician, Control Department,
Statens Seruminstitut, Copenhagen.

Dr. E.J. Hart, Port Angeles, WA., USA.

Dr. W.L. McLaughlin, X-Ray Physics Section, Center for
Radiation Research, National Bureau of Standards, Washington,
D.C., USA.

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<p>Title and author(s)</p> <p>Accelerator Department - Annual Progress Report for the period 1 January - 31 December 1978.</p> <p>Acceleratorafdelingens årsrapport 1 januar - 31 december 1978.</p>	<p>Date May 1979</p> <p>Department or group</p> <p>Accelerator</p> <p>Group's own registration number(s)</p>
<p>32 pages + tables + 3 illustrations</p>	
<p>Abstract</p> <p>A description is given of work in the fields of irradiation technology, chemical dosimetry, radiation chemistry, physical dosimetry and radiation bacteriology research, as well as of the operation of various irradiation facilities.</p> <p>Available on request from Riss Library, Riss National Laboratory (Riss Bibliotek), Forsøgsanlæg Riss), DK-4000 Roskilde, Denmark Telephone: (03) 37 12 12, ext. 2262. Telex: 43116</p>	<p>Copies to</p>