Pacific Northwest Laboratory Annual Report for 1985 to the DOE Office of Energy Research

**Part 4 Physical Sciences
February 1986**

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
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L. H. Toburen and Staff Members
of Pacific Northwest Laboratory

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Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This 1985 annual report from Pacific Northwest Laboratory (PNL) to the Department of Energy (DOE) describes research in environment, health, and safety conducted during fiscal year 1985. The report again consists of five parts, each in a separate volume.

The five parts of the report are oriented to particular segments of our program. Parts 1 to 4 report on research performed for the DOE Office of Health and Environmental Research in the Office of Energy Research. Part 5 reports progress on all research performed for the Assistant Secretary for Environment, Safety and Health. In some instances, the volumes report on research funded by other DOE components or by other governmental entities under interagency agreements. Each part consists of project reports authored by scientists from several PNL research departments, reflecting the multidisciplinary nature of the research effort.

The parts of the 1985 Annual Report are:

Part 1: Biomedical Sciences

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Part 2: Environmental Sciences

Program Manager - R. E. Wildung

R. E. Wildung, Report Coordinator
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Part 3: Atmospheric Sciences

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E. L. Owczarski, Editor

Part 4: Physical Sciences

Program Manager - L. H. Toburen

L. H. Toburen, Report Coordinator
J. E. Danko, Editor

Part 5: Overview and Assessment

Program Manager - L. G. Faust

L. G. Faust, Report Coordinator
R. W. Baalman, Editor

Activities of the scientists whose work is described in this annual report are broader in scope than the articles indicate. PNL staff have responded to numerous requests from DOE during the year for planning, for service on various task groups, and for special assistance.

Credit for this annual report goes to many scientists who performed the research and wrote the individual project reports, to the program managers who directed the research and coordinated the technical progress reports, to the editors who edited the individual project reports and assembled the five parts, and to Ray Baalman, editor in chief, who directed the total effort.

A highlight this past year was the appointment of a Scientific Advisory Committee. Honoring us by accepting our invitation to serve on the committee are:

Dr. Franklin I. Badgley	University of Washington
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1953	HW-30437, HW-30464
1954	HW-30306, HW-33128, HW-35905, HW-35917
1955	HW-39558, HW-41315, HW-41500
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1961	HW-72500, HW-73337
1962	HW-76000, HW-77609
1963	HW-80500, HW-81746
1964	BNWL-122
1965	BNWL-280; BNWL-235, Vol. 1-4; BNWL-361
1966	BNWL-480, Vol. 1; BNWL-481, Vol. 2, Pt. 1-4
1967	BNWL-714, Vol. 1; BNWL-715, Vol. 2, Pt. 1-4
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1971	BNWL-1650, Vol. 1, Pt. 1-2; BNWL-1651, Vol. 2, Pt. 1-2
1972	BNWL-1750, Vol. 1, Pt. 1-2; BNWL-1751, Vol. 2, Pt. 1-2
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1978	PNL-2850, Pt. 1-5
1979	PNL-3300, Pt. 1-5
1980	PNL-3700, Pt. 1-5
1981	PNL-4100, Pt. 1-5
1982	PNL-4600, Pt. 1-5
1983	PNL-5000, Pt. 1-5
1984	PNL-5500, Pt. 1-5

FOREWORD

Part 4 of the Pacific Northwest Laboratory Annual Report for 1985 to the DOE Office of Energy Research includes those programs funded under the title "Physical and Technological Research." The Field Task Program Studies reports in this document are grouped by budget category and each section is introduced by an abstract that indicates the Field Task Proposal/Agreement reported in that section. These reports only briefly indicate progress made during 1985. The reader should contact the principal investigators named or examine the publications cited for more details.

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Analytical Studies

• Chemical Basis for Biological Response to Complex Organic Mixtures

Several studies to identify biologically active components in complex mixtures derived from coal and shale oil have been completed during the past year. Knowledge gained from this work will be applied to develop more reliable methods for determining potential risk from exposure to complex organic mixtures arising from production and use of fossil fuel energy resources.

These studies continue to make important contributions to general knowledge about biological effects of complex mixtures in the environment. Fundamental aspects of this research are funded by the Office of Health and Environmental Research (OHER). Research related more specifically to processes and process modification is funded by the Office of Fossil Energy.

