



Meterological measurement programs for air pollution studies

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ACCELERATOR DEPARTMENT
Annual Progress Report
1 January - 31 December 1985

Abstract. A description is given of research in the fields of reaction kinetics, physical dosimetry, radiation physics and technological application of radiation and irradiation technology, as well as of the operation of various irradiation facilities.

INIS-descriptors. ACCELERATOR FACILITIES; DOSEMETERS; DOSIMETRY; IRRADIATION DEVICES; REACTION KINETICS; RESEARCH PROGRAMS; RISØE NATIONAL LABORATORY

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PREFACE

The Accelerator Department conducts research in chemical reaction kinetics, dosimetry, and in processes based on ionizing radiation contributing to its industrial utilization.

At our disposal are three electron accelerators of 10-, 2-, and 0.4-MeV, cobalt-60 facilities of 3- and 10-kCi, and a Multigas Excimer Laser equipped with various experimental facilities.

A major research effort is directed towards understanding basic chemical processes better, in particular the reaction kinetics. The applied techniques include pulse- and steady-state radiolysis, laser flash-photolysis, stop-flow experiments, and computer simulations.

A proper measurement of the absorbed dose is a prerequisite for application of radiation in research and industry. Research is conducted into the response mechanisms of dosimeters and in the development and calibration techniques of dosimeters for gamma, electron, and heavy charged particle irradiations.

Commercial test irradiations are carried out at two of the electron accelerators and at both cobalt-facilities, but only the 10-MeV linear electron accelerator is equipped for full-scale industrial purposes. Consultive and advisory assistance is rendered in connection with commercial irradiation, including, e.g. calibration of a customer's dosimeter systems.

This report describes the principal activities in these fields for the period 1 January - 31 December 1985.

The contributions marked * are abstracts of published papers.

1. REACTION KINETICS

1.1 Thermal decomposition of O₃ in acid solution

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

Ozone decays in acid solutions in a slow thermal process, which does not seem to involve radical species. The decay rate depends on pH, the concentrations of oxygen and ozone and on the temperature. The mechanism is complex and only the first half-life can be expressed in terms of second-order kinetics. The rate of decomposition is 3-4 times faster in "oxygen-free" solutions than in oxygen-saturated solution, which may suggest that the reaction



known in the gas phase takes place in aqueous media as well. The pH dependence suggests that H⁺ or ClO₄⁻ may also be involved. The decay rates are proportional to the concentrations of neither H⁺/ClO₄⁻ nor oxygen; but the reciprocal of the cube root of the concentrations of ClO₄⁻ or O₂ gives a reasonably good approximation in the pH region 0 to 4 and in the oxygen concentration range 10⁻⁵ - 10⁻³ M.

The temperature coefficient is independent of both the pH and the oxygen concentration in the temperature range 0° to 55°C. The activation energy of the decomposition reaction is very close to the activation energy of the dissociation reaction (1) in the gas phase again indicating that this reaction is involved in the decomposition mechanism. To verify this conclusion, investigations of isotopic exchange between O₃ and O₂ are undertaken.

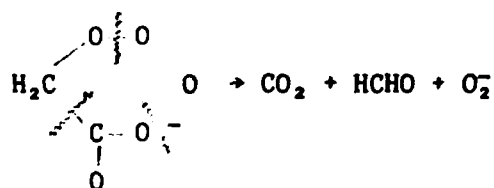
1.2 Pulse radiolysis of acetate-ozone solutions

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

In pulse radiolysis of "oxygen free", N₂O-saturated ozone-acetate solutions, pH = 9-10, the formation of O₂⁻ radical ions is observed as a build-up at the 430 nm band. The O₂⁻ radicals form at the expense of acetate radicals. The kinetics at the 350 nm band (acetate radical) reveals that the formation of O₂⁻ takes place in two kinetically coupled steps.

The decrease in absorption at 260 nm confirms the stoichiometry of two ozone molecules used for every O_3^- formed. At pHs below 7 in buffered solutions the O_3^- radicals are rapidly converted into OH radicals and a chain decomposition of O_3 is observed at 260 nm. Oxygen was found to reduce $G(O_3^-)$ at higher pHs, while a pronounced inhibition of the O_3 chain decomposition by oxygen is demonstrated below pH 7. The influence of oxygen is explained as a scavenging of acetate radicals to form the relatively unreactive peroxy acetate radical.

The chain length of ozone destruction (pH < 7) by 1-2 krd γ -radiation was found to be between 4-25 depending on the dose and the ozone concentration. The products of this chain decomposition were carbon dioxide and formaldehyde. Amounts of CO_2 , measured gaschromatographically and HCHO, measured spectrophotometrically, correspond to the depletion in ozone. Therefore, an addition of ozone to the acetate radical is suggested. The addition product decomposes by an internal electron transfer process into smaller molecules through a six-membered transition state



1.3 The hydrated electron and its reactions at high temperatures*

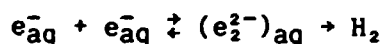
(H. Christensen (Studsvik Energiteknik AB, Nyköping, Sweden) and K. Sehested)

The spectrum of the hydrated electron was determined in the temperature range 5-300°C using strongly alkaline solutions and high hydrogen pressure. At temperatures up to about 150°C the temperature coefficients of E_{\max} and $\Delta E_{1/2}$, are $-2.8 \times 10^{-3} \text{ eV K}^{-1}$ and $2 \times 10^{-3} \text{ eV K}^{-1}$, respectively. E_{\max} is the energy at maximum absorption and $\Delta E_{1/2}$ is the half-width of the spectrum. The temperature coefficient of $\epsilon_{\max}G$ is $50 \pm 10 \text{ dm}^3 \text{ mol}^{-1} \text{ cm}^{-1} \text{ K}^{-1}$ in the temperature range 5-300°C. $\epsilon_{\max}G$ at 21°C is $[1.18 \pm 0.15] \times 10^5 \text{ dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}$ in these solutions. The temperature coefficient of $G(e_{\text{aq}}^-)$ is probably close to zero.

The rate constant of the second-order decay (2k) is $[1.00 \pm 0.05] \times 10^{10} \text{ dm}^3 \text{ mol}^{-1} \text{ s}^{-1}$ at 20°C, independent of pH. The activation energy of the reaction is $23 \pm 1 \text{ kJ mol}^{-1}$ ($5.4 \pm 0.2 \text{ kcal mol}^{-1}$) at temperatures up to 150°C.

The decay at temperatures above 150°C becomes slower with increasing temperatures, but still follows second-order kinetics for 2-3 half-lives. At these high temperatures it is not possible to avoid dissolution of silicon dioxide from the synthetic quartz cell. The silicon dioxide dissolves as silicate, which lowers the pH of the starting solution. As long as the final pH is higher than ~ 10, the decay rate still decreases with temperature, but if the final pH becomes lower than ~ 9 because of dissolved silicate, the decay rate increases. The decay is independent of the concentration of silicate. Also, silicate even at the highest concentrations does not change the electron spectrum, half-width or $\epsilon_{\max}G$ to any significant extent at ambient and higher temperatures.

The simplest mechanism capable of describing the kinetic data at various temperatures is the equilibrium expression



where the dissociation reaction has a higher activation energy than the dimerization reaction. Calculations show that the activation energy of the dissociation reaction is higher than 30 kcal.

1.4 O_2^- reactions at elevated temperatures

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

The purpose of this work is to study the spectrum and reactions of the superoxide radical at temperatures up to 250-300 °C to simulate conditions in nuclear reactors. Spectral changes in solutions at pHs 2 to 10 have been measured at temperatures up to 150°C at hydrogen pressures up to 15 MPa in the presence of O_2 . All radicals are therefore converted to O_2^- . The influence of temperature on the equilibrium $O_2^- + H^+ \rightleftharpoons HO_2$ and the rate constants $O_2^- + HO_2$ and $HO_2 + HO_2$ will be determined. This work is continuing and is financially supported by Studsvik Energiteknik AB, Sweden.

1.5 Activation energy for the reaction $H + OH^- \rightarrow e_{aq}^-$. Kinetic determination of the enthalpy and entropy of solvation of the hydrated electron*

(B. Hickel (CEA - CEN/Saclay, Gif-Sur-Yvette Cedex, France) and K. Sehested)

The reaction between atomic hydrogen and hydroxide ion in aqueous solutions $H + OH^- \rightarrow e_{aq}^- + H_2O$ has been studied by pulse radiolysis. The rate constant was measured at pH 11.7 and 12 by following the growth of the hydrated electron absorption at 600 nm. The activation energy of the reaction has been determined over the temperature range 15-60°C as 6.3 ± 0.6 kcal/mol (26.4 ± 2.5 kJ/mol). From this value and the activation energy of the reverse reaction, the e_{aq}^- enthalpy of formation $\Delta H_f = -32.6 \pm 1.6$ kcal/mol (-136.4 ± 6.7 kJ/mol) and its standard entropy $S^\circ = 16.7 \pm 5.4$ cal/mol deg (69.8 ± 22.5 J/(mol deg)) were calculated. The high entropy of solvation $\Delta S_s = 11.7 \pm 5.4$ cal/(mol deg) (49 ± 22.6 J/(mol deg)) when electrons are transferred from gas phase into aqueous solution indicates that the hydrated electron is a structure breaker.

1.6 Activation energy of the reaction $OH + NH_3$

(B. Hickel (CEA - CEN/Saclay, Gif-Sur-Yvette Cedex, France) and K. Sehested)

The reaction of OH radicals with ammonia was studied at elevated temperatures. As the radical NH_2 formed in this reaction has a very low extinction coefficient and therefore is difficult to observe, we chose a competitive study with $Fe(CN)_6^{4-}$, SCN^- and SCN^-/H_2 . The activation energy of $Fe(CN)_6^{4-} + OH$ is known and that of $SCN^- + OH$ was determined to 3 kcal by competition with hydrogen up to 200°C. The activation energy for $OH + NH_3$ could be determined only up to 100-150°C in these systems, as other reactions such as complexing reactions with NH_3 take place above these temperatures. The activation energy is found to be close to zero, which indicates that the reaction $OH + NH_3 \rightarrow NH_2$ is not a simple hydrogen abstraction reaction, but rather an addition of OH followed by an elimination of water. Further experiments are in progress.

1.7 Selenium (V). A pulse radiolysis study*

(U.K. Klänning (Chemistry Dept., Aarhus University) and K. Sehested)

Two Se(V) species, assumed to be SeO_3^- and $HSeO_3^{2-}$, in equilibrium with each other were observed in pulse radiolysis of aqueous selenite and selenate solutions. The following reactions and equilibria were observed: 1) $SeO_3^- +$

$\text{OH}^- \rightleftharpoons \text{HSeO}_4^{2-}$, $K_1 = 0.78$; 2) $\text{SeO}_3^{2-} + \text{OH}^- \rightleftharpoons \text{HSeO}_4^{2-}$, $k_2 = (3.5 \pm 0.2) \times 10^9$
 $\text{dm}^3 \text{mol}^{-1} \text{s}^{-1}$, $k_{-2} = (7.3 \pm 0.5) \times 10^5 \text{ s}^{-1}$; 3) $\text{O}^- + \text{H}_2\text{O} + \text{SeO}_3^{2-} \rightleftharpoons \text{HSeO}_4^{2-} + \text{OH}^-$,
 $k_3 = (1.1 \pm 0.1) \times 10^7 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$, $k_{-3} = (3 \pm 1) \times 10^5 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 4) $\text{HSeO}_3^- +$
 $\text{OH}^- \rightarrow (\text{H}_2\text{SeO}_4^-) \rightarrow \text{SeO}_3^- + \text{H}_2\text{O}$, $k_4 = (1.6 \pm 0.1) \times 10^8 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 5) $\text{H}_2\text{SeO}_3 + \text{OH}^-$
 $\rightarrow (\text{H}_3\text{SeO}_4) \rightarrow \text{SeO}_3^- + \text{H}_3\text{O}^+$, $k_5 = (1.0 \pm 0.1) \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 6) $2\text{SeO}_3^- \rightarrow \text{Se(IV)}$
 $+ \text{Se(VI)}$, $k_6 = (5.2 \pm 0.5) \times 10^8 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 7) $2\text{HSeO}_4^{2-} \rightarrow \text{SeO}_3^{2-} + \text{SeO}_4^{2-} + \text{H}_2\text{O}$,
 $k_7 \sim 5 \times 10^8 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 8) $\text{HSeO}_4^{2-} + \text{SeO}_3^- \rightarrow \text{SeO}_3^{2-} + \text{SeO}_4^{2-} + \text{OH}^-$, $k_8 \sim 10^9$
 $\text{dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 9) $\text{CO}_3^{2-} + \text{SeO}_3^- \rightarrow \text{CO}_3^- + \text{SeO}_3^{2-}$, $k_9 = (6 \pm 1) \times 10^6 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 10)
 $\text{SeO}_4^{2-} + e_{\text{aq}}^- \xrightarrow{\text{H}^+} \text{HSeO}_4^{2-}$, $k_{10} = 1.1 \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$; 11) $\text{HSeO}_4^- + \text{H}^+ \rightarrow$
 $(\text{H}_2\text{SeO}_4^-) \rightarrow \text{SeO}_3^- + \text{H}_2\text{O}$, $k_{11} \sim 10^6 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$. The standard Gibbs energy of
 formation of SeO_3^- and HSeO_4^{2-} , $\Delta_f G_{\text{aO}}^\circ(\text{SeO}_3^-) = -207 \text{ kJ mol}^{-1}$ and $\Delta_f G_{\text{aO}}^\circ(\text{HSeO}_4^{2-})$
 $= -364 \text{ kJ mol}^{-1}$, and the standard reduction potentials $E_{\text{aO}}^\circ(\text{SeO}_3^-/\text{SeO}_3^{2-}) = 1.68$
 $\pm 0.01 \text{ volt}$ and $E_{\text{aO}}^\circ(\text{HSeO}_4^{2-}/\text{SeO}_3^{2-}) = 1.57 \pm 0.01 \text{ volt}$ were determined from K_1
 and k_2/k_{-2} and the standard Gibbs energy of formation of OH^- , OH^- , H_2O and
 SeO_3^{2-} . The relatively small values for $\Delta_f G_{\text{aO}}^\circ(\text{SeO}_3^-)$ and $\Delta_f G_{\text{aO}}^\circ(\text{HSeO}_4^{2-})$ agree
 with the observation that one-equivalent oxidation of Se(IV) or reduction of
 Se(VI) oxoacids and oxoanions generally are fast and suggest that one-equiva-
 lent oxidations of selenite may take place by electron transfer from selenite
 or by transfer of O^-/OH to selenite.

1.8 Laser flash photolysis of Se^{IV} and Se^{VI} oxoacids and oxoanions at 193 nm
 (U.K. Klänning (Chemistry Dept., Aarhus University) and K. Sehested)

The primary processes in laser flash photolysis of SeO_3^{2-} and HSeO_4^- were studied:



as well as the photolysis of H_2SeO_3 and SeO_4^{2-}



A comparison of the ratios of yields of e_{aq}^- and O^-/OH in the photolysis of analogous sulfur and selenium oxoanions and oxoacids with the corresponding changes in the standard Gibbs energy for these compounds suggests that the

precursors of O⁻/OH production are populated after relaxation rather than directly by the light-absorption process.

1.9 Laser flash photolysis of fluoroxosulfate SO₄F⁻ at 193 nm

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

The observation of the sulfate radical formed at a rate matching the laser pulse suggests a primary process



(similar to the primary process in the photolysis of other hypohalites) by which the oxygen-halogen bond is broken.

No absorption, such as could be assigned to the fluorine atom, was observed in neutral or in 1 mol dm⁻³ HClO₄. However, the kinetics of decay of SO₄^{·-} agree with an immediate formation of OH in the initial concentration equal to [SO₄^{·-}]. These observations indicate that the fluor atom is a strong acid



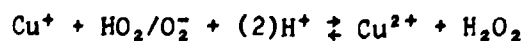
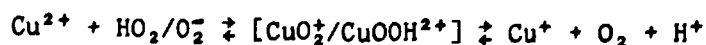
and that FOH⁻ dissociates in less than 10⁻⁷ - 10⁻⁸ s according to



1.10 The reactivity of Cu²⁺-arginine complexes with HO₂/O₂⁻ radicals

(D. Cabelli (Brookhaven National Laboratory, Upton, N.Y., USA) and J. Holcman)

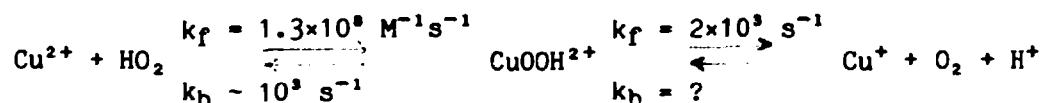
Although Cu²⁺ complexes are known to react with HO₂/O₂⁻ according to the following general mechanism



CuO₂⁺/CuOOH²⁺ has never been observed. Recent theoretical studies suggested

that arginine plays a role in the dismutase function of the superoxide dismutase enzyme (SOD-Cu²⁺) as the arginine has a non-bound amino group that can both stabilize the O₂⁻ radical and donate a proton. A study of the Cu²⁺-arginine complexes with HO₂/O₂⁻, using pulse radiolysis and the pressure cell that allows a very high O₂ concentration, might allow spectroscopic observation of CuO₂⁺/CuOOH²⁺ and elaboration of a Cu²⁺ dismutase mechanism.

The aforementioned mechanism was studied between pH 1.5-10.5 as a function of Cu²⁺ concentration and, at pH 2.0 and 6.0, arginine concentration. Preliminary results indicate that at pH 2.0 HO₂ reacts with Cu²⁺-arginine to form a transient which subsequently disappears at a rate, k ~ 2×10³ s⁻¹. Spectrally, the transient (postulated to be CuOOH²⁺) has a similar absorption profile but lower OD than that of HO₂. The kinetic and mechanistic results thus far are:



At high pH the reaction is much more complex as the rate of disappearance of O₂⁻ changes with pH, suggesting that not all forms of Cu²⁺-arginine complexes react at similar rates. At pH 6.0 the rate also changes with arginine concentration, possibly because the free copper present reacts at a much faster rate than the pertinent Cu²⁺-arginine complexes. Finally, the dismutation rate slows significantly (as much as a factor of 4) at high O₂ concentration. This work is still in progress.

1.11 Pulse radiolysis of aqueous solutions containing methionyl di- and tripeptides. Reactions with OH radicals

(K. Bobrowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

The reactions of hydroxyl radicals with a series of di- and tripeptides containing methionine were investigated in acid and neutral pH range by pulse radiolysis method. Two kinds of absorption bands were formed simultaneously by the OH-attack on methionyl-methionine and methionyl-methionyl-methionine at low concentrations (10⁻⁴). The position of the more intensive band was dependent on the pH. At pH = 1 the absorption spectrum has two maxima: at λ = 290 nm and λ = 480 nm. At pH = 5 absorption maximum at λ = 290 remains, but the second one is blue-shifted to ~ 390 nm. These results suggest the formation of intramolecular radical cation complexes, predominantly at low pH, which are characterized by a three-electron bond between two sulphur atoms of

the same peptide molecule and absorption maximum around 480 nm. With increasing pH another intramolecular process takes over in which the establishment of S...N three-electron bond is involved. An intramolecularly formed S...N three-electron bonded radical cation has been identified as a transient intermediate at higher pH with $\lambda_{\max} = 390$ nm.

In solutions of methionyl-glycine, methionyl-serine, and (Met-X)-type dipeptides at pH = 4.95, transient absorption bands with $\lambda_{\max} = 390$ nm are observed as well. These species decay exclusively by a second-order process. However, different spectra are obtained for the corresponding solutions of glycyl-methionine and seryl-methionine. The 390 nm band is now absent and instead a broad, flat, nonspecific absorption is seen at ca. 360 nm. Part of this absorption is short-lived and decays exponentially with $\tau_{1/2} \sim 4$ μ s. All spectra include also a peak with $\lambda_{\max} = 290$ attributable to α -(alkylthio)-alkyl radicals. These results show an inability of formation of the intramolecular three-electron bond between nitrogen and sulphur atoms (contrary to intramolecular three-electron bond between two sulphur atoms) if these two atoms are separated by peptide bond.

1.12 Pulse radiolysis of aqueous solutions containing methionyl di- and tripeptides. Peptide-halide systems

(K. Bobrowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

The reactions of dibromide radical-anions Br_2^- with a series of model di- and tripeptides containing methionine were preliminary investigated at pH 1 and pH 5.1. The absorption spectra formed during the electron pulse in all solutions has a maximum at $\lambda = 270$ -280 nm and another at $\lambda = 360$, the latter identified as Br_2^- . In methionyl-methionine (pH 1, and pH 5.1), methionyl-methionyl-methionine (pH 5.1), glycyl-methionine (pH 5.1), and seryl-methionine (pH 5.1) the initial absorption decays, leaving a spectrum with a peak at ~ 380 -390 nm. This peak can be attributed to a transient with a three-electron bond between sulphur and bromide. In solutions of methionyl-glycine, and methionyl-serine at pH 5.1, no absorption corresponding to the >S...Br species was observed. Furthermore, an increase in optical absorption occurs in the range 250-300 nm, concomitantly with the disappearance of the 360 nm absorption band. Both phenomena, e.g. the formation of >S...Br type species and the build-up of 270 nm-species concomitant with the decay of Br_2^- were observed in methionyl-methionyl-methionine solutions at pH 1.

Some interesting behaviour was observed during OH radical induced oxidation of methionyl-methionine and methionyl-methionyl-methionine in the presence of iodide ions at pH 1. Contrary to the behaviour established for the methionine system no absorption attributable to a reaction product of I^- with the sulphur-centered radical cations is observed. Furthermore, the decay of the 480 nm absorption band parallels the formation of the 270 nm absorption band. Further experiments are needed for a better understanding of phenomena occurring in the methionyl peptides-halide systems.

1.13 Pulse radiolysis of aqueous solutions containing methionyl di- and tripeptides. Tl^{2+} -induced oxidation

(K. Bobrowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

Preliminary experiments concerning Tl^{2+} -induced oxidation of methionyl-methionine and methionyl-methionyl-methionine at low concentrations (3×10^{-5} M, 10^{-4} M) have shown the formation of a 480 nm absorption band in both acid (pH = 1) and slightly acid (pH = 4.1) aqueous solutions. These data indicate the formation of intramolecular sulphur-centered radical cation complexes. Further experiments are in progress.

1.14 The reaction of Pseudomonas Azurin with N_3^{\cdot} and $Br_2^{\cdot-}$ radicals

(K. Bobrowski (Institute of Biochemistry and Biophysics, Polish Academy of Sciences, Warsaw, Poland) and J. Holcman)

The reactions of Pseudomonas Azurin (the electron mediating cupric blue protein involved in the bacterial cytochrome chain) with azide radicals N_3^{\cdot} and dibromide radical anions, $Br_2^{\cdot-}$ have been undertaken using the pulse radiolysis method. Preliminary experiments have shown that N_3^{\cdot} radicals selectively attack the tryptophan residue (in spite of its deep burial in the azurin molecule). However, coincident with the $Br_2^{\cdot-}$ radical attack, the formation of a 270 nm absorption band is observed. The similarity of this band to the 270 nm band in some model methionyl peptides suggests methionine as the attacked residue site in the azurin molecule.

1.15 Pulse radiolysis of bromide and chloride in aqueous solutions containing cyanide*

(Z. Draganić (Belgrade, Yugoslavia), K. Sehested, I. Draganić (Belgrade, Yugoslavia), and E. Bjergbakke)

Mixtures of cyanide and bromide or chloride in aqueous solutions (N_2O -saturated, pHs 3.1 and 10.3) were examined by fast kinetic spectrophotometry with a 3 krd pulse from the 10 MeV electron accelerator. A crossing of reaction pathways of cyanide and halide was observed and attributed to reactions of the OH-adduct of cyanide with Br_2^- and Cl_2^- . A computer fitting of the experimental results was used to calculate the rate constants of the reactions. Some complementary experiments were made with pure halide solutions and the molar extinction coefficients of Br_2^- and Cl_2^- were derived by computer simulation: $\epsilon_{Br_2^-} = 22,000 \pm 1,000 M^{-1}cm^{-1}$ at $\lambda = 270$ nm, and $\epsilon_{Cl_2^-} = 20,500 \pm 1000 M^{-1}cm^{-1}$ at $\lambda = 230$ nm. The competition of bromide and molecular hydrogen for OH radicals was followed at elevated hydrogen pressures in the optical pressure cell and the rate constant for $OH + Br^-$ was determined at pHs 3.1 and 10.3.

1.16 Radiolytic products in various waters*

(E. Bjergbakke, Z. Draganić (Belgrade, Yugoslavia), K. Sehested, and I. Draganić (Belgrade, Yugoslavia))

A survey of results obtained by computer simulations of some processes in laboratory and in nature is prepared.

The chemical action of ionizing radiation on water is of considerable interest because of the widespread use of water as a solvent in radiochemistry and nuclear technology, its use as a coolant and moderator in nuclear reactors, and its presence at radioactive waste disposal sites in deep underground or at the bottom of the ocean.

Radiolytic products and their dependence on the irradiation conditions are, nevertheless, quite often unknown or neglected by many of those dealing with radioactivity and radiation.

We present a survey of results obtained by computer simulations of some processes under laboratory conditions and in nature. The existence of a well-established model of the radiolysis of pure water combined with abundant data

on the rate constants and intermediates of many reactions, enables us to effectively model processes in aqueous solutions with a computer. To illustrate the effect of absorbed energy (the dose), energy deposition rate (the dose-rate) and the quality of radiation (its type and energy), we present the formation of H_2 , H_2O_2 and O_2 under different conditions. Dilute aqueous solutions were used to examine some cases of more general interest to laboratory practice. Data on the formation of radiolytic products are presented for various common solutes such as oxygen, hydrogen, carbon dioxide, metal ions, and representative, simple, organic compounds such as methane and ethanol. In the case of natural water, the results of the computer simulations are given for fresh water, underground waters (bed rock and salt brine) and sea water. The formation of radiolytic products in a boiling water nuclear reactor (BWR) is presented in some detail. We also show some results relevant to the product formation in waters of Precambrian Earth, e.g. in natural nuclear reactors like those discovered in Oklo, and in primordial oceans due to the energy deposition from the radioactive decay of ^{40}K .

1.17 Computer simulations of the reactions in the Fricke dosimeter with varying Cl^- concentrations

(E. Bjergbakke)

This work is performed in collaboration with J. Swallow, Christie Hospital and Holt Radium Institute, Manchester, and B. Parsons, Kelterston College, North East Wales Institute.

The experimental work has been completed and a report is being written. A good agreement between experimental data and computed values has been obtained.

1.18 Contract work for Studsvik Energiteknik AB

(H. Christensen (Studsvik Energiteknik AB, Nyköping, Sweden), E. Bjergbakke, and O. Lang Rasmussen (Computer Installation))

The work consists of simulation of radiation chemical reactions and mechanisms associated with radioactive waste disposal and reactor chemistry.

1.19 References

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2. RADIATION PHYSICS, PHYSICAL DOSIMETRY, AND TECHNOLOGICAL APPLICATIONS OF RADIATION

2.1 Track structure theory

(J.W. Hansen, K.J. Olsen (Dept. of Radiophysics, University Hospital of Copenhagen, Herlev) and M. Waligorski (Institute of Nuclear Physics, Krakow, Poland))

The applicability of the track structure theory has been tested by comparing predictions based on the theory with experimental high-LET dose-response data for an amino acid alanine and a nylon based radiochromic dye film radiation detector. The linear energy transfer, LET, has been varied from 28 to 20,200 MeVcm²/g using ion beams ranging from protons to sulphur ions. The low-LET reference radiations were beams of fast electrons and ⁶⁰Co γ-rays. At doses well below saturation the two detectors act upon low-LET radiation in close accordance with the theoretical considerations, but at marginal high doses dose-rate effects are observed obscuring the evaluation of important param-

ters. Radical-radical interaction, diffusion of radicals, and energy migration in large molecules and along crystal lattices, all depend strongly on the density and time scale of deposited energy. Thus, the model that attempts the prediction of bulk effects from ionizing radiation must include functions describing the circumstances under which the energy is deposited and how this energy deposition influences the final chemical products. The work is in progress.

The comparison of calculated data with experimental data from irradiation of alanine with cyclotron-produced fast neutrons at the Institute of Nuclear Physics, Krakow, has not yet been completed. Because of differences between the Risø REINT computer program and the CHIT.MRC program developed at the University of Lincoln, USA, for neutron irradiations an improvement is being made of the CHIT.MRC program in respect to the dose deposition in the track of heavy particles. This work is performed during Dr. Waligorski's stay with Prof. R. Katz (Lincoln, Nebraska, USA).

2.2 Investigation of the alanine dose meter

(J.W. Hansen and K.J. Olsen (Dept. of Radiophysics, University Hospital of Copenhagen, Herlev))

Manufacturing of alanine pills has demanded the development of special equipment added to the tablet machine for delivering an accurate dosage of alanine powder into the die in order to produce mechanically stable pills. Further, additional equipment was designed for transporting the pills away from the die area automatically, for adequately selecting the mechanically stable pills, and for cleaning the pills of dust before storage. For testing the mechanical stability a special gauge was constructed which in terms of force per unit area indicates the pressure for rupture of the pill.

A three-year contract proposal to the European Communities, Biology, Radiation Detection, and Medical Research, has been accepted to start January 1, 1986. The contract concerns development of alanine as a personal accidental dose meter. The dose meter should be investigated for response to low- and high-LET radiation with emphasis put on charged particles simulating fast neutron spectra in tissue. Special emphasis will be put on detection of doses below 1 Gy.

Our alanine dose meter has been exposed to 740 MeV ^{40}Ar -particles at Gesellschaft für Schwerionenforschung (GSF) in Darmstadt, W. Germany. Measured effectiveness relative to response from ^{60}Co γ -ray photons conforms with calculated effectiveness within 4% which is within the experimental uncertainty. The idea behind the irradiation of alanine at GSF was to measure the response to saturation doses after exposure to a particle with a high average LET in the sample. The data should be used in the investigation of the relationship between the change in saturation response with the dose rate in the particle's track. We did not get the saturation doses because the exposure time was too long.

2.3 Dosimetry

(A. Miller)

Dosimetry for application in radiation processing is carried out in the fields of reference dosimetry, dose validation measurements, development of dosimeters and testing of dosimeter properties.

During 1985 a dose intercomparison with National Physical Laboratory (NPL), UK, was carried out. Rise film and alanine dosimeters were irradiated with Cobalt-60 γ -rays at NPL, and the readings carried out here showed an agreement of better than 2% for 5-50 kGy and 5% at 2 kGy for alanine and 2-5% over the range 2-50 kGy for the film dosimeters.

An international dose assurance service (IDAS) was established in 1985 by the IAEA, and by using dosimeters from this service the dose of the Rise 10 kCi cobalt facility was checked. The agreement was better than 2%.

We have taken an active part in the establishment of IDAS cobalt facilities, and we also take part as consultant to the IAEA for continuing this program with the aim of establishing a similar service for industrial electron accelerator facilities. To provide a needed common reference for industrial electron beam dosimetry, dose measurements were carried out at the NPL 10 MeV linac using graphite calorimeters from Rise, National Bureau of Standard (NBS), Washington and NPL. The Rise calorimeters showed agreement with NPL readings within 3%. Various dosimeters from Gesellschaft für Strahlen- und Umweltforschung (GSF), München, NBS, NPL and Rise have later been irradiated at NPL in order to identify possible transfer dosimeters. These readings are not finally evaluated. Similar irradiations should have been carried out at

the Risø linac, but the current klystron problems have so far prevented that. This work is supported by IAEA Research Contract No. 3583, and a collaboration on this work is also maintained with Dr. A. Kovacs, Institute of Isotopes, Budapest.

The graphite calorimeters mentioned above have been tested for their applicability as a reference at the 400-keV accelerator. Several readings, however, show lack of coherence between the expected dose values (calculated) and the values measured with a calorimeter. At the same time there is a reasonable agreement between film dosimeter readings and calculations. These measurements, which were carried out in collaboration with Dr. Igor Janovský, a visiting scientist from Nuclear Research Institute, Rez, Czechoslovakia, will be continued in 1986.

Calibration irradiation of dosimeters to reference doses both at the 10 MeV electron accelerator and the cobalt-60 facilities were in several cases carried out for customers in Denmark and abroad, and measurement of dose distributions in products irradiated both at Risø and at other irradiation facilities were carried out in order to demonstrate the compliance of the irradiation process with national regulations and codes of practice. Film and alanine dosimeters were also sent to customers' irradiation facilities in order to verify administered doses. 13 customers have used these services in 1985.

The development of film dosimeters has continued, and industrial production of test batches has taken place in amounts of 250 m length, 40 cm width. The properties of the dosimeter film are being tested and contacts have been made with a firm which has started development work on a reader for this dosimeter.

The same film dosimeter has also been used to make a prototype dosimeter for application at CERN, Geneva. The dosimeter will here be used to measure the radiation level at accelerator components, and is being developed under a contract with CERN.

Lyoluminescence of glutamine has been investigated as a routine dosimeter in the range from 10 Gy to 100 kGy. The response to Cobalt-60 gamma and 10 MeV electron irradiation has been found to be equal within the precision of the dosimeter ($\pm 5\%$) and relative humidity was also found not to influence the response. (This work was carried out in collaboration with Mrs. Xie Liqing,

Beijing, China).

We participate in a working group under ICRU in order to produce a set of guidelines for "Dosimetry for Radiation Processing". This report is near completion and a final meeting of the working group was arranged at Risø in October 1985.

2.4 Radiation processing

(A. Miller)

Irradiation of commercial products on the linear accelerator has continued through 1985. These irradiations and irradiations for other Risø departments and various institutions are normally carried out on 2 days a week, but because of problems with the klystron, more time has been needed to fulfil our obligations. "Various institutions" include Sct. Hans Hospital, Roskilde, where a research project uses radiation to determine the size of brain receptors. This research has so far been presented in 15 publications.

The commercial irradiations include radiation sterilization of medical and laboratory devices, crosslinking of polymers, modification of semiconductor parameters and various test irradiations of, e.g. foods. In December 1985 Risø got clearance for irradiation of spices, but this has not yet begun commercially. An information meeting for the Danish food industry, regulatory and other institutions was arranged in 1985 with approx. 100 participants. 43 customers made use of the irradiation service in 1985 including some irradiations at the 400 keV accelerator and at the cobalt facilities.

A close collaboration with the Polymer Group (Chemistry Dept.) is maintained, and we participate in various development projects with this group and its commercial partners. We participated with the Polymer Group in the Scandinavian Rubber Conference in Copenhagen with an exhibition booth.

2.5 References

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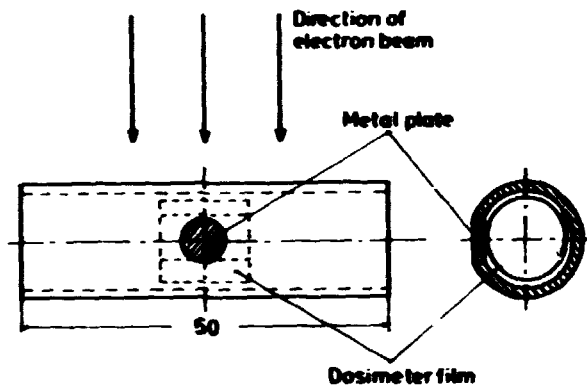
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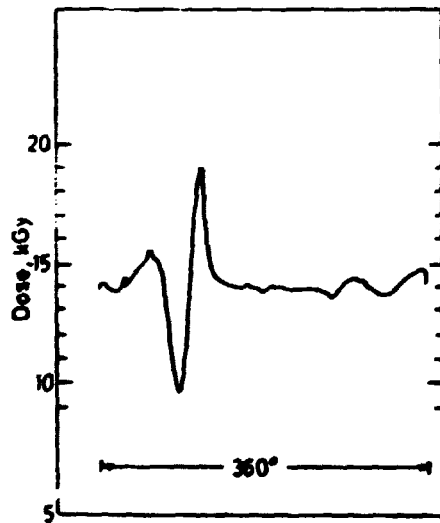
Figure 1.

A

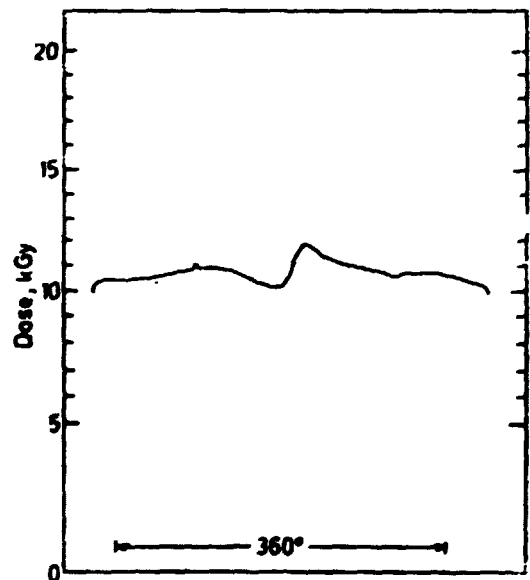


Example of dose distributions measured in 15 mm^ø plastic tubes irradiated with unidirectional 10 MeV electrons A). The surprisingly large dose gradients are due to a small metal plate at the side of the tube. The measurement in B) was made with the tube located in the product box close to the entrance side of the electron beam, while in C) it was located further away from the beam. The effect of the more scattered radiation is to diminish the dose gradients.

B



C



3. OPERATION AND MAINTENANCE OF THE IRRADIATION FACILITIES

3.1 HRC electron linear accelerator

(J. Fenzer and B. Lynggård)

During the year the accelerator operation has been influenced by abnormal situations. Time-consuming repair and replacement of vital components were carried out in the first third of the year. The operation in the last third of the year was restricted due to a defective klystron. The defective klystron, with gradually reduced power, has caused the accelerator to be in operation for about a 15% longer time than usual. The distributed use of the accelerator for the last five months has been 60% for service irradiation on the conveyor and 30% for experimental irradiation in connection with the pulse radiolysis equipment. Maintenance of the facility covered 10% of the time.

The experience of the latest year of operation shows that it is necessary to secure a continuous operation of the ten-year old accelerator with more costly spare parts especially in regard to the commercial irradiations. Within the last year the following maintenance works were carried out:

Injector system. The cathode in the electron gun was replaced. The old cathode has been in operation for 15,000 hours.

Vacuum system. On several occasions a defective vacuum valve in the injector system has caused the gun cathode to be exposed to the atmosphere. Reprocessing of the gun cathode may last up to two weeks. To avoid these problems the organic sealed valve was replaced with an all-metal sealed valve with an expected lifetime of up to 10,000 cycles before servicing.

The aluminium window for the 90° bent beam is being replaced once a year in order to avoid vacuum leakage or implosion of the window due to metal fatigue.

A Faraday cup was installed in connection with the ± 50 keV resolution energy analyzing slit, both designed in the Department. The Faraday cup and slit are integrated in the accelerator vacuum system and together with the switching magnet SM-1 serve as a spectrometer which has been calibrated in terms of energy. An output window port, designed in the Department, was mounted on the

switching magnet SM-2 in the target room.

The fast closing vacuum valve has been overhauled; the organic seal has to be replaced from time to time due to x-ray degradation.

A secondary vacuum gauge was installed in the injector system.

Heat from a collimator in the injector system caused an oil seal into the gun tank to leak. Temporarily the problem was solved by air cooling of the seal. Steps will be taken to replace the collimator with a water-cooled one.

Microwave system. In August after 12,500 hours of operation the klystron S/N 1023 was replaced due to decreased microwave power. Unfortunately, the spare klystron, which previously had been in operation for 2000 hours, malfunctioned after only 100 hours of operation. The klystron S/N 1023 was reinstalled, but operated with approximately half the rated output level. Immediately a new klystron was ordered from ITT in USA, but the export license from the US Ministry of Commerce was delayed and the klystron was not received in 1985.

Due to the very low microwave power level it has been necessary to increase the daily operation hours by 15% in order to cover commercial service irradiations.

Two directional couplers in the microwave system have malfunctioned. One of them, in connection to the klystron, was temporarily repaired but will be replaced when the new klystron is installed.

The other directional coupler, at the input to the accelerator wave guide, developed a vacuum leak for the second time. It was impossible to tighten the leak, and a spare adapter was temporarily installed. Steps will be taken to repair the coupler.

3.2 Febetron, field-emission accelerator

The field-emission accelerator was used for pulse radiolysis of gases. A defective top module caused a malfunction and was replaced by a new spare module. Also the beam tube was replaced after 5 years of operation.

3.3 ICT, low-energy accelerator

The low-energy accelerator was used for dosimetry and polymerization experiments. The water-cooled beam stopper was replaced due to corrosion.

3.4 10,000 Ci ^{60}Co -facility

The 10,000 Ci ^{60}Co -facility was used for radiation research and for customer services. The operation was trouble-free.

3.5 5,000 Ci ^{60}Co -facility

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

3.6 3,000 Ci ^{60}Co -facility

The 3,000 Ci ^{60}Co -cell was used for research and test irradiations in radiation chemistry, radiation dosimetry, and customer services. The operation was trouble-free.

3.7 Lambda Physik Excimer laser

The laser has been used for fast ESR experiments at the Institute of Chemistry, Århus University, and for Raman spectroscopy and flash photolysis in the Accelerator Department. The operation and maintenance of the laser has been time consuming mainly due to transportation and reinstallation at the three different locations. The halogen-gas valves and the valves in the gas mixing system has caused troubles and some valves were repaired or replaced. The thyatron has at several occasions malfunctioned, but the problem was solved by careful frequent rinsing and/or replacement of the air filter in the laser.

3.8 Computer for flash-photolysis

An old PDP-11/10 computer with disk and magnetic tape units (earlier used in the Metallurgy Department) was installed in 1984 to be used for the flash-photolysis set-up. However, the computer never became operational due to malfunctioning of the disk unit, and it was discarded by the Electronic Department. A new PDP-11/23 (founded by the Danish Natural Sciences Research Council) was purchased for the Raman spectroscopy experiments, and it will be used for the flash-photolysis experiments as well.

3.9 Pulse radiolysis set-up

(J. Fenger and B. Lynggård)

Measuring of weak transient absorption signals assumes that a light detector with high signal/noise ratio is available. Two multiplier configurations were designed with signal/noise ratio better than 1000, covering the range from 330 - 700 nm. One of the photomultipliers was designed by an IAEA fellow, Rafal Kubis, Institute of Applied Chemistry, Lodz, Poland.

The disk and tape units on the 15-year old PDP8 computer have malfunctioned, and programs can be read-in only from paper tape. As the staff of the Electronic Department no longer has experience in repairing these old computers, it will be repaired only in the event of an emergency requiring its use. Steps will be taken to have it replaced with a new computer system.

4. EDUCATIONAL ACTIVITIES AND PUBLICATIONS

4.1 Lectures

K. Bobrowsky, Three-electron-bonded Radical Cations Formation in Pulse Radiolysis of Methionine Di- and Tripeptides. Risø, August.

H. Christensen and E. Bjergbakke, Alpha-radiolysis of Aqueous Solutions. Ninth International Symposium on the Scientific Basis for Nuclear Waste Management, 9-11 September, Stockholm. Materials Research Society, Swedish Nu-

clear Fuel and Waste Management Co (SKB).

J.W. Hansen and K.J. Olsen, High-LET Radiation and High-level Dosimetry. 19th Annual Meeting on the European Society of Radiation Biology, Prague, August.

E.J. Hart, K. Sehested, E. Bjergbakke and J. Holcman, Y-ray Initiated Chain Decomposition of Aqueous Ozone Solutions. Radiation Research Society, Los Angeles, May.

A. Miller, Orienteringsmøde om bestråling af levnedsmidler. Risø, March.

A. Miller, "Bestråling af Levnedsmidler" Rotary, Roskilde, February.

K.J. Olsen and J.W. Hansen, Experimental Data from Irradiation of Physical Detectors Disclose Weaknesses in Basic Parameters in the Delta-ray Theory of Track Structure. 9th Symposium on Microdosimetry, Toulouse, May.

K.J. Olsen and J.W. Hansen, Alanine, a Tissue-equivalent Free Radical Dosimeter for Low and High LET Radiation. Medical Physics and Biomedical Engineering Seminar, University of Wisconsin-Madison, Madison, WI, September.

K. Sehested, Behaviour of Hydrated Electrons at Temperatures up to 300°C. Institut für Experimentalphysik und Institut für Strahlenchemie der Universität Wien, May.

K. Sehested, J. Holcman, E. Bjergbakke and E.J. Hart, The Use of Pulse Radiolysis for the Study of the Chemistry of Aqueous Ozone and Ozonide Solutions. Symposium PULS 85, Łodz, September.

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4.2 Publications

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H. Christensen and E. Bjergbakke, Radiolys i 0-1 i Samband med Vätedosering. Studsvik Report NW-85/978.

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J.W. Hansen, Matematisk-Fysiske Modeller til Bestemmelse af Tunge Ladede Partiklers Relative Effektivitet. Risø-I-201.

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J.W. Hansen, Basic Concepts of Radiation Physics and Dosimetric Units. In Techniques of Radiation Dosimetry, Ch. 2, eds. K. Mahesh, D.R. Vis. Published by Mohinder Singh Sejwal for Wiley Eastern Limited, New Delhi, India.

B. Hickel and K. Sehested, Poster: Kinetic Determination of Some Thermodynamic Properties of e_{aq}^- . Miller Conference, Windemere, Scotland, April.

B. Hickel and K. Sehested, Activation Energy for the Reaction $H + OH^- \rightarrow e_{aq}^-$. Kinetic Determination of the Enthalpy and Entropy of Solvation of the Hydrated Electron. J. Phys. Chem. 89, 5271.

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S. Solar, W. Solar, N. Getoff, J. Holcman and K. Sehested, Reactivity of OH and O⁻ with Aqueous Methyl Viologen Studied by Pulse Radiolysis. J. Chem. Soc., Faraday Trans. I, 81, 1101-1112.

5. 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	0.01 - 4 μ s
Pulse repetition rates	single pulses and 12.5, 25, 37.5, 50, 100, 150, and 200 pps.

Accelerator room beam facilities:

1. A bent electron beam with scan width of 60 cm in connection with a conveyor system provides a capacity of irradiating e.g. 400 kg at 35 kGy per hour.
2. Two horizontal beam ports, full average beam power, for electron and X-ray irradiation.
3. One horizontal beam port, reduced average beam power (12.5 pps) in connection with an energy-analyzing spectrometer.

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 MeV
Pulse length	50 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 conveyor to permit pilot-plant irradiation.

4. Excimer Laser, Lambda Physik Model EMG102 E¹

Specifications:

Active medium	<u>F₂</u>	<u>ArF</u>	<u>KrCl</u>	<u>KrF</u>	<u>XeCl</u>	<u>N₂</u>	<u>XeF</u>
Wavelength	157	193	222	249	308	337	351 nm
Pulse Energy (mJ)	10	200	30	250	150	7	100 mJ
Pulse width (FWHM)		14	9	16	10	6	14 ns
Single pulse and repetition rates up to	80	80	100	100	100	100	50 Hz

⁶⁰Co-Facilities

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

Designed for very homogeneous irradiation of samples with a maximum diameter of 180, 100, or 60 mm. The corresponding maximum dose rates (4,860 Ci (1.8×10¹⁴ Bq), 1 January 1986) are 3.0×10⁵, 8.3×10⁵, and 1.9×10⁶ rad·h⁻¹ (0.87, 2.3, 5.2 Gy·s⁻¹), respectively.

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

Designed for laboratory use and fitted with a 123 mm^Ø×150 mm irradiation chamber. The dose rate in the center of the chamber (2,950 Ci (1.1×10¹⁴ Bq), 1 January 1986) is 2.5×10⁵ rad·h⁻¹ (0.70 Gy·s⁻¹). The cell is located at the Control Department, Statens Seruminstitut, Copenhagen.

¹An appropriation by the Danish Natural Science Research Council, shared with Aarhus University.

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

Designed for laboratory use and fitted with a 120 mm^ø×220 mm irradiation chamber. The dose rate in the center of the chamber (1,800 Ci (6.7×10¹³ Bq), 1 January 1986) is 1.8×10⁵ rad·h⁻¹ (0.50 Gy·s⁻¹).

6. VISITING SCIENTISTS

- K. Bobrowski, Institute of Biochemistry and Biophysics, Warsaw, Poland.
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- I. and Z. Draganić, Beograd, Yugoslavia.
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- E.J. Hart, Port Angeles, WA., USA.
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<p>Abstract</p> <p>A description is given of research in the fields of reaction kinetics, physical dosimetry, radiation physics and technological application of radiation and irradiation technology, 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>