Accelerator Department. Annual progress report, 1 Januar - 31 December 1979

Forskningscenter Risø, Roskilde

Publication date:
1980

Document Version
Publisher's PDF, also known as Version of record

RISØ-M-2231

ACCELERATOR DEPARTMENT
Annual Progress Report 1 January - 31 December 1979

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, RISOE NATIONAL LABORATORY.

UDC 061.62 : 621.039 (489) Risø National Laboratory
UDC 621.384.6

April 1980
Risø National Laboratory, DK 4000 Roskilde, Denmark

ISBN 87-550-0670-1
ISSN 0418-6435

Risø Repro 1980

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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 1979. The contributions marked with § are abstracts of published articles.

1. OPERATION AND MAINTENANCE OF IRRADIATION FACILITIES

(J. Fenger and B. Lynggård)

1.1 HRC electron linear accelerator

The accelerator has been used to a normal extent. The weekly schedule for the operation is four days for experimental irradiation, mostly in connection with the pulse radiolysis equipment, half a day for service irradiation, and half a day for maintenance. During shut-down periods work has been carried out as follows:

Microwave transmitter: A new high voltage rectifier (delivered by Haimson Research Corp.) has been installed to replace the original, as the diodes used no longer are marketed. After half a year in operation, problems still arose due to breakdown of the diodes. These problems have been examined, and a rectifier based on a much more conservative concept might be designed by the department.

Injector system: The light pipes (lead-silica-core fiber) used for control of the 200 kV high voltage electron injector has been replaced. Due to radiation the absorption in the light pipes increases too fast, so they have to be replaced within a year. The light pipes can be recovered to some extent by heat treatment, but the lifetime is still too short and a better solution is under consideration.

Vacuum system: During a long period with too high a pressure in the beam handling system (up to 10^{-6} mm Hg), the vacuum system has been opened for replacement of a leaking bellows. One of the three 100 l/s pumps was replaced, and another was supplied with new pump modules. In the waveguide system connecting the klystron and the accelerator, a 30 l/s pump has been replaced, and the old one repaired by the factory.

For diagnostic purposes a new independent pressure detector system on the accelerator vacuum system has been installed.

The beam output window on the 90^{0} beam port has been replaced due to a leak caused by deformation of the thin, 0,5 mm, aluminum window. The deformation was caused by overheat; precaution

is now taken to avoid overheat by scanning the electron beam across the window area at all beam power levels.

Water cooling system: The water cooling system for the accelerator was in 1977 extended with a double bed deionizer in order to reduce the conductivity of the cooling water to less then 1.6 μS · cm. Unfortunately we got problems with the new system as the aggressive deionized cooling water in combination with the natural content of oxygen dissolves the copper from the cooling channels, and this copper shortened the deionizer. To solve the problem an oxygen filter has been installed and all components, except the ones made of copper and stainless steel, were removed. After half a year in operation the system still works satisfatorily.

1.2 Febetron, field emission accelerator

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

1.3 ICT, low energy accelerator

The low energy accelerator was used for dosimetry experiments. Due to a defect in the beam catcher the accelerator is now unserviceable. A new beam catcher will be installed in May 1980.

1.4 ^{60}Co-facilities

(E. Engholm Larsen and E. Bjergbakke)

Upgrading of the three ^{60}Co-facilities (the 3,000 Ci cell in the Accelerator Department, the 5,000 Ci cell at Statens Serum-institut, and the 10,000 Ci facility in the Agricultural Department).

In the autumn 1979 all three ^{60}Co-facilities were upgraded to the maximum capacity. The radioactive ^{60}Co was produced in

Research Reactor DR3 and the incapsulation was performed in Hot Cells. The capsules and various tools were constructed at the Engineering Workshop.

Upon removal of the eight lifting rods from the 10,000 Ci facility, they were checked for radioactive contamination. As no contamination was found, the complicated cleaning procedure of the ^{60}Co sources was omitted. The ^{60}Co sources from the 3,000 Ci cell were removed and transferred to Hot Cells for destruction. The old ^{60}Co sources from the 10,000 Ci facility were then transferred to the 3,000 Ci cell.

Two out of eight new ^{60}Co sources could not pass the leak test and had to be replaced. This delayed the reloading of the 10,000 Ci facility for more than one month. Some minor changes in the loading procedure and temporary shielding were introduced. The 5,000 Ci ^{60}Co-cell was upgraded by two new sources of 1,500 Ci each. Finishing the reloading all three facilities were reassembled and calibrated.

1.4.1 10,000 Ci ^{60}Co-facility

The 10,000 Ci ^{60}Co-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.

1.4.2 5,000 Ci ^{60}Co-facility

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

1.4.3 3,000 Ci ^{60}Co-facility

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

1.5 Reference

E. Engholm Larsen and E. Bjergbakke. Udskiftning af ^{60}Co-kilder. To be published as Risø-M-report.

2. EXPERIMENTAL EQUIPMENT

2.1 Light intensifier

(J. Fenger)

In connection with the pulse radiolysis equipment an apparatus for intensity increase of the light level of a Varian VIX-150W Xenon lamp has been developed. The light intensity is increased by a factor of 125 at 220 nm. The lamp is operated with a constant current of 12 A and for a short period of time up to 10 ms boosted with a pulse current up to 250 A; maximum tolerated pulse energy dissipated in the lamp is 10 J.

2.2 Experimental equipment for irradiation of the dye film dose meter with high LET particles

(J.W. Hansen and M. Wille)

In order to measure the exact dose to the dye film when irradiated with heavy ions it has been necessary to construct special equipment which satisfies requirements demanded for the microdosimetric investigations. The dose is determined by the fluence of particles picked up in a Faraday cup and measured as an integrated charge. The problem arises when the absorption in the film material is considerable, or when a change of charge state is possible.