CHARACTERIZATION OF TUMORIGENIC COMPONENTS IN SHALE-DERIVED OIL

C. W. Wright, J. A. Franz, E. K. Chess, and B. W. Wilson

Shale-derived oils are more complex than their coal-derived counterparts having similar distillation temperature ranges. This additional complexity arises from the presence of several functional groups, including carbonyl moieties, not generally found in coal liquids, and from extensive alkylation of polycyclic aromatic compounds (PAC). The highly aliphatic nature of shale oils versus coal liquids is shown in the nuclear magnetic resonance (NMR) spectra (Figure 1). The complexity of shale oils versus coal liquids is also shown by the gas chromatograms of the components eluting in the polycyclic aromatic hydrocarbon (PAH) fractions. These fractions were isolated by alumina column chromatography from the 700-750°F distillates of both materials. Little to no resolution of components was achieved with the shale oil by gas chromatography or gas chromatography/mass spectrometry. Low-voltage high resolution mass spectrometry was used for homologous series identification of possible structures found in the complex shale oil matrix.

Shale oil materials have generally exhibited lower mutagenic and tumorigenic activity than comparable coal-derived materials. Attempts to characterize and identify specific compounds, or chemical classes of compounds, responsible for these biological responses have been largely unsuccessful. In early studies using fractional distillation, the mutagenicity and carcinogenicity of shale-derived oils, unlike that of coal liquids, showed little, if any, dependence on boiling

temperature. Less biological activity was detected in the distillates boiling above 650°F than was detected in the crude, full-boiling-range shale oil. In addition, the microbial mutagenicity of the shale oil was not associated with the nitrogen-containing PAC (NPAC) fraction resulting from alumina column chromatography as it has been with coal-derived materials.

Because separation of compounds by boiling point, and hence molecular weight, was not effective in isolating chemical toxicants, development of methods to provide specific chemical class fractions for biological testing and further chemical analyses became an important priority on this project. A normal phase, high-performance liquid chromatography (HPLC) method, with separation based on polarity, yielded sufficient quantities of materials for initiation/promotion (I/P) testing and microbial mutagenicity testing. An HPLC chromatogram showing the conditions of the separation and the fractions collected from the analysis of Paraho shale oil is given in Figure 2. Results of the microbial mutagenicity testing of HPLC fractions are shown in Figure 3. Because mutagenic activity was essentially isolated in HPLC fractions 4, 5, and 6, these were combined to form "HPLC fraction 4" for the I/P assay. Results of the I/P assay are given in Table 1.

The dosages of each chemical class (HPLC) fraction applied to the test animals were proportional to their concentration in the crude shale oil. Thus some conclusions can be drawn from the I/P data regarding the relative importance of aliphatics, neutral PAH and alkylated neutral PAH, moderately polar compounds (including pyrroles and azaarenes), and highly polar compounds to the

FIGURE 1. Gas Chromatographic and Nuclear Magnetic Resonance Comparisons of Coal- and Shale-Derived Oils.

overall tumorigenicity of the shale oils tested. The majority of tumorigenic activity was isolated in HPLC fraction 2 of the shale oil. Components eluting in this fraction included the neutral parent and alkylated PAH of three to six rings. HPLC fractions 1 and 3 showed similar tumorigenic activity, but significantly less activity than HPLC fraction 2. Components eluting in these fractions included neutral aliphatic hydrocarbons

(AH) and aromatic hydrocarbons of one to two rings in fraction 1, and parent and alkylated pyridines and pyrroles in moderately polar fraction 3. HPLC fraction 4 (which displayed the greatest amount of microbial mutagenicity) was the least tumorigenic fraction of the shale oil. The highly polar components (including some multi-heteroatomic compounds) were eluted in this fraction.

FIGURE 2. Normal Phase High-Performance Liquid Chromatography Fractionation of Crude Paraho Oil Shale.

FIGURE 3. Mutagenic Activity of Shale Oil High-Performance Liquid Chromatography (HPLC) Fractions with TA 98.

COMPARATIVE INHIBITION OF BENZO[A]PYRENE BINDING TO DNA BY CHEMICAL CLASS FRACTIONS FROM COMPLEX MIXTURES

D. L. Springer, D. W. Later, M. E. Frazier, and O. D. Mahlum

An important effort in the past year has been to measure DNA adducts as indicators of risk associated with exposure to complex chemical mixtures. Recent studies have demonstrated that expression of carcinogenic compounds

TABLE 1. Skin Tumor Initiating Activity of Shale Oil and HPLC Fractions

Treatment Group	Tumor Incidence (in %)	No. of Tumors per Mouse ^(a)
Control (methylene chloride/acetone)	100	0.1
Shale oil (25 mg)	43	0.5
HPLC #1 ^(b)	23	0.3
#2 ^(b)	50	0.7
#3 ^(b)	23	0.3
#4 ^(b)	17	0.2
Benzo[a]pyrene (25 μg)	100	7.1

(a) Based on groups of 30 mice.

(b) Doses administered were proportional to amount in parent material (crude shale oil).

added to coal liquids may be inhibited by components in the complex mixture. The mechanisms of inhibition are central to understanding biologic response to complex mixtures.

Narrow boiling point range distillate cuts from a coal liquid, specific chemical class fractions from one of the distillate cuts, and a specific amount of radiolabeled benzo[a]pyrene (BaP) were applied to the skin of male rats. Using radiocounting techniques on DNA isolated from skin, the amount of bound BaP was determined at several time points.

The 800-850°F and the 850°F+ distillate cuts from solvent refined coals (SRC) II were used as test materials, along with chemical class fractions derived from the distillates, including aliphatic, polycyclic aromatic hydrocarbon (PAH), nitrogen-containing polycyclic aromatic compounds (NPAC), and hydroxy-PAH.

Binding of BaP alone at a dose of 50 mg/animal resulted in approximately 22 picomoles of bound BaP per mg of DNA recovered from the skin. When a similar amount of BaP was co-administered with 25 mg of distillate, binding was decreased by approximately 99%. In a subsequent experiment, the amount of distillate administered was decreased to

500 micrograms, with the 850°F+ distillate exhibiting significantly less inhibition than the 800-850°F cut.

When BaP was co-administered with the aliphatic fraction, binding of BaP to DNA was not affected. The other three fractions, each of which contained aromatic compounds, showed significant inhibition of binding to DNA. Of the latter fractions, the neutral PAH showed the greatest inhibition (binding was decreased by 47%), followed by the NPAC and hydroxy PAH fractions. Thus, the greatest inhibition of binding arose from the fraction most similar chemically to BaP.

DISTILLATION TEMPERATURE AS A PREDICTOR OF CARCINOGENIC AND MUTAGENIC ACTIVITY IN ENERGY-RELATED COMPLEX MIXTURES

C. W. Wright, J. P. Fillo, J. C. Craun, D. W. Later, and B. W. Wilson

Distillation parameters, bioassay results, and chemical composition data for materials from several processes, including SRC-I, SRC-II, H-Coal, integrated two-stage liquefaction (ITSL), and EDS, have been compiled and evaluated to determine similarities and differences in the effect of distillation temperature on biologic activity.

Regardless of process technology, microbial mutagenicity of coal liquid generally increased as boiling range temperature increased. Microbial mutagenicity, associated mainly with the nitrogen-containing polycyclic aromatic compounds (NPAC) fractions of coal liquids, tended to correspond to the detection of three- to four-ringed amino-PAH. Amino-PAH content and, hence, microbial mutagenicity, was significantly less for the EDS and ITSL coal liquefaction materials as compared to the SRC and H-Coal materials. Mutagenic neutral PAHs were isolated in the high-performance liquid chromatographic subfractionation of the PAH fraction; the most active subfractions contained the highest concentrations of benzo[a]pyrene (BaP).

For each process from which samples were available, significant tumor-initiating activity was found for narrow range distillates above 700°F. As boiling point (bp) increased, both the incidence of tumor and

total number of tumors increased; the highest boiling distillate was the most potent skin tumor initiator for all materials. The neutral PAH fractions of both the SRC-II 800-850°F and 850°F+ distillates showed the greatest amount of initiating activity compared to their respective aliphatic hydrocarbon (AH), NPAC and hydroxy-PAH chemical class fractions. In addition, the high-performance liquid chromatography (HPLC) subfractions of the active PAH fractions containing the highest levels of BaP were the most active in the initiation/promotion (I/P) assays for mouse skin tumorigenicity.

The highest boiling point distillates of SRC-I, SRC-II and ITSL materials gave the highest mutagenic response in the Chinese hamster ovary (CHO) cell assay. Similar to I/P results, the PAH fractions were the most active chemical class fraction in this assay.

Distillates boiling above 700°F were mainly composed of four- to six- (or more) ringed polycyclic aromatic compounds (PAC) plus their alkylated homologs. Characterization of SRC and H-Coal chemical class fractions demonstrated that similar species were present in these coal liquids. EDS and ITSL materials tended to contain chemical species that were more highly alkylated and hydrogenated and had lower concentrations of amino-PAH and azaarene compared to process materials without catalytic hydrogenation. In addition, EDS materials had a slightly higher average molecular weight than corresponding materials of the same distillation range from other processes. Phenyl-substituted PAC (such as phenylnaphthalene and hydroxybiphenyl) was found in significantly less concentration in the EDS material compared to SRC-II or H-Coal materials.

Several distillation variables were evaluated as predictors of tumorigenic and mutagenic activity of full boiling range materials. The "95% over" temperature (i.e. the temperature at which 95 wt% of the starting material has distilled over) was found to be a good indicator of toxicological response. Statistical analyses showed that as a group, full boiling range materials from the SRC-II, H-Coal, EDS, and ITSL processes were mutagenic and/or tumorigenic ($P < 0.05$) if their 95% distillation cut point was above 660°F, provided the dose was 20 to 30 mg or greater. Materials with 95% over distillation cut

points below 660°F did not show statistically significant activity ($P < 0.05$).

Thus the 95% over distillation temperature appears to be a useful indicator of mutagenic and tumorigenic activity for coal-derived complex mixtures that have not undergone post-production hydrotreatment. It should also be noted that certain low molecular weight amino PAH, including the carcinogen 2-aminonaphthalene, are not readily detected in complex mixtures by the bioassays used in these studies, and that materials with 95% distillation temperatures below 660°F may contain low concentrations of such compounds.

IDENTIFICATION OF PREVIOUSLY UNCHARACTERIZED COMPOUNDS IN ENERGY-RELATED COMPLEX MIXTURES

E. K. Chess, D. W. Later, and J. A. Franz

Identifying specific compounds that either act as toxicants or are potentially important in understanding chemical mechanisms in certain biological processes is of continuing interest in the chemistry component of the complex mixtures program. Over the last year, two previously uncharacterized carbazole isomers and a novel hydrogenated pericondensed three-ring compound have been isolated and identified in coal liquids.

Nitrogen-containing polycyclic aromatic compounds (NPAC) are generally found in products of coal utilization from processes such as liquefaction, gasification, combustion, and coking. Carbazole, a three-ring pyrrole compound, is typically one of the most concentrated NPACs found in such materials. In studying the chemical composition of the pyrrolic-PAH isolate of a solvent refined coal (SRC) liquid, other isomers of carbazole (nominal molecular weight 167) were detected by gas chromatography-mass spectrometry (GCMS). From subsequent analyses, carbazole and two additional $C_{12}H_9N$ pyrrolic isomers have been identified: 1H-benz(g)indole and 3H-benz(e)indole. Complete isolation of the three $C_{12}H_9N$ isomers was achieved by an integrated separation method using adsorption, liquid, and gas chromatography. Molecular structures and elemental compositions were determined by high resolution mass spectrometry (HRMS), ultraviolet (UV) spectroscopy, and nuclear magnetic resonance (NMR) spectroscopy. Carbazole, 1H-benz(g)indole and

3H-benz(e)indole were identified in an SRC distillate liquid and a commercial creosote oil. These isomers are of interest because carbazole benzologs are known to be important skin carcinogens in certain coking by-product tars and oils.

2,3-Dihydro-1H-phenalene, a partially hydrogenated, three-ring, pericondensed polynuclear aromatic hydrocarbon (PAH), was found to be a major component in the neutral PAH fractions of catalytic two-stage liquefaction process materials. The identification was based on results from analyses by GC, MS, and NMR of the coal liquid fractions and a synthesized reference compound. A simple synthetic route to this compound from the readily available starting material, phenalene-1-one, was devised and used. Because pericondensed compounds and their partially hydrogenated analogs have been identified as important species in hydrogen shuttling during catalytic hydrogenation in coal liquefaction processes, the identification of relatively large amounts of 2,3-dihydro-1H-phenalene in the products of two different catalytic two-stage liquefaction processes is noteworthy, as it suggests that this compound may play a previously unrecognized role in the liquefaction process.

STUDIES RELATING TOXICOLOGICAL ACTIVITY AND CHEMICAL COMPOSITION DURING A DEMONSTRATION RUN

C. W. Wright, E. K. Chess, D. D. Mahlum, and D. W. Later

Understanding effects of catalyst age and other process conditions on the biological activity of process streams is important in optimizing process operations from the occupational health standpoint. In studies described here, samples were obtained on selected days during a 25-day demonstration run of a catalytic two-stage liquefaction (CTSL) process. These samples were assayed for tumorigenicity and mutagenicity and were characterized chemically. Resulting data were compared to process conditions such as catalyst age, etc.

In the CTSL process studied a catalyst is used in both the first- and second-stage reactors. The first stage is operated at relatively lower temperatures. Results from

the CTSL process were compared with those obtained on materials from other two-stage coal liquefaction processes: the integrated and nonintegrated two-stage liquefaction processes (ITSL and NTSL). Both these processes used catalysts in the second-stage hydrogenation reactor but did not incorporate a catalyst in the first-stage thermal reactor.

Data were acquired on the pressure filter liquid (PFL), an internal post-second-stage process material, and the product distillate blend (PDB), more representative of a final product. In addition, both first- and second-stage materials from a previous experimental run of this coal liquefaction process were analyzed. Samples were chemically analyzed by column adsorption chromatography, high-resolution gas chromatography (GC), GC/mass spectrometry (GCMS), low-voltage probe-inlet MS, and proton nuclear magnetic resonance. Toxicological testing was performed using the histidine reversion assay for microbial mutagenicity and the initiation/promotion assay for tumorigenicity in mouse skin.

The following results were obtained from the analyses of PFL materials. The major components were aliphatic hydrocarbons (AH), parent and alkylated polycyclic aromatic hydrocarbons (PAH), and parent and alkylated hydroaromatic PAH. There was an increase in heteroatomic, nitrogen-containing polycyclic aromatic compounds (NPAC) and hydroxy-substituted PAH (hydroxy-PAH) content as the time of pilot plant operation increased. The proportion of PFL material boiling above 975°F, as well as the concentration of heavy molecular weight components (greater than four rings), also increased as run time increased. A concomitant decrease in AH and hydroaromatic content was noted in the PFL materials as the demonstration run progressed. Microbial mutagenicity of the PFL materials increased with pilot plant operation time, and PFL materials from later in the 25-day demonstration run showed a tendency toward increased tumorigenic response compared to those from earlier periods. Although the most pronounced changes in chemical composition occurred during the initial days of operation when temperatures

were equilibrating, the overall changes indicated the deleterious effects of catalyst degradation during the demonstration run.

Chemical components in the CTSL PDB were similar to those detected in the PFL, except that they were of a lower molecular weight range (an effect of the blended lower-boiling distillates used to make the PDB). The PDB gave a lower tumorigenic response than did the PFL; likewise, a lower mutagenic response was noted for the PDB compared to the PFL materials from after about 10 days of pilot plant operation. The PDB also displayed some degree of direct-acting mutagenesis.

Results of chemical analyses were similar for both the first- and second-stage materials from the experimental CTSL run, with the exception that the first-stage material had more heavy molecular weight components. The tumorigenic response of the first-stage material tended to be slightly greater than that of the second-stage material. Both the tumorigenic response and the molecular weight range of these materials were less than those from the 25-day demonstration run.

Compared to the ITSL and NTSL processes, the CTSL materials generally had lower concentrations of AH, PAH and hydroaromatic PAH. The degree of alkylation and hydrogenation of CTSL materials was the same or less than that of the ITSL and NTSL materials. The CTSL PDB gave lower tumorigenic response than did the ITSL and NTSL materials, lower mutagenic response than the NTSL materials, and slightly higher mutagenic response than the ITSL materials. The chemical composition of the CTSL PDB most closely resembled that of the NTSL material. Greater reductions in tumorigenicity were noted in first- and second-stage materials from the ITSL and NTSL processes than in the CTSL process, indicating that a catalyst in the first-stage reactor may reduce the tumorigenic potential of first-stage coal liquefaction products.

Chemical analysis and biological testing data on the CTSL process materials correlated with previous data showing that mutagenic potency was generally contained in NPAC fractions and that tumorigenic potential was generally a function of increased boiling point.

• Supercritical Fluid Analytical Methods

Supercritical fluids are providing the basis for new and improved methods across a range of analytical technologies. New methods are being developed to allow the detection and measurement of materials that are incompatible with conventional analytical methodologies. Characterization of process and effluent streams for synfuel plants requires instruments capable of detecting and measuring high molecