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Accelerator Department. Annual Progress Report 1 January - 31 December 1986

Forskningscenter Risø, Roskilde

Publication date: 1987

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

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Accelerator Department Annual Progress Report

1 January - 31 December 1986

Risø National Laboratory, DK-4000 Roskilde, Denmark February 1987 RISØ-M-2633

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ACCELERATOR DEPARTMENT Annual Progress Report 1 January - 31 December 1986

<u>Abstract</u>. 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.

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February, 1987 Risø National Laboratory, DK 4000 Roskilde, Denmark

CONTENTS

PR EF	ACE		5
1.	REACTIO	N KINETICS	6
	1.1	Decomposition of ozone in aqueous acidic solution	6
	1.2	Oxygen isotope exchange in the decay of ozone in	
		acid solution	6
	1.3	Ozone decomposition in aqueous acetate solutions	7
	1.4	The importance of radical-radical reactions in pulse	
		radiolysis of aqueous carbonate/bicarbonate	8
	1.5	The reactions of HO_2 and O_2^- at elevated teperatures	8
	1.6	Radiolysis of aqueous ClO ₂ solutions	9
	1.7	Oxygen isotope exchange between OH and H_2O	9
	1.8	Y-radiolysis of aqueous $Br(0^{10})_3^-$	10
	1.9	Pulse radiolysis of aqueous solutions of halides in the	
		presence of hydrocyanic acid or potassium cyanide	10
	1.10	The interaction between copper(II)-arginine complexes and	
		HO_2/O_2 radicals. A pulse radiolysis study	11
	1.11	Superoxide dismutase	11
	1.12	Intramolecular tree-electron bonds formation in methionyl	
		homopeptides /di-L-Met, /tri-L-Met	12
	1.13	Pulse radiolysis of aqueous solutions of methionine	
		containing oligopeptides	12
	1.14	Intramolecular charge transfer between tryptophane and	
		tyrosine in Trp-(Pro)n Tyr Peptides	13
	1.15	Pulse radiolysis of vitamins (nicotinic acid)	13
	1.16	A quantitative description of the action of high-dose pulses	
		of radiation on aerated acid solutions containing ferrous	
		and chloride ions	14
	1.17	Contract work for Studsvik Energiteknik AB	14
	1.18	References	14

1

1 1 1

2/3

.

Page

Page

2.	RADIATION PHYSICS, DOSIMETRY, AND RADIATION PROCESSING				
	2.1	Particle track structure	15		
	2.2	Investigation of the alanine dosimeter	17		
	2.3	A polymer-alanine film for local dose measurements	18		
	2.4	Radiation processing	18		
	2.5	Dosimetry for radiation processing	19		
	2.6	References	22		

.

3.	OPERATION	AND MAINTENANCE OF THE IRRADIATION FACILITIES	23
	3.1	HRC electron linear accelerator	23
	3.2	Febetron, field-emission accelerator	25
	3.3	ICT, low-energy accelerator	25
	3.4	The *°Co-facilities	26
	3.5	Lambda Physik Excimer laser	27
	3.6	Flash-photolysis set-up	27
	3.7	Pulse radiolysis set-up	27

4.	EDUCATION	AL ACTIVITIES AND PUBLICATIONS	27
	4.1	Lectures	27
	4.2	Publications	29

5.	IRRADIATION FACILITIES AT THE ACCELERATOR DEPARTMENT	32
6.	VISITING SCIENTISTS	34
7.	STAFF OF THE ACCELERATOR DEPARTMENT	35

I I I

1 Figure.

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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 experimen al facilities.

A major research effort is directed towards understanding basic chemical processes better, in particular 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 1986.

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The contributions marked * are abstracts of published papers.

1. REACTION KINETICS

1.1 Decomposition of ozone in aqueous acidic solution

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

The decomposition of ozone in acid solution is a slow thermal process, which is dependent on pH, concentration of ozone and oxygen, and the temperature. Experiments have shown that the anions $(ClO_{+}^{-} \text{ or } SO_{+}^{2-})$ at concentrations above 10^{-2} M take part in the decomposition process. At pH 3 an increase in perchlorate ions enhances the decomposition rate, which indicates a reaction with the oxygen atom from the dissociation of ozone or with ozone itself. The decomposition rate decreases with pH even in the range 2-0 which means that the proton takes part in the decomposition as well. The decomposition rate at very low ozone concentrations ($< 5 \times 10^{-5}$ M) approaches a limiting value, which is independent of pH and the range 1-4. Whether this value is the rate of ozone dissociation or the rate of reaction with water is not yet known. The limiting rate has a very high activation energy and yields a pure first-order reaction.

Experiments with ozone solutions in heavy water were performed in order to study the influence of a possible excited oxygen molecule $(O_2({}^1\Delta_g))$, as this oxygen molecule can be produced in the decomposition of ozone. It has a twenty times longer lifetime in deuterated water than in light water. The results showed no detectable differences in the decomposition rate between heavy and light water indicating that $O_2({}^1\Delta_g)$ plays no role in the process.

1.2 Oxygen isotope exchange in the decay of ozone in acid solution (K. Sehested, J. Holcman, E.J. Hart (Port Angeles, WA, USA) and Ch.-H. Fischer (Hahn-Meitner-Institut, W. Germany))

An oxygen isotope exchange study of mixtures of 1^{3} , 1^{6} ,

- 6 -

change is enhanced by the temperature which also increases the decomposition rate of ozone. In all experiments a loss in ${}^{36}O_2$ indicates an O_3 -mediated exchange of oxygen with water during the ozone decomposition which may mean that one of the pathways of ozone decomposition goes through reaction with water (probably through hydrogen peroxide).

Decomposition of aqueous ozone by light (UV or visible) enhances the exchange rate by a factor of 5-10 which is in accordance with the known light dissociation reaction of ozone

$$0_{3} \stackrel{hv}{+} 0_{2} + 0$$

From this observation we may conclude that the dissociation of ozone in aqueous acidic solution contributes to t⁺ thermal destruction of ozone to only a minor extent.

1.3 Ozone decomposition in aqueous acetate solutions^{*}
(K. Sehested, J. Holcman, E. Bjergbakke and E.J. Hart (Port Angeles, WA,
USA))

The acetate radical ion reacts with ozone with a rate constant of $k = (1.5 \pm 0.5) \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$. The products from this reaction are CO₂, HCHO, and O₂. By subsequent reaction of the peroxy radical with ozone the acetate radical ion is regenerated through the CH radical. A chain decomposition of ozone takes place. It terminates when the acetate radical ion reacts with oxygen forming the unreactive peroxy acetate radical. The chain is rather short as oxygen is developed, as a result of the ozone consumption. The inhibiting effect of acetate on the ozone decay is rationalized by OH scavenging by acetate and successive reaction of the acetate radical ion with oxygen. Some products from the bimolecular disappearance of the peroxy acetate radicals, however, react further with ozone, reducing the effectiveness of the stabilization.

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

Due to relatively slow rates of reaction between OH radicals and carbonate and bicarbonate ions $(k(OH + HCO_3^-) = 8 \times 10^6 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1} \text{ and } k(OH + CO_3^-) = 4 \times 10^6 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1})$, radical-radical reactions influence observed yields and kinetics of formation of the CO_3^- radicals. In the present work the rate constant of the reaction, OH + CO_3^-, k = $3.5 \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$ has been determined by computer modelling of experimentally observed CO_3^- yields. This reaction appears to be more important than the previously recognized reactions: OH + OH, OH + O_2^- or CO_3^- + CO_3^-.

Computer modelling, using a complete set of reactions, has been employed to quantitatively evaluate the effect of radical-radical reactions on results obtained with pulse radiolysis of the bicarbonate/carbonate system. The impact of this effect upon previously published data will be discussed.

1.5 The reactions of HO₂ and O_2 at elevated temperatures^{*}

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

The spectra of the HO_2 and O_2^- radicals were determined in the temperature range 20-300°C in aqueous solutions using 0.2 MPa oxygen and about 10 MPa hydrogen pressure. In this temperature range the wavelengths at maximum absorption, λ_{max} and the half-widths of the spectra change very little with temperature. The temperature coefficient of $\epsilon_{max}G$ is 14 and 32 dm³mol⁻¹K⁻¹ for HO2 and O2, respectively. The corresponding relative temperature coefficients are 0.15 and 0.25% K⁻¹. For HO₂ λ_{max} = 223 nm and ε_{max} = 1 490 dm³mol⁻¹cm⁻¹ at 20°C. For $O_2 \lambda_{max} = 243$ nm and $\varepsilon_{max} = 2150$ dm³mol⁻¹cm⁻¹ at 20°C. The pK_A of HO₂ was determined in the temperature range 20-285°C. In the range 20-100°C pK_A changes little, but above 100°C pK_A increases with an increasing rate up to a value of 6.15 at 285°C. The value at ambient temperature is 4.8. In acid solutions, pH $\langle 2$, the HO₂ radical decays in a bimolecular reaction with a rate constant of 8.4×10^5 dm³mol⁻¹s⁻¹ at 20°C. The activation energy is 20.6 kJ mol⁻¹ (4.9 kcal mol⁻¹). At higher pH the decay is caused at lower temperatures (≤ 100 °C) by the reaction between 10_2 and 0_2° radicals with a rate constant of 9.6×10^7 dm³mol⁻¹s⁻¹ and an activation energy of 7.6 kJ mol⁻¹ (1.8 kcal mol⁻¹).

At higher temperatures ($\geq 200^{\circ}$ C) the decay may be explained by the equilibrium $0_{2}^{-} + 0_{2}^{-} \stackrel{?}{\rightarrow} 0_{4}^{2^{-}}$ followed by the reaction H⁺ + $0_{4}^{2^{-}} \rightarrow HO_{2}^{-} + 0_{2}$ with an activation energy higher than 55 kJ mol⁻¹ (13 kcal mol⁻¹) for the reaction $0_{2}^{-} + 0_{2}^{-} \rightarrow 0_{4}^{2^{-}}$.

1.6 Radiolysis of aqueous ClO₂ solutions

(U. Kläning (Chemistry Dept., Aarhus University), K. Sehested and E. Bjergbakke)

As a continuation of the pulse radiolysis experiments on aqueous ClO_2 solutions, N₂O-saturated solutions at 7 < pH < 13 containing $[ClO_2] < 10^{-3}$ mol dm⁻³ were Y-irradiated with doses between 50-300 Gy (5-30 krd). Analysis of O₂ (gas chromatographic, Van Slyke), ClO_2^- , ClO_2^- , and ClO_3^- (iodometric and by Fe²⁺) formed by the irradiation, shows that ClO_2 reacts with OH and O⁻ in a branching reaction

 $C10_2 + 0^-$, (OH) $C10_3^- + 0_2^ C10_3^-$

with a branching ratio G_1/G_2 which for 0⁻ equals 0.20 and for OH equals 0.55.

1.7 Oxygen isotope exchange between OH and H_2O

(U. Kläning (Chemistry Dept., Aarhus University), E. Bjergbakke and E. Larsen (Chemistry Dept.))

¹^oO enriched water saturated with N_2O with ¹^oO in natural abundance was studied by Y-irradiation and subsequent mass spectrometric analysis of isotopic O_2 formed by the irradiation. Preliminary results show that the predominant exchange reaction is

¹•OH + OH⁻ → ¹•OH⁻ + OH.

1.8 Y-radiolysis of aqueous $Br(1^{\circ}O)_{1}^{-}$ solutions

(U. Kläning (Chemistry Dept., Aarhus University), E. Larsen (Chemistry Dept.) and K. Sehested)

 $Br(1^{\circ}O)_{3}^{-}$ in aqueous solution at pH ~ 7 containing BrO_{4}^{-} in varying concentrations were Y-irradiated with 1 kGy (100 krd). The ¹⁸O content of the oxygen formed by the irradiation was measured by mass spectrometric analysis. The preliminary results indicate that ¹⁶O¹⁶O is formed by the reactions

 $Br(1^{\circ}0)_{3}^{-} + e_{aq}^{-} + Br(1^{\circ}0)_{2}^{-} + 1^{\circ}0H^{-} + 0H^{-}$

$$Br(10)_2 + 16OH + 16O10 + Br^{10}OH$$

Exchange reactions such as

$$Br({}^{10}O)_{3}^{-} + {}^{16}OH \xrightarrow{k} Br({}^{18}O)_{2}({}^{16}O)^{-} + {}^{16}OH$$

is not observed indicating that $k < 10^5 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$.

1.9 Pulse radiolysis of aqueous solutions of halides in the presence of hydrocyanic acid or potassium cyanide[#]

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

The early stages of radiolysis of aqueous solutions of halides (bromide or chloride) containing hydrogen cyanide at pH 3-4 and 10.3 were examined by a pulsed electron beam and fast kinetic spectrophotometry. Computer simulations of the reaction processes were used in fitting the experimental data and gaining a better insight into the reaction mechanism. It was found that in the radiolysis of the mixtures the same reaction pathways appear that are known to be involved in solutions containing halides or cyanides only. Some crossings of reaction pathways were found to take place in the bromide-cyanide system and were attributed to the reactions BrOH⁻ + HCN and Br + CN⁻. The mixed intermediates are formed with $k_{10} \approx 1 \times 10^7 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$ and $k_{10} \approx 2.5 \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$, and by reaction with Br_2^- , $k_{21} \approx 5 \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1}$. A rate constant for the reaction OH + Br⁻ + BrOH⁻, k(OH + Br⁻) = (5.0 \pm 0.5) \times 10^9 \text{ dm}^3 \text{mol}^{-1} \text{s}^{-1} was obtained with H_2 as competitor. The complete reaction scheme makes a prediction of the concentrations of intermediates in the

bromide-cyanide system possible. The crossing of pathways was not found in solutions of chloride containing cyanide.

1.10 The interaction between copper(II)-arginine complexes and HO_2/O_2^- radicals. A pulse radiolysis study[#]

(D.E. Cabelli, B.H.J. Bielski (Chemistry Dept., Brookhaven National Laboratory, Upton, N.Y., USA) and J. Holcman)

Superoxide radicals were shown to react with all four Cu(II)-arginine complexes that exist between pH 1.5-12.5; $k_{10}(O_2^- + CuArgH^{2+}) = 3.3 \times 10^9 M^{-1}s^{-1}$, $k_{12}(O_2^- + Cu(ArgH)_{2}^{2+}) = 1.3 \times 10^7 M^{-1}s^{-1}$, $k_{13}(O_2^- + CuArg_2H^+) = 1 \times 10^6 M^{-1}s^{-1}$, $k_{1*}(O_2^- + CuArg^+) = 1 \times 10^9 M^{-1}s^{-1}$. The system was studied in detail at pH 2.0 and pH ≥ 7 . At low pH the mechanism was shown to proceed via a CuO₂H²⁺ complex while, at higher pH, a superoxide complex of the Cu(II)-arginine complexes was formed. The results are discussed in terms of the currently accepted mechanism by which SOD-Cu²⁺ dismutates O_2^- radicals.

1.11 Superoxide dismutase

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

Superoxide dismutase (SOD) is known to disproportionate O_2^- radicals to O_2 and H_2O_2 . However, when exposed to H_2O_2 SOD shows catalase activity resulting in gradual inactivation of the enzyme. Deactivation of SOD by H_2O_2 is supposedly due to site specific formation of OH radicals. Our experiments using formate as a scavenger for site specific OH radicals and nitroblue-tetrazolium chloride (NBT) as an indicator/scavenger for CO_2^- radicals confirm site-specific formation of the OH radicals. This relatively simple system has proved to be an effective probe for reactions taking place within the active site of the SODs. The competition between NBT and O_2 for the CO_2^- radicals was studied. The results show that the ability of oxygen to compete with NBT for the CO_2^- -radical is substantially enhanced within the active site relatively to that in the bulk of the solution.

1.12 Intramolecular tree-electron bonds formation in methionyl homopeptides /di-L-Met/, /tri-L-Met*

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

Tree-electron-bonded sulphur-centered radical cations have recently gained prominence, as possible intermediates in electron transfer processes in biological systems, e.g. peptides, especially since these odd-electron species are unreactive toward molecular oxygen. In the present study we report the generation of three-electron-bonded species in methionyl homopeptides: methionyl-methionine /di-L-Met/ and methionyl-methionyl-methionine /tri-L-Met/.