The equipment consists of a vacuum chamber which includes a Faraday cup and a water cooled collimator to limit the defocused beam to a suitable geometry for the film target. The film is mounted on the periphery of a rotating wheel and is in this way

irradiated in sequences inbetween which the beam current is measured.

An electronically regulated motor drives the wheel and the electronic equipment, and besides counting the number of revolutions starts and stops the ion beam. The vacuum chamber is mounted on top of a diffusion pump and is pumped down to 10^{-6} torr. The equipment is designed in such a way that it can be used directly on the Van de Graaff accelerator at Niels Bohr Institute as well as on the Linac in the department.

2.3 Position monitors for the linear accelerator

(J.W. Hansen and M. Wille)

Monitors for determination of the beam position and beam current is under construction. Two beam tubes with welded flanges are fabricated and vacuum-leak tested. The tubes contain the coaxial line connections to the loopes picking up the electromagnetic field generated by the substructure of the electron beam.

2.4 Calorimeter for the ICT accelerator

(J.W. Hansen and M. Wille)

A calorimeter for absolute measurement of dose from the low energy accelerator has been modified with respect to the temperature sensing circuit and the beam absorbing calorimetric body. The temperature response is now much faster and more exact. The calorimeter will be used for the determination of energy and dose rate dependence of the dye film dose meter as well.

2.5 Reference

J. Fenger, High Intensity Stabilized Pulsed Analyzing Lamp for Ultraviolet Transient Spectrometry. To be published.

3. PHYSICAL DOSIMETRY AND TECHNOLOGICAL APPLICATION OF RADIATION

3.1 Radiochromic dye film dose meters

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

Thin plastic dose meters containing radiochromic dyes are being produced. A continued effort to optimize the properties of these dose meters is maintained. A part of this work is performed under a research contract with IAEA (2051/RB). Two progress reports have been submitted during 1979 and papers including these and other measurements of the properties of plastic dose meters were published. During a visit to the University of Mexico three lectures were given describing general dosimetry problems and the state of the art of radiochromic film making and measurements.

During the year work has been centered on the polyvinyl butyral (PVB) film with hexa-(hydroxyethyl)-pararosaniline cyanide. The measurements indicated that the sensitivity of the dose meter decreased during storage and that the induced optical density increased 6-10% after irradiation. The latter effect has been effectively reduced by a new formulation of the solution from which the film is cast, while the former effect still seems to exist.

The dose meter films are sensitive to UV-light. We have tried to utilize this effect to enhance the response of the dose meters by placing a dose meter between phosphor tablets that emit UV-light during irradiation. The response has been increased 2-8 times using this technique, but drawbacks in the form of changes of properties are possible.

3.2 Dose measurements

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

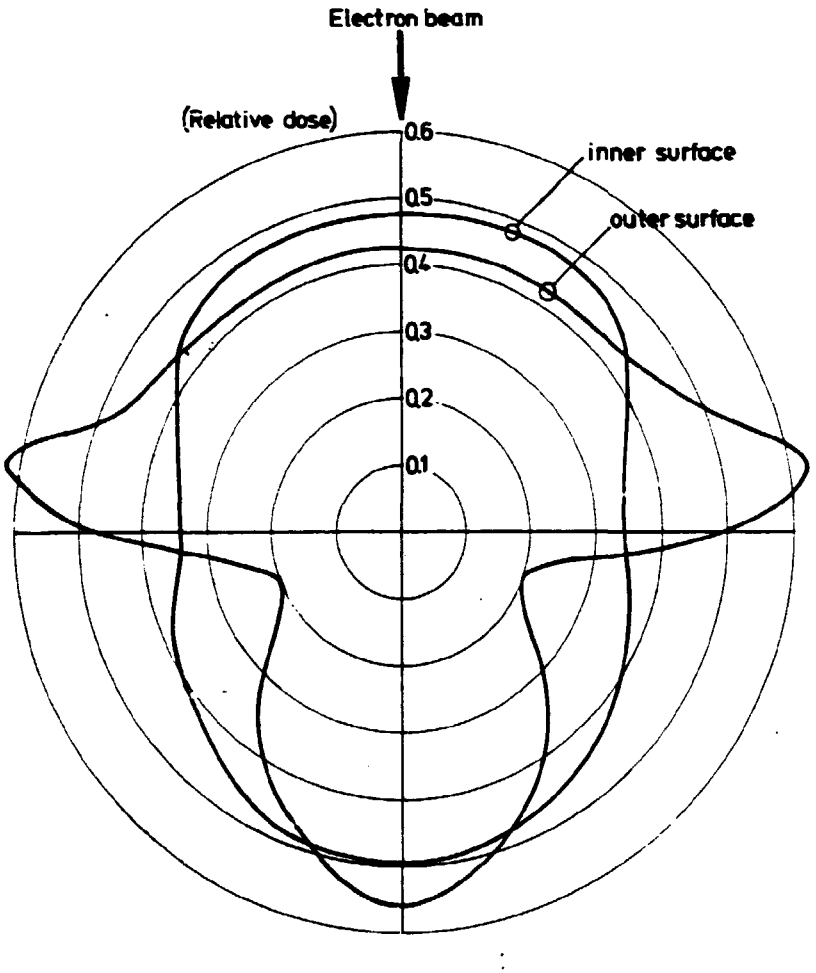
The dose meter films were used for several dose and dose distribution measurements at different experimental set-ups at the linear electron accelerator. Papers were published about dose distributions in cylindrical targets and earlier interferometric measurements.

3.3 Microdosimetry

(J.W. Hansen, M. Jensen (Statens Strålskydds Institut, Stockholm, Sweden), R. Katz (University of Nebraska-Lincoln, Lincoln, NE, USA), and W.L. McLaughlin (National Bureau of Standards, Washington, D.C., USA))

The microdosimetrical use of the thin radiochromic dye film in a test of the track theory of Katz has caused detailed investigations of the film behaviour at high doses, where the dose response characteristic, contrary to expectation, deviates from the characteristic of a one-hit detector. In order to sort out phenomenalistically what determines the difference in maximum optical density, the bleaching effect, fading, and build-up, when the film is irradiated with low LET radiation and heavy ions to saturation doses, the influence from oxygen and dose rate has been investigated for electrons and γ-ray photons. When irradiated in a nitrogen atmosphere the film shows a less pronounced bleaching and fading effect, whereas the difference in saturation optical density is unchanged. The build-up effect is much more pronounced when irradiated in vacuum.

A chamber for irradiating the film samples in vacuum is under construction. The purpose is to measure the beam current to the film during irradiation with heavy ions. Some of these ions are stopped in the film and some are changing charge state when penetrating the film. Further the chamber will provide the same irradiation condition for all radiation qualities except for γ-ray photons.



Dose distribution on the outer and inner surface of a polyethylene tube (diameter 100 mm, wall thickness 10 mm) irradiated from one side with 10 MeV electrons.

The group has assisted visiting scientists with beam parameter measurements and calculations in which computer programs for the HP 9830 desk top calculator have been used.

3.4 International dose intercomparison

(A. Miller)

IAEA is organizing a study with the aim of establishing an international dose measurement service for commercial irradiation plants. At a meeting in Budapest the program status and IAEA's coordinated research program for high-dose standardization were reviewed. The research contract (2051) is a part of this research program and a lecture was given summarizing the results so far.

3.5 Induced radioactivity

(A. Miller and P. Hedemann Jensen (Safety Dept.))

The absorption of high energy electrons creates bremsstrahlung that through nuclear reactions can create isotopes, some of which may be radioactive. In cooperation with the Safety Dept. a contract proposal has been sent to IAEA, suggesting to investigate the possibility of creating radioactive isotopes at the 10 MeV linear accelerator.

3.6 Commercial activities

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

In cooperation with the Chemistry Dept. an effort is made to promote ionizing radiation as an industrial process. Test irradiations have been performed for several firms; we took part in a development program for polymer modification, firms were invited to a meeting at Risø with Prof. J. Silverman, University of Maryland, we produced posters for exhibitions, and an article was published in the Danish journal "ingeniøren".

3.7 References

A.G. Karadjov and Johnny W. Hansen, Estimation of Electron Dose Delivered by a 0.4 MeV Accelerator from Bremsstrahlung Dose Measurements. Radiat. Phys. Chem., accepted for publication.

Johnny W. Hansen and Bent Lynggård, Calculator Programs for Transient Temperature and Stress Calculations on Thin Targets. Risø-M-2112, to be published.

Johnny W. Hansen, Conceptual Basis for the Radiochromic Dye Film Dose Meter as a Test of Particle Track Theory. To be published as Risø-M-report.

4. CHEMICAL DOSIMETRY AND RADIATION CHEMISTRY

4.1 Calibration of ^{60}Co-facilities[§]

(H. Corfitzen, E. Bjergbakke and K. Sehested)

After the upgrading the three ^{60}Co-facilities were recalibrated by the Fricke and the "super" Fricke dosimeter. The 10,000 Ci facility was calibrated in all three channels with 8,4 and 1 source.

4.2 Computer simulation of the reactions in the Fricke dosimeter with varying Cl^- concentrations

(E. Bjergbakke)

This work is performed in collaboration with J. Swallow, Christie Hospital & Holt Radium Institute, Manchester. Based on experimental data obtained in Manchester, we tried to work out the reaction mechanism for the Fricke dosimeter containing chloride. Computer simulation was used to check the mechanism. The mechanism is still incomplete, and we plan to look into the reactions of Cl_2 and H_2O_2 or the intermediates in their reactions with Fe^{++} to find a possible explanation.

4.3 Pulse radiolysis of Cl^- solutions

(J. Holcman and E. Bjergbakke)

OH radicals form a radical ion with chloride ions, which is in equilibrium with free OH radicals $ClOH^- \rightleftarrows Cl^- + OH$. In N_2O

saturated solutions, however, the observed build-up of the absorption cannot be ascribed to this equilibrium. The build-up had tentatively been ascribed to a protolytic reaction of $ClOH^-$, ($ClOH^- \rightleftarrows ClO^{--} + H^+$). This reaction, however, can be excluded as we find that an increase of pH has no effect or the opposite effect than expected on the build-up kinetics and optical absorption. Pulse radiolytic experiments, however, indicate the reaction $ClOH^- + Cl^- \rightarrow Cl_2^- + OH^-$ (1). An analogical reaction is known for bromide ions. Because of a relatively slow rate and a complex competition from other reactions, reaction (1) cannot be separated and definitively proved alone by pulse radiolytic experiments. Therefore computer simulation has been applied. The results of the computer simulation are consistent with the pulse radiolytic experiments allowing for an estimation of the rate constant of the reaction $k_1 = 2 \times 10^5\ M^{-1}s^{-1}$.

4.4 Pulse radiolysis of hydrogen pressurized aqueous NaCl solutions

(J. Holcman, E. Bjergbakke and K. Sehested)

One of the problems in radioactive waste disposal in salt formations is the radiolysis of brine, which will migrate towards buried waste containers. Under radiolysis of brine, mixtures of oxygen and hydrogen are formed. The steady state concentrations under long term irradiation are therefore of great interest. The aim of the present study is to clarify the mechanisms of the hydrogen consumption. It was found that hydrogen is consumed by $ClOH^-$ radicals with the rate constant of the same order of magnitude as for the OH radical. A reaction between Cl_2^- and hydrogen has not been observed at pressures up to 100 atm H_2. The study is continued with the aim to measure the yield of Cl_2^- formed in spurs and/or by the direct effect (direct energy absorptions) as function of the chloride ion concentration.

4.5 Radiolysis of salt-brine[§]

(E. Bjergbakke)

This work was done according to the agreement between Risø and ELSAM/ELKRAFT concerning advisory assistance from Risø to ELSAM/ELKRAFT's waste management project, phase 2.

Salt formations of the type considered for radioactive waste disposal contain a small fraction of brine inclusions. The brine will migrate towards a buried waste container and will thus be exposed to γ-radiation. In this work a reaction mechanism for the radiolysis of concentrated sodium chloride is constructed on the basis of the experimental and theoretical data available. Some experiments have been performed in order to make an estimate of the rate constants of a few unknown reactions. The reaction mechanism forms the basis for the computation of the radiolytic decompositon yields by numerical integration of the complete set of chemical equation. Radiolysis of concentrated NaCl solutions leads to formation of H_2 and O_2. The highest obtainable H_2 concentration is of the order of 0.2 M corresponding to a partial hydrogen pressure of 300 atm at room temperature. Variations in dose rate do not change the maximum concentration significantly, but the rate of the build-up of hydrogen concentration is proportional to the dose rate. At a dose rate of 20 krad/hour the maximum concentration will be reached in 1 year. At a dose rate of 2 krad/hour in 10 years and at a dose rate of 1 rad/hour in 20.000 years. In a time scale of many years the system cannot be regarded as a closed system; the hydrogen will disappear by diffusion and high hydrogen concentrations will never be obtained.

4.6 Direct effect of radiation on ClO_4^-

(K. Sehested, E. Bjergbakke, and E.J. Hart (Port Angeles, WA, USA))

The ClO_4^- ion is extremely unreactive with the primary radicals from the water radiolysis. Therefore this ion is of potential interest for studying direct effects from radiation in very concentrated solutions. Many attempts have been made, but up

to now the mechanism is not well understood. By employing pulsed irradiations and an advanced gaschromatographic technique together with other product determination methods it should be possible to get a better knowledge of the reaction mechanism. This project has also some implications on the $O(^3P)$ study.

The measurements in concentrated solutions (1 to 6 M $NaClO_4$) have shown a formation of ClO_3^-, O_2, and H_2O_2, probably also Cl^-. The main difference from the previous determinations are in the ratio of O_2 and H_2O_2 yields. These two products must be protected from attacks from the water radicals, which is achieved in a better way by using a short powerful electron pulse than in γ-irradiations. Our preliminary results indicate that the excited perchlorate ion mainly emits an oxygen atom forming chlorate ions. It is suggested that this oxygen atom is in the singlet state as very little triplet state oxygen atoms are found. The singlet oxygen atom is known to react very fast with water forming hydrogenperoxide, and indeed we find an increased yield of H_2O_2. This mechanism may account for the main reaction, but does not complete the material balance. The small amount of O_2 and possibly also Cl^- may either come from competing reactions or from another decay pathway of the excited perchlorate ion.

4.7 $O(^3P)$ yields in alkaline aqueous solutions

(E. Bjergbakke, K. Sehested and E.J. Hart (Port Angeles, WA., USA))

The mechanism causing the increase in $O(^3P)$ yield at pH 12 to 14 is assumed to be direct exitation of OH^-: $(OH^-)^* \rightarrow O(^3P) + H^-$ and $H^- + H_2O \rightarrow H_2 + OH^-$. In the system, formate, N_2O and O_2, H_2 will be protected from radical attack. We measured an increase in H_2 yield of 5% with the increase of pH from 10 to 13. The uncertainty of the gaschromatographic measurement of small amounts of H_2 in the presence of big amounts of O_2 is however of the order of 5%, so further experiments are needed to establish a 5% increase.

4.8 Pulse radiolysis at high temperatures and high pressures

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

The experiments have been concentrated on the spectrum of the OH radical as function of the temperature and the activation energy of the reaction OH + OH → H_2O_2. The spectrum is changed very little below ~280 nm up to temperatures of 150-170°C, whereas the Gε seems to increase above this wavelength. Some complications arose at higher temperatures (200-220°C) because of impurities from the irradiation cell. These impurities react with the primary radicals giving absorptions at a longer wavelength. A new cell of a purer suprasil is now employed. The spectra were taken in N_2O-saturated neutral water with a low dose per pulse. The determination of the activation energy of the reaction OH + OH → H_2O_2 was measured from the decay kinetics in a similar solution, but with a much higher dose per pulse (10 krad), to obtain a reasonable good second order plot over at least 2 half lives. The activation energy is evaluated to ~10.5 kJ x mol^{-1} (2.5 kcal x mol^{-1}) up to 150°C. Additional experiments were performed in Ar-saturated solutions of H_2O_2.

Another reaction of interest is Fe^{2+} + OH → $Fe^{2+}(OH)$. We determine the activation energy to ~9 kJ x mol^{-1} (2.2 kcal x mol^{-1}) up to 220°C. The reaction was studied in N_2O-saturated solution at different pH's ($HClO_4$) and with different ferrous concentrations. The build-up of $Fe^{2+}(OH)$ was measured in UV (240-300 nm), and the first order rate constant is found to vary from 3 x 10^8 $M^{-1}s^{-1}$ at 20°C to 2 x 10^9 $M^{-1}s^{-1}$ at 220°C.

4.9 Iodine (VI) in aqueous solution[§]

A Laser Flash Photolysis and Pulse Radiolysis Study.

(U.K. Kläning (Kemisk Institut, Aarhus Universitet), K. Sehested, and Th. Wolff (Universität Gesamthochschule, Siegen, Germany))

Laser flash photolysis and pulse radiolysis of aqueous iodate and periodate solution show that iodine in the unstable oxidation state six may be formed in the same solution as three different species depending on the reaction in which the species

is formed. The reactions of iodine (VI) species with the solvent with formation of a more stable iodine (VI) species are extremely slow. The possible structures of various iodine (VI) species are discussed on the basis of characteristic features of the reaction in which iodine (VI) species are formed.