Pulse radiolysis of 10^{-*} M di-L-Met and tri-L-Met, N₂O saturated aqueous solutions at pH - 5 provided two transient absorption bands with maxima at 290 nm and 390 nm. A different spectrum is obtained for the corresponding solution at pH 1. The 390 nm band is now absent and instead a broad, rather intensive absorption is seen with maximum at ca. 480 nm. A similar absorption band was generated during Tl^{2+} - induced oxidation of di-L-Met and tri-L-Met at low concentration in both acid and slightly acid solutions. These results suggest the existence of two various three-electron-bonded radical cations: intramolecular sulphur-centered radical cation complex with absorption maximum at 480 nm, and S.•.N three-electron-bonded radical cation identified as transient intermediate with absorption maximum at 390 nm. These were formed intramolecularly. All spectra include also a peak with $\lambda_{max} = 290$ nm attributable to α -/alkylthio/- alkyl radicals.

1.13 Pulse radiolysis study of aqueous solutions of methionine containing oligopeptides

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

Oxidation of oligopeptides by OH radicals was studied by means of pulse radiolysis. Absorption spectra and kinetics were observed for the following oligopeptides Met-Gly-Met, Met-Ala-Met, Met-Gly-Met-Met. The same type bonds as for previously studied peptides Met-Met and Met-Met-Met were observed, e.g. the band at $\lambda \cong 390$ nm attributed to the S.*.N bond and $\lambda \cong 490$ nm attributed to the S.*.S bond were observed. In contrast to the previous study a more complex kinetic behaviour suggesting equilibrium between S.*.S and S.*.N

- 12 -

species has been observed. Stability range for the S.*.S species was found to be shifted by approximately 2 pK units when going from Met-Gly-Met ($pK_a = 3.05$) to Met-Gly-Met-Met ($pK_a = 5.10$).

1.14 Intramolecular charge transfer between tryptophane and tyrosine in Trp-(Pro)n Tyr_peptides

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

Trp-(Pro)n Tyr, n = 0, 1, 2, peptides were oxidized by N_3^* radicals generated in pulse radiolysis. The selective formation of the radical at the tryptophyl residue was followed by electron transfer from the tyrozyl residue. Rate constants for this process were measured for the temperature range 5-65°C. Low activation energy for the intramolecular electron transfer - 0.2 eV suggests participation of electron tunneling in this process.

1.15 Pulse radiolysis of vitamins (nicotinic acid)

(N. Getoff, S. Solar, Institut für Strahlenchemie, Vienna, Austria),
J. Holcman and K. Sehested)

Reactions of nicotinic acid with H, e_{aq} and OH have been studied by pulse radiolysis. The hydrated electron forms an adduct with $pK_a = 5.5$ and $\lambda_{max} =$ 380 nm and 410 nm for basic and acid form, respectively. The hydroxyl radicals form an adduct with $pK_{a_1} = 1.3$ and $pK_{a_2} = 4.80$ with absorption maxima at 315 and 330 nm. In reaction with H-radicals three absorption maxima 285, 340 and 410 nm are observed. This demonstrates the formation of two distinct products: a cyclohexadienyl-type radical and a radical identical to the electron adduct. Indications are found that the cyclohexadienyl-type radical converts into the electron adduct radical by irreversible proton dissociation. 1.16 A quantitative description of the action of high-dose pulses of radiation on aerated acid solutions containing ferrous and chloride ions[#] (E. Bjergbakke, S. Navaratnam, B.J. Parsons (The North East Wales Institute, Deeside, Clwyd, UK), and A.J. Swallow (Paterson Institute for Cancer Research, Christie Hospital & Holt Radium Institute, Manchester, UK))

Kinetic measurements of the production of ferric complexes have been made for up to 100 seconds following a 1 µs pulse of 10-MeV electrons (165 Gy) given to aerated, acid solutions containing ferrous and chloride ions. The yields of ferric ions have been quantitatively accounted for by computations involving a scheme with 33 reactions. The rate constants necessary for a good fit are consistent with published values except for the oxidation of ferrous ions by chlorine atoms for which we now propose a value of 1.3×10^{10} dm³mol⁻¹s⁻¹.

1.17 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.

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2. RADIATION PHYSICS, DOSIMETRY, AND RADIATION PROCESSING

2.1 Particle track structure

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

<u>Track structure theory</u>. The radiation properties of the amino acid L- α -alanine has been investigated in respect to formation and recombination of radiation-induced free radicals at very high ionization densities. The rate of recombination of the CH, CHCOOH radical in irradiated alanine depends on the ionization density and is less than 1% per year for low-LET exposures (LET = Linear Energy Transfer) to doses below 10° Gy. At exposures to a high ionization density, e.g. 10⁵ Gy, the recombination amounts to approximately 10% in 1000 hours after the irradiation and after which stabilized conditions are obtained. These results from low-LET exposures indicate that the recombination is consistent with the assumption of a minimum distance between adjacent stable radicals. This provides a probe for studying the strongly inhomogeneous distribution of energy in the track of heavy charged particles for which increasing recombination rates at low average ionization density, single tracks, have been observed for increasing LET. Thus a high ionization density leads to a high concentration of trapped radicals which in organic solids, as it is observed, cannot stabilize closer together than approximately 10 nm. Excess radicals formed either by high doses of low-LET radiations of high dose rate or in the track core of high-LET particles, will recombine until an equilibrium is obtained. At a saturation dose of 10⁵ Gy an average distance between neighbouring radicals is theoretically found to be 3 nm, which is a factor of 3 below the distance considered to be the minimum for stabilized conditions. After exposure to 10* Gy from 6 and 16 MeV protons, single tracks, a recombination of approximately 6 and 3%, respectively, has been observed. Calculations show that in the tracks of 6 and 16 MeV protons in an alanine pellet 2 mm thick, 15 and 6%, respectively, of the particle energy is deposited at doses above 10^5 Gy. Formation of excess radicals and subsequent recombination after high-LET exposures must then be referred to the high doses inhomogeneously deposited in the track of a heavy charged particle and is independent of average dose to the sample.

Calculations on energy density distribution and observed radical recombination after high-LET exposures in a range in LET of $30-2\cdot10^4$ MeV cm²g⁻¹ are searched correlated as a function of velocity and charge of the bombarding particle. These investigations are related to theoretical considerations on model description for heavy charged particle and neutron interactions with tissue equivalent radiation detectors, and concern observed effects obscuring the evaluation of important parameters in the model.

<u>Tracks effects in alanine with heavy ions</u>. Electron spin resonance spectral analysis has been initiated in order to identify the radiation-induced radicals in alanine after exposure to high ionization densities. Differences in the spectra for alanine are observed after exposure to low-LET radiation to low and saturation doses. A comparison between ESR spectra obtained for alanine irradiated to a low dose of fast electrons and to a low average dose, single tracks, of 64 MeV oxygen ions also reveals a difference which points to a formation of transformed species in the track of a heavily ionizing particle. This supports the assumption of different chemical reactions going on along an area close to the primary particle path. In this region the terminal radical to radical reactions may be different from those in the thindown region of delta-rays at a larger distance from the part of the particle. From the point of view of ionization density, consequent radical concentration, and formation of other species, there is a justification in calling the area close to the path a track core. At high energies and low charge, e.g.

- 16 -

fast protons, the energy loss events are more separated and in that situation a core may actually not exist. Thus the concept of core is rather a high-LET phenomenon which is seen from a comparison of data and calculations for 16-MeV protons (LET = 30 MeVcm²g⁻¹) and, e.g. 64-MeV oxygen ions (LET = $7.2 \cdot 10^3$ MeVcm²g⁻¹).

<u>Fast neutron exposure of alanine</u>. Calculations on relative response for alanine exposed to cyclotron-produced fast neutrons at the Institute of Nuclear Physics, Krakow, have not yet been completed due to improvements to be made in our computer programs. During Dr. Waligorski's stay with Professor R. Katz (Lincoln, Nebraska, USA) work has been finished on a corrected formula for the radial distribution of dose around the path of a heavy particle. This formula is based mainly on our power-law model for slowing down low-energy electrons.

Part of this work has been supported by the Commission of the European Communities under contract number BI-6-0028-DK(B). The ESR spectral analysis has been performed at the Bruker ER 200 spectrometer at the Department of Physical Chemistry, University of Aarhus.

2.2. Investigation of the alanine dose meter

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

The simple amino acid L- α -alanine has been investigated for use as a radiation detector in low- and high-LET radiation fields. Data concerning detector sample preparation, dose-response relationships, fading of radiation induced response, and measured and calculated radiation sensitivity to heavy charged particles are presented. A new binding material, polyvidone, has been tested and compared to cellulose and paraffin. It offers better mechanical and thermal stability coupled with absence of zero dose signal and low sensitivity to radiation. Dose meters were prepared by pressing microcrystalline alanine powder with 5% polyvidone binder into small pellets, which after exposure to ionizing radiation were measured by means of electron spin resonance, ESR, spectroscopy. The radiation detector is cheap, robust and easy to handle. The low-LET radiation-induced free radicals are stable at normal laboratory conditions for doses below 10° Gy over a long period of time, which makes the detector useful for intercomparison and documentation purposes. Fading of radiation induced response has been observed in alanine detectors exposed to The work is supported by the Commission of the European Communities under contract number BI6-0028-DK(B). Irradiations with 740 MeV $^{\circ}$ Ar-ions were carried out at GSI, Darmstad.

2.3 A polymer-alanine (ilm for local dose measurements"

(J.W. Hansen and I. Janovský (Nuclear Research Institute, Řež, Czechoslovakia))

A 0.35 mm thick film consisting of a microcrystalline L- α -alanine imbeddec into an ethylene-vinyl acetate copolymer has been prepared in a cooperative project between the Nuclear Research Institute, Reź, and Risø. The alanine film has been investigated with respect to dosimetric properties as useful dose range, dose rate dependency, post-irradiation stability, and temperature effects. Results show a useful dose range of 25 to 10^5 Gy with a standard deviation on reproducibility of $\pm 4\%$ for doses above 100 Gy. No dose rate dependence has been found in the range of 1.2 to 10^7 Gy/s. Dose-response at elevated temperatures has been found to increase by approximately 10% over a temperature range from 25 to 80°C being dependent on dose. Radiation-induced response at doses in the linear dose range has been found to be stable within the experimental uncertainty over a period of 2500 hours after irradiation. The polymer alanine film proves to be suitable for various radiation technologies. This is due to a number of causes (a) the very broad useable dose range, (b) the relatively high radiation sensitivity, and (c) tissue equivalency in clinical dosimetry for lateral and depth dose measurements and for mimicking doses to the skin.

The Nuclear Research Institute, $\tilde{R}_3\tilde{z}$, will try to take out a patent in which Risø will have equivalent rights as the Czechoslovak Institute.

2.4 Radiation processing

(A. Miller)

Product irradiations at the-10 MeV linear accelerator have continued through 1986 at a normal level, which is two days a week including irradiations for other Risø departments and public institutions. Two exceptions from this guide occurred, namely at the beginning of 1986, where klystron problems caused product irradiations to be carried out three days a week, and during a 2-week breakdown at a Danish commercial irradiation facility, where we carried out product irradiations for this facility thus fulfilling our back-up obligation.

The product irradiations include radiation sterilization of medical and laboratory equipment, crosslinking of polymers, modification of semiconductor parameters and treatment of small amounts of spices, which were cleared for irradiation in 1985. Several test irradiations were carried out both at the 10 MeV accelerator, at the 400-keV accelerator and at the cobalt-60 facilities. A total of 43 customers made use of the Department's irradiation services during 1986.

Contact with and consulting for potential users of radiation were carried out, often in collaboration with the Polymer Group (Chemistry Department).

Students from Roskilde University Center and from the Agricultural University have carried out research projects on food irradiation, reflecting thereby the renewed interest in this conservation method.

2.5 Dosimetry for radiation processing

(A. Miller)

<u>Dose validation measurements</u>. The approval of radiation processes, in particular radiation sterilization and food treatment, depends essentially on initial dose validation measurements, which must prove that the absorbed dose is within approved limits. During 1986 we carried out several such measurements for products irradiated at Risø and at Danish irradiation facilities. National or international rules havve not been established that specify the degree of details needed for such measurements, however, and discussions have therefore been opened with Danish and - in particular - English authcrities with the aim of approaching agreement on such rules. Collaboration on this subject is maintained with the Danish irradiation facilities as well.

<u>Dose calibrations</u>. In several cases we have irradiated dosimeters to standard doses for customers who wish to establish calibration data for their dosimetry system. Such irradiations were carried out at the cobalt facilities and at the 10-MeV linear accelerator. Reference dosimeters have been sent for irradiation at a customer's facility in order to verify the doses administered. A total of 19 customers made use of the dosimetry services including dose validation measurements in 1986.

<u>Traceability</u>. Dose measurements and dosimeter calibrations must be traceable to primary standards. We are maintaining traceability by intercomparisons with National Bureau of Standards (NBS), USA, National Physical Laboratory (NPL), UK, and by irradiation of dosimeters through IAEA's Dose Assurance Service.

During 1986 an apparent discrepancy between our stated doses and those measured by the above institutions was discovered. The discrepancy was up to 5% which may be within the uncertainty obtainable. However, because it seems to be consistent, the possible causes for the difference are being investigated.

<u>Collaboration with IAEA</u>. The International Dose Assurance Service (IDAS) of the IAEA is now in operation for gamma irradiation facilities world-wide. An extension of this service to electron accelerator facilities is planned, and irradiation of candidate dosimeters from GSF, Munich, NPL and NBS were carried out at Risø and at NPL in the spring of 1986.

We took part in the evaluation of these irradiations as consultants to the IAEA. The results showed very good agreement between nominal doses at Risø and NPL, and measured doses as stated by GSF, NPL and NBS. Problems regarding the geometry of the dosimeters remain, however, and the work will continue at Risø and other places in order to find a useful dosimeter system. We have been receiving financial support to this work from IAEA under Research Contract No. 3583.

<u>Calorimetry</u>. Calorimeters serve as primary standards for irradiation at 10-MeV accelerators at Risø and elsewhere. We use graphite calorimeters for reference measurements and water calorimeters for routine measurements, and in a collaboration with Dr. Igor Janovský, Nuclear Research Institute, Czechoslovakia, we are investigating possible differences between graphite, water and aluminum calorimeters. Also in collaboration with Dr. Janovský we have attempted to use similar calorimeters as standards at the 400-keV accelerator, but as described in the previous progress report, several problems seemed to prevent this approach. During a visit to Risø by Dr. Janovský these problems were, however, successfully solved and it now seems possible to use the simple semi-adiabatic calorimeters at industrial accelerators in the energy range 0.4-10 MeV. Thin film dosimetry. Radiochromic dye dosimeter films are used extensively at the dose validation measurements, both for absolute dose determination and for dose distribution measurements. We use both commercially available films, and films which are produced as a result of a development project with a German firm. We have used and tested this dosimeter film to our satisfaction, but before deciding upon the marketing and sale of this film dosimeter, a further improvement is being tested.

Another dosimeter for reflected light measurement in the form of a tape is being developed in collaboration with the same firm as above under a contract with CERN, Geneva, who want the dosimeter for accelerator component testing. A prototype of this dosimeter has been produced, and it is also being investigated, if this dosimeter can be used routinely for product irradiation.

Educational activities. We have received 4 IAEA fellows during 1986, who during 4-6 week sojourns had an opportunity to familiarize themselves with dosimetry techniques for radiation processing. In several cases they also took an active part in the ongoing measurement programs.



- Figure 1: Graphite calorimeter for dose calibration of industrial low-energy electron accelerators.
 - 1. Graphite body, diameter 136 mm, thickness 10 mm.
 - 2. Thermistor for temperature measurement.
 - 3. Polystyrene foam for thermal insulation.
 - 4. Thin polyethylene film acting as protective cover.
 - 5. Fiber plate that defines the irradiated area of the graphite calorimeter body.

Temperature measurement before and after irradiation provides an accurate dose measurement due to the adiabatic behaviour of the calorimeter (Janovský and Miller, 1987).

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3. OPERATION AND MAINTENANCE OF THE IRRADIATION FACILITIES

3.1 HRC electron linear accelerator

(J. Fenger and B. Lynggård)

Operation of the accelerator has been influenced by abnormal situations. During March and September the operation was interrupted due to replacement and repair of vital accelerator components. Since September 1985 and until March 1986 the beam power was reduced due to a defective klystron, and the operation time for commercial irradiation was extended. The accelerator has been used 31% of the time for commercial irradiation, 7% for service irradiation, 32% for experimental irradiation and 30% for repairs and maintenances.

In connection to the approval of the accelerator for food irradiation the liability of the radiation safety for the accelerator was taken over by "Statens Institut for Stralehygiejne".

The spare part situation was improved during the year; two klystrons, one thyratron and one microwave coupler were purchased.

Within the year the following repairs and maintenances were carried out:

Injector system. The only non-cooled collimator in the beam optics was replaced by a water-cooled one. The new unit designed in the Department, has solved heating-up problems of the injector vacuum valve and has reduced the heat-up of the beam tube, thus eliminating oil leck from a gasket between the beam tube and the gun-oil tank. The electronics for the gun steering was modified in order to obtain a more precise shape of the gun pulse. The regulation of the 250-kV gun high voltage was improved.

<u>Microwave system</u>. The old defect klystron, S/N 1023, was replaced after 8000 hours of operation. The original klystron directional coupler, temporarily repaired in 1985, was replaced with the spare coupler, and the original one was repaired in the Department. The 30 l vacuum pump in the rectangular waveguide system was replaced with a renovated pump.

<u>Vacuum system</u>. The collimator/monitor assembly at the output of the accelerator waveguide leaked from the water chamber into the vacuum system. The assembly was repaired by Haimson Research Corporation in the USA. The repairs took one month, and the whole vacuum system was carefully leak-tested by use of a He-leak detector, kindly lent by the Plasma Physics Group (Physics Department). A ceramic insulating tube and a ceramic feed-through leaked. A new insulating tube was designed in the Department. A new feed-through was designed and delivered from Vacuum Service in Sweden.

Due to corrosion in the flange for the scanner window, leaks developed from time to time, and after 10 years of operation the window flange was machined and electroplated.

Beam handling system. A new beam scanning system, scanner coil, and power supply for the 90° magnet were installed. Both the scanner system and the 90° magnet power supply were designed in the Department. The design of the new scanner is based on energy oscillation between the scanner coil and a capacitor bank.

The water cooling system was overhauled, the filter media in the oxygen filter was renewed. Some of the components in the cooling system are corroded and will be replaced.

The conveyor system for industrial irradiation was overhauled, and some improvements were introduced. The old recorder for secondary emission current was replaced by a new 6-channel recorder; the other channels are used for recording other beam parameters.

3.2 Febetron, field-emission accelerator

The field-emission accelerator was used for pulse radiolysis of gases. The following maintenance was carried out:

A module failed and was replaced. The switch that shifts between negative and positive pulser voltages failed due to aging; as we use only negative pulses the contacts were shorted in that position, and the accelerator can no longer be used for positive pulses (x-ray tubes). Losses of dry air (insulating gas) have gradually increased; the seals were checked, and the cables were sealed with epoxy between the inner conductor and the insulation.

3.3 ICT, low energy accelerator

The low-energy accelerator was used for dosimetry and polymerization experiments. Minor repairs were carried out on the conveyor system.

3.4 ⁶ ^oCo-facilities

In the spring of 1986 all three ^{6°}Co-facilities (the 10,000 Ci-facility in the Agricultural Department, the 5,000 Ci-gammacell at the Danish State Serum Institute and the 3,000 Ci-gammacell in the Accelerator Department) were upgraded to the maximum capacity. The radioactive ^{6°}Co was produced in Research Reactor DR3 and the encapsulation was performed in the Hot Cells facility. The capsules and various tools were constructed at the Engineering Workshop.

The ⁶°Co sources from the 3,000 Ci-cell were removed and transferred to Hot Cells for storage, and the cell was taken to the Engineering Workshop for a general overhaul. The facility was installed in another room in the Accelerator Department and loaded with the old ⁶°Co sources from the 10,000 facility.

The 10,000 Ci-facility was loaded with eight new sources, and the 5,000 Ci-cell was upgraded by two new sources of 1,500 Ci each. After completing the reloading all three facilities were reassembled and calibrated.

10,000 Ci * Co-facility

The 10,000 Ci ⁶⁰Co-facility was used for radiation research and for customer services.

5,000 Ci ⁶⁰Co-facility

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

3,000 Ci ⁶ Co-facility

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

3.5 Lambda Physik Excimer laser

The laser was used for fast ESR experiments at the Chemistry Department, Aarhus University, and for Raman spectroscopy and flash photolysis at Risø. The operation of the laser was trouble-free.

3.6 Flash-photolysis set-up

The flash-photolysis equipment consisting of the light source, optics, detectors, light shutters and synchronizing electronics was installed in the new Laser Laboratory.

3.7 Pulse radiolysis set-up

The equipment was used intensively during the year, also by several visiting scientists.

The analyzing light intensity was increased by use of a 300 W xenon lamp instead of the normally used 150-W lamp.

4. EDUCATIONAL ACTIVITIES AND PUBLICATIONS

4.1 Lectures

K. Bobrowski and J. Holcman, Pulse Radiolysis Generation of Three Electron Bonded Species in di- and tri-Peptides Containing Methionine. 7th Conference of Polish Radiation Research Society, Kodz, Poland, 17-19 March.

K. Bobrowski and J. Holcman, Intramolecular Three-Electron Bonds Formation in Methionyl Homopeptides / di-L-Met, /tri-L-Met. Int. Meet. on Radiation Chemistry and Processing, Brdička Days 1986. Mariánské Lázne, Czechoslovakia, 21-25 April. K. Bobrowski and J. Holcman, Pulse Radiolysis Generation of Three Electron Bonded Species in di- and tri-Peptides Containing Methionine. Troisiemes Journees de'Etudes sur la Chimie des Radiations, Austrans, France, 10-13 June.

K. Bobrowski, Charged Transfer in Simple Model Peptides. Intramolelcular
 radical transformations involving methionine, tryptophan and tyrosine units.
 Risø, 23 December.

J.W. Hansen, Detection of Low- and High-LET Radiation with Alanine. Int. Meet. on Radiation Chemistry and Processing, Brdiàcka Days 1986. Mariánské Lázne, Czechoslovakia, 21-15 April.

J.W. Hansen, Heavy Charged Particle Dosimetry, Theory and Application. Symposium of Radiological Physicist, Smolenice, Czechoslovakia, 22-25 September.

J. Holcman, E. Bjergbakke and K. Sehested, The Importance of Radical-Radical Reactions in Pulse Radiolysis of Aqueous Carbonate/Bicarbonate. 6th Tihany Symposium on Radiation Chemistry, Balatonszéplak, Hungary, 21-16 September.

A. Miller and W. Batsberg, "Sterilisation og tværbinding af polymerer ved bestråling", Polymerteknisk Selskab. Risø, 13 marts.

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K.J. Olsen and J.W. Hansen, Radiation Detector Mimic Human Tissue. University of Wisconsin, Madison, Wisconsin, USA, 23 June.

K. Sehested, Fotokemi og Strålingskemi. Forelæsninger ved Kemisk Institut, Århus Universitet, 29-30 January, 18-19 February and at Risø 3 March.

4.2 Publications

Accelerator Department - Annual Progress Report, 1 January - 31 December, 1985. Risø-M-2559.

D. Berenstein, Prophage Induction by Ultraviolet Light in Acinetobacter calcoaceticus. Journal of General Microbiology **132**, 2633-2636.

K. Bobrowski and J. Holcman, Intramolecular Three-Electron Bonds Formation in Methionyl Homopeptides (di-L-Met), (Tri-L-Met). Radiat. Phys. Chem 28, No. 5/6, pp. 555 (Abstract).

H. Christensen and K. Sehested, The Hydrated Electron and Its Reactions at High Temperatures. J. Phys. Chem. **90**, 186.

H. Christensen and E. Bjergbakke, a-Radiolysis of Bentonite/Water Mixtures. Studsvik Technical Note NS-86/23.

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5. IRRADIATION FACILITIES AT THE ACCELERATOR DEPARTMENT

Electron Accelerators

1. Linear Electron accelerator, Haimson Research Corp. Model HRC-712

Accelerator room beam facilities:

- 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 A Pulse length 50 ns 3. Low-energy Electron Accelerator, High Voltage Eng. Corp. Model EPS 400-IND

 Specifications:

 Electron energy

 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	F2	ArF	KrC1	KrF	XeC1	N ₂	XeF
Wavelength	1 57	193	222	249	308	337	351 nma
Pulse energy (min)	10	2 0 0	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

* Co-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 (9.680 Ci $(3.60\times10^{1+} \text{ Bq})$, 1 January 1987) are 6.0×10^{5} , 1.66×10^{6} , and $3.8\times10^{6} \text{ rad}\cdot\text{h}^{-1}$ (1.74, 4.6, 10.4 Gy·s⁻¹), respectively.

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

Designed for laboratory use and fitted with a 123 mm⁹×150 mm irradiation chamber. The dose rate in the center of the chamber (5.360 Ci $(2.0\times10^{14}$ Bq), 1 January 1987) is 4.2×10^{5} rad·h⁻¹ (1.17 Gy·s⁻¹). The cell is located at the Control Department, National Serum Institute, Copenhagen.

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

3,000 Ci **Co-cell (built at Rise 1968)

Designed for laboratory use and fitted with a 120 mm^{0} ×220 mm irradiation chamber. The dose rate in the center of the chamber (4.256 Ci (1.58×10¹* Bq), 1 January 1987) is 4.35×10⁵ rad·h⁻¹ (1.21 Gy·s⁻¹).

Sources were replenished in all 3 facilities in April 1986.

6. VISITING SCIENTISTS

- K. Bobrowski, Institute of Biochemistry and Biophysics, Warsaw, Poland.
- D. Cabelli, Brookhaven National Laboratory, Upton, L.I., N.Y., USA.
- H. Christensen, Studsvik Energiteknik AB, Nyköping, Sweden.
- A. Dodbiba, Institute of Nuclear Physics, Tirana, Albania.
- N. Getoff, Institut für Strahlenchemie, Vienna, Austria.
- E.J. Hart, Port Angeles, WA., USA.
- I. Janovský, Nuclear Research Institute, Reź, Czechoslovakia.
- U. Kläning, Kemisk Institut, Aarhus University, Aarhus.
- W.L. McLaughlin, National Bureau of Standards, Gaithersburg, MD., USA.
- E. Martinez Pardo, Nuclear Center ININ, Mexico, Mexico, D.F.
- D. Rexha, Institute of Nuclear Physics, Tirana, Albania.
- P. Sharpe, National Physical Laboratory, Teddington, Middlesex, England.
- S. and W. Solar, Institut für Strahlenchemie, Vienna, Austria.
- W. Wanisuksombut, Office of Atomic Energy for Peace, Bangkog, Thailand.
- W. Warasawas, Office of Atomic Energy for Peace, Bangkok, Thailand.
- L. Xie, Institute of Low Energy Nuclear Physics, Beijing Normal University, Beijing China.

7. STAFF OF THE ACCELERATOR DEPARTMENT

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Rise National Laboratory	Rise – M – 2633			
Title and arthor(s)	Date February 1987 Department or group Accelerator Groups own registration numberts			
Accelerator Department Annual Progress Report for the period 1 January - 31 December 1986.				
Acceleratorafdelingens årsrapport 1 januar – 31 december 1986.				
	Project/contract no.			
Pages 35 Tables Hhustrations 1 References	ISBN 87-550-1308-2			

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

Descriptors _ INIS

ACCELERATOR FACILITIES; DOSEMETERS; DOSIMETRY; IRRADIATION DEVICES; REACTION KINETICS; RESEARCH PROGRAMS; RISOE NATIONAL LABORATORY

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