On basis of kinetic measurements of the reaction $IO_3^- + O^- \rightleftarrows IO_4^{2-}$ and publis ed thermodynamic data the standard Gibbs energy of formation of IO_4^{2-} was estimated to be -57 kJ x mol^{-1}.

4.10. The photcchemistry and radiochemistry of perxenate and xenon trioxide in aqueous solution

(E. Appelman (Argonne National Laboratory, Argonne, Ill., USA), U.K. Kläning (Kemisk Institut, Aarhus Universitet), K. Sehested, and Th. Wolff (Universität Gesamthochschule, Siegen, Germany))

Pulse radiolysis of aqueous perxenate and xenon trioxide solutions show that the e_{aq}^--adducts and the O^- (OH) adducts of perxenate and xenon trioxide, Xe^V, Xe^{VII} and Xe^{IX} in general have properties very similar to the analogous iodine species I^{IV}, I^{VI} and I^{VIII}. A characteristic difference is however that the dinuclear xenon species (Xe^{VII},Xe^{VIII}), (Xe^{VIII},Xe^{IX}) and Xe^{VI},Xe^{IX}) are formed more willingly than the corresponding iodine species and playing a more important role in subsequent reactions than the corresponding iodine species.

Photochemical experiments including steady state, flash photolysis and laser flash photolysis measurements show that a primary process in the photolysis of perxenate is $Xe^{VIII} \rightarrow Xe^{VII} + O^-$, a process which is analogous to the primary process in the photolysis of periodate: $I^{VII} \rightarrow I^{VI} + O^-$. In addition there is some indirect evidence for a primary process in which molecular oxygen is formed, a process which is not observed in photolysis of periodate.

4.11 Activation energy of formation and decay of radical cations and phenoxy radicals

(K. Sehested and J. Holcman)

The activation energy of various reactions is determined in a thermostated optical cell between 20° and 90°C.

The formation and decay of the isodurene radical cation as measured at pH 4 and 1 respectively at the optical absorption at 450 nm both yield an activation energy of 25 kJ x mol^{-1} (5.9 kcal x mol^{-1}). The activation energy for the radical cation formation from various methoxy compounds was also determined.

The activation energy for the formation of phenoxy radicals from phenol is found to be 22.5 kJ x mol^{-1} (5.3 kcal x mol^{-1}), whereas from hydroquinone it is measured to 18 kJ x mol^{-1} (4.3 kcal x mol^{-1}).

The reaction of anisole radical cation with anisole has a rather small activation energy of 8 kJ x mol^{-1} (1.9 kcal x mol^{-1}).

4.12. A pulse radiolysis study of the protonated dimethylaniline reaction with OH and e^-_{aq}

(I. Karaivanov (Institute of Nuclear Research and Nuclear Energy, Sofia, Bulgaria) and J. Holcman)

Aqueous solutions of dimethylaniline (DMA) saturated with Ar, N_2O and O_2, at pH 7 and lower, have been studied. The rate constant for the reaction of e^-_{aq} with the protonated dimethylaniline ($DMAH^+$) was found to be $K(e^-_{aq} + DMAH^+) = 5 \times 10^9\ M^{-1}s^{-1}$. On the basis of this value and the known values of K(OH + DMA) and $K(OH + DMAH^+)$ it is assumed that all e^-_{aq} and OH radicals react with $DMAH^+$ at pH 3, if high concentrations of $DMAH^+$ (10^{-1} M) is used. A concentration effect can thus be studied. The absorption bands at 300 - 340 nm and 400 - 490 nm increase considerably when the $DMAH^+$ concentration is increased in Ar-saturated solutions at pH 3-5. A reaction between the adduct

of e^-_{aq} with $DMAH^+$ and DMA(OH) and/or $DMAH^+$(OH) could be responsible for this concentration effect.

At pH lower than 3, when all e^-_{aq} are converted into H atoms, this effect was not observed. Under these conditions the absorption at 460 nm decays by second order kinetics which probably reflect a recombination reaction between the OH and H adduct of $DMAH^+$.

In order to get information about the reaction between e^-_{aq} and $DMAH^+$, Ar-saturated solution at pH 5 containing butanol or methanol have been studied. No absorption was found in these systems, nevertheless the absorption at 720 nm shows that all e^-_{aq} are scavenged by $DMAH^+$.

4.13. The kinetics of dye formation by pulse radiolysis of pararosaniline cyanide in aqueous or organic solution[§]

(W.L. McLaughlin (National Bureau of Standards, Washington, D.C. USA), M.M. Kosanić (Faculty of Science, University of Novi Sad, Novi Sad, Yugoslavia), V.M. Marković and M.T. Nenadović (Radiation Chemistry Dept., Boris Kidrić Institute of Nuclear Science, Vinča, Belgrade, Yugoslavia), J. Holcman and K. Sehested)

The radiation-induced conversion of the leuco-cyanide of pararosaniline dye, $[H_2N\langle\bigcirc\rangle]_3{\equiv}C\text{-}CN$, to the highly colored salt-isomer of the dye, $[H_2N\langle\bigcirc\rangle]_2{=}C{=}\langle\bigcirc\rangle{=}NH_2^+ + CN^-$, in acidic aqueous solution (wavelength of maximum absorption λ_{max} = 540 nm) or polar organic solution (λ_{max} = 550 nm), takes place in two separate processes. The first is very fast (within <50 ns), and the second much slower following first-order kinetics with a rate constant that varies from 4×10^3 s^{-1} to $\sim 10^6$ s^{-1}, as the acidity or concentration of an oxidizing agent increases. In oxygen-free acidic aqueous or organic solutions (Ar-sat.) there is an unstable transient species (λ_{max} = 380 nm). When using O_2 or N_2O-saturated aqueous or organic solution, there is no intermediate absorption band at 380 nm, but the slow process of dye formation at 540 or 550 nm is still sequential to the initial fast process having somewhat faster kinetics than in Ar-saturated solution.

4.14. Oxygen production in the reduction of ferric tris-bipyridyl complex with OH^-

(B. Pedersen, G. Nord (H.C. Ørsted Institut), and E. Bjergbakke)

The Fe-bipyridyl complex was proposed for a homogeneous aqueous system for production of hydrogen by use of solar energy. The suggested overall reactions were:

$$Fe^{+2}(bipy)_3 + h\nu \rightarrow Fe^{+3}(bipy)_3 + \tfrac{1}{2}H_2$$

and the reduction

$$Fe^{+3}(bipy)_3 + OH^- \rightarrow Fe^{+2}(bipy)_3 + \tfrac{1}{4}O_2$$

We studied the reduction reaction by measuring the O_2 yield using the Van Slyke GC system. By dissolving dry crystals of $Fe^{+3}(bipy)_3$ in a strong NaOH solution (O_2-free), we proved that no oxygen is formed in the reduction reaction. When we dissolve the $Fe^{+3}(bipy)_3$ in acid solution and then mix it with a strong alkaline solution, oxygen is formed. The yield of oxygen increases with the time t measured from the dissolution to the mixing until it reaches a maximum at t_{max} and then the yield decreases with increasing time. t_{max} depends on the pH of the acid solution; it also depends on the concentration of other ligands in the solution (Cl^-, SO_4^-). We conclude that the intact $Fe^{+3}(bipy)_3$ complex does not produce O_2 by reduction with OH^-, whereas one of the intermediates in the dissociation of the complex do produce oxygen.

4.15. References

H. Corfitzen, E. Bjergbakke and K. Sehested, Calibration of the ^{60}Co-facilities. To be published as Risø-M-report.

E. Bjergbakke, Radiolysis of Salt-Brine.
This report has been worked out according to the agreement between Risø National Laboratory and ELSAM/ELKRAFT concerning advisory assistance from Risø to ELSAM/ELKRAFT's waste management project, phase 2. February 1980.

H. Christensen and K. Sehested, Pulse Radiolysis at High Temperatures and High Pressures. Accepted for publication in Radiat. Phys. Chem.

U.K. Kläning, K. Sehested and Th. Wolff, Iodine (VI) in Aqueous Solution. A Laser Flash Photolysis and Pulse Radiolysis Study. Submitted to J. Chem. Soc. Faraday Trans. I.

E. Appelman, U.K. Kläning, K. Sehested and Th. Wolff, The Photochemistry and Radiochemistry of Perxenate and Xenon Trioxide in Aqueous Solution. Submitted to J. Chem. Soc. Faraday Trans. I.

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. Proteins induced by irradiation of M. radiodurans

(M. Trier Hansen)

Four proteins, α, β, γ and δ, preferentially synthesized in ultraviolet light treated cells of M. radiodurans, were characterized in terms of their molecular weights and isoelectric points. Within the sublethal dose-range the differential rate of synthesis for these proteins increased linearly with the inducing UV-dose. The degree of derepression reached one hundred fold and the most abundant protein, β, amounted to approximately two per cent of the total newly synthesized protein after irradiation. Damage caused by ionizing electron radiation or by treatment with mitomycin C also provoked the synthesis of the four proteins. In contrast to treatments which introduced damage in the cellular DNA, the mere arrest of DNA replication, caused by nalidixic acid or by starvation

for thymine, failed to elicit the synthesis of either protein. The induced proteins are proposed to participate in the versatile and efficient processes of DNA repair in M. radiodurans.

5.2. Repair of psoralen-DNA adducts in E. coli

(M. Trier Hansen)

A mutation, which leads to greatly increased sensitivity to psoralen in E. coli, was shown to map at 10 minutes on the bacterial chromosome. Mutants at this site (acrA) have recently been found lacking in outer membrane phospholipid. This alters membrane permeability and leads to hypersensitivity to hydrophobic antibiotics. Besides this general unspecific effect, phospholipids have been suggested to facilitate repair DNA-synthesis. To test this, experiments were initiated to compare repair capabilities of the acrA mutant and wild type strains. Reactivation of bacteriophage λ, which had been damaged outside the cell before infection, was used as a measure of repair proficiency.

5.3. W-reactivation in Acinetobacter calcoaceticus

(D. Berenstein)

Studies on W-reactivation in Acinetobacter calcoaceticus were 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. Phage development in bacteria induced for W-reactivation was compared to phage development in unirradiated bacteria by the so-called one-step experiments. Phage stocks used for these experiments were prepared in big volumes, concentrated more than one hundredfold and carefully purified to assure good adsorption.

The experiments revealed that induction of W-reactivation decreases the average phage yield; the decrease is twofold for the unirradiated phage, and about fivefold for the irradiated one. Also, there is a delay in phage production where the phage

is UV-irradiated. The delay is independent of W-reactivation. The extended period of phage production may be caused by the presence of radiation damage in the phage DNA. Production of a smaller phage yield by the irradiated cells may suggest that cellular metabolism is somewhat impaired by the irradiation dose.

5.4. Examination of geothermal water Aars 1-a for occurrence of sulphate-reducing bacteria

(D. Berenstein)

This work was done according to the agreement between Risø and Dansk Olie & Naturgas A/S (DONG) concerning geothermal water.

Three different procedures were employed in order to detect the sulphate-reducers: 1) incubation of water samples in fluid, deoxygenized medium; 2) incubation of plates in anaerobic jars; 3) incubation of water samples mixed with nutrient agar in tubes. None of the methods detected sulphate-reducing bacteria in the geothermal water.

5.5. 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 cerius, 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.

5.6. 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.

5.7. References

M. Trier Hansen, Four Proteins Synthesized in Response to DNA Damage in Micrococcus radiodurans. J. Bacteriol. 141 (1980) 81-86.

D. Berenstein, Examination of Geothermal Water Aars 1-a for Occurrence of Sulphate-Reducing Bacteria.
This work was done according to the agreement between Risø and Dansk Olie & Naturgas A/S (DONG) concerning geothermal water.

6. EDUCATIONAL ACTIVITIES AND PUBLICATIONS

6.1. Lectures

D. Berenstein, W-reactivation in Acinetobacter. Sixth International Congress of Radiation Research, Tokyo, Japan, May 16.

D. Berenstein, Strålesterilisation af medicinsk éngangsudstyr (Radiation Sterilization of Medical Disposable Equipment) for studerende ved Danmarks Sygeplejeskole. Risø, June and October.

E. Bjergbakke, Radiolysis of Concentrated Aqueous NaCl Solutions. 6th Danish-Polish Symposium on Radiation Chemistry, Warsaw, Poland, October 22-24.

J.W. Hansen, Conceptual Basis for the Radiochromic Dye Film Dose Meter as a Test of Particle Track Theory. 6th Danish-Polish Symposium on Radiation Chemistry, Warsaw, Poland, October 22-24.

M. Trier Hansen, Radioresistance of Micrococcus Radiodurans. Laboratoriet for Mikrobiologi, Danmarks tekniske Højskole, August 28.

E.J. Hart, O(^{3}P) Atom Formation in γ-Ray Irradiation and Photolyzed Aqueous Solution. Risø, December 13.

R. Katz, The Structure of Particle Tracks, with Applications to Radiation Dosimetry and Radiobiology. Risø, July 17.

A. Miller, Thin Film Dosimetry at the Risø Accelerator Department. National Bureau of Standards, Washington, D.C., USA, August 30.

A. Miller, Radiochromic Dye Film Dosimetry. Part 1: General. Part 2: Properties of the radiochromic dye dose meter and other plastic dose meters. Part 3: Applications of the radiochromic dye film dose meters. Universidad Nacional Autonoma de Mexico, Mexico 20, D.F., September 3-14.

A. Miller, Physical Dosimetry at the Accelerator Department, Risø. Instituto Nacional de Investigaciones Nucleares, Mexico City, Mexico, D.F., September 12.

A. Miller, Stability of the Radiochromic Dye Film Dose Meter. 6th Danish-Polish Symposium on Radiation Chemistry, Warsaw, Poland, October 22-24.

A. Miller, Investigation of the Radiochromic Dye Film Dosimeter. IAEA Coordinated Research Meeting on High Dose Standardization and Intercomparison for Industrial Processing. Budapest, Hungary, November 11-16.

6.2. Publications

Accelerator Department Annual Progress Report (1979), 1 January - 31 December, 1978. Risø-M-2173.

E. Bjergbakke (1979), The Radiation Chemistry of Electron- and Gamma-irradiated Aqueous Solutions of Fe^{2+} and Cu^{2+}. Nukleonika, Vol. 24, No. 9, 825-845.

Z.D. Draganić, I.G. Draganić, and K. Sehested (1979), The Radiation Chemistry of Aqueous Solutions of Dicyandiamide. J. Phys. Chem. 83, 220.

J.W. Hansen and P. Lundsager (1979), A Technique for Temperature and Ultimate Load Calculations of Thin Targets in a Pulsed Electron Beam. Nuclear Instruments and Methods 160, 203-210.

J. Holcman and K. Sehested (1979), Pulse Radiolysis Study of Aqueous p-Toluonitrile. Nukleonika, Vol. 24, No. 9, 879-885.

J. Holcman and K. Sehested (1979), The Relation Between the Acid-Base Behaviour of the OH Adducts of Substituted Benzenes and Their Ionization Potential. Nukleonika, Vol. 24, No. 9, 887-892.

H. Levine, W.L. McLaughlin, and A. Miller (1979), Temperature and Humidity Effects on the Gamma-Ray Response and Stability of Plastic and Dyed Plastic Dosimeters. Radiat. Phys. Chem. 14, 551-574.

W.L. McLaughlin, M.M. Kosanić, V.M. Marković, M.T. Nenadović, J. Holcman, and K. Sehested (1979), The Kinetics of Dye Formation by Pulse Radiolysis of Pararosaniline Cyanide in Aqueous or Organic Solution. Risø-M-2202.

W.L. McLaughlin, A.C. Lucas, B.M. Kapsar, and A. Miller (1979), Electron and Gamma-Ray Dosimetry Using Radiation-Induced Color Centers in LiF. Radiat. Phys. Chem. 14, 467-480.

W.L. McLaughlin, J.C. Humphreys, B.B. Radak, A. Miller and T.A. Olejnik (1979), The Response of Plastic Dosimeters to Gamma Rays and Electrons at High Absorbed Dose Rates. Radiat. Phys. Chem. 14, 535-550.

A. Miller (1979), Mere end 300 bestrålingsanlæg. Industriel anvendelse af stråling. Ingeniøren, Nr. 10.

A. Miller (1979), 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 Nov. 1978 - 30 Apr. 1979, and 1 May - 31 Oct. 1979). IAEA Research Contract 2051/R1/R2/RB. Risø-I-15.

A. Miller (1979), Beam Spot Measurements on a 400 keV Electron Accelerator. Radiat. Phys. Chem. 13, 1-4.

A. Miller, W.L. McLaughlin, W. Batsberg Pedersen, and K. Pejtersen (1979), Absorbed Dose Distributions in Small Copper Wire

Insulation due to Multiple-Sided Irradiations by 0.4 MeV Electrons. Radiat. Phys. Chem. 13, 181-186.

A. Miller (1979), Holography and Interferometry in Dosimetry. Nukleonika, Vol. 24, No. 9, 907-925.

A. Miller (1979), Radiochromic Dye Film Dosimetry. Part 1: General. Part 2: Properties of the radiochromic dye dose meter and other plastic dose meters. Part 3: Applications of the radiochromic dye film dose meters. IFUNAM-79-24-25-26. Universidad Nacional Autonoma de Mexico, Mexico 20, D.F.

A. Miller and W.L. McLaughlin (1979), Absorbed Dose Distributions in Irradiated Plastic Tubing and Wire Insulation. Radiat. Phys. Chem. 14, 525-533.

K. Sehested and J. Holcman (1979), Radical Cations of Ethyl-, Isopropyl- and Tert-Butylbenzene in Aqueous Solution. Nukleonika, Vol. 24, No. 9, 941-950.

6.3. Test-irradiations

Test-irradiations were carried out for:

Ciconia, Åbybro
Danmarks tekniske Højskole, Lyngby
Den kgl. Veterinær- og Landbohøjskole, København
K. Balling Engelsen A/S, Maribo
Fibiger-Laboratoriet, København
Fiseninstitutet, København
Gambro AB, Lund, Sweden
Københavns Universitet:
    Audiologopædisk Forskningsgruppe
    Proteinlaboratoriet
Microtronic Aps, Roskilde
Mölnlycke Steritex A/S, Espergærde
Novo Industri A/S, Bagsværd
Ortopædisk Hospital, Århus
Peerless Fabrikkerne A/S, København
Radiflex Aps, Hedehusene
Statens Seruminstitut, København

Planteavlsleder V. Schelbeck, Agedrup

Triax A/S, Hornsyld

Århus Universitet, Århus.

6.4. Visiting scientists

H. Nantin, Tetra Pak, Lund, Sweden.

K. Ennow, Statens Institut for Strålehygiejne, København.

H. Christensen, Studsvik Energiteknik AB, Studsvik, Sweden.

R. Jess Jørgensen, Institut for Veterinær Mikrobiologi og Hygiejne, København.

H. Dan, Cattle Research Institute, Corbeanca, Romania.

F. Dér, Kaposwar, Hungary.

U. Kläning, Kemisk Institut, Århus Universitet, Århus.

H.J. Peters, High Voltage Engineering Europa B.V., Amersfoort, The Netherlands.

S. Madsen, A/S NIRO Atomizer, Søborg.

D.A. Vroom, IRT Corporation, San Diego, CA., USA.

P.O. Gravesen, Mölnlycke Steritex A/S, Espergærde.

M. Symons, Leicester University, Leicester, England.

M. Rae, National Radiological Protection Board, Harwell, England.

R. Katz, Behlen Laboratory of Physics, University of Nebraska-Lincoln, Lincoln, USA.

W.L. McLaughlin, National Bureau of Standards, Washington, D.C. USA.

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

I. Karaivanov, Institute of Nuclear Research and Nuclear Energy, Sofia, Bulgaria.

7. IRRADIATION FACILITIES AT THE ACCELERATOR DEPARTMENT

Electron Accelerators

1. Linear Electron Accelerator, Haimson Research Corp. 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 μs
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 ± 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 μs interval, better than ..... ± 1%

Accelerator room beam facilities:

1. A bent beam with scan width of 40 cm providing a process irradiation capacity of 1000 - 1500 Mrad kg/hour.
2. A horizontal beam, full average beam peak power, for electron and X-ray irradiation.
3. A horizontal beam, reduced average beam power (12.5 pps) in connection with a ± 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.

^{60}Co-Facilities

10,000 Ci ^{60}Co-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 (10,700 Ci, 1 January 1980) are 6.5×10^5 rads/h, 1.8×10^6 rads/h, and 4.2×10^6 rads/h, respectively.

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

Designed for laboratory use and fitted with a 123 mm$^{\varnothing}$ x 150 mm irradiation chamber. The dose rate in the center of the chamber (6,500 Ci, 1 January 1980) is 5.6×10^5 rads/h. The cell is located at the Control Department, Statens Seruminstitut, Copenhagen.

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

Designed for laboratory use and fitted with a 120 mm$^{\varnothing}$ x 200 mm irradiation chamber. The dose rate in the center of the chamber (4,100 Ci, 1 January 1980) is 4.1×10^5 rads/h.

8. STAFF OF THE ACCELERATOR DEPARTMENT

Head: Knud Sehested
Office staff: Ebba Haugaard and Ruth Madsen

Scientific staff

Dvora Berenstein
Erling Bjergbakke
Jørgen Fenger
Ivan Karaivanov (from 26 September)
Mogens Trier Hansen
Johnny W. Hansen
Jerzy Holcman
Bent Lynggård
Arne Miller

Technical staff

Svend Bøjlund Andersen
Dorte Egelund Andersen (15 January - 30 April), Apprentice
Margit Elm Andersen (from 1 July)
Karen Boysen
Erling Cederström (from 1 October)
Karen Christiansen (1 May - 30 October), Apprentice
Hanne Corfitzen
Edvard Grys (1 January - 1 July)&(from 28 August)
Ina Hansen
Inge Høegh
Torben Johansen
Erik Engholm Larsen
Fritz Larsen
Inge Merete Larsen
Laurits Nielsen
Per Broen Pedersen (until 1 August)
Kresten Pejtersen

Consultants

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

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

W.L. McLaughlin, National Bureau of Standards, Washington, D.C., USA.

Risø National Laboratory **Risø - M - 2231**

Risø - M - 2231

Title and author(s)

Accelerator Department - Annual Progress Report for the period 1 January - 31 December 1979.

Acceleratorafdelingens årsrapport
1 januar - 31 december 1979.

Date April 1980

Department or group

Accelerator Dept.

Group's own registration number(s)

37 pages + tables + 1 illustrations

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.

Copies to

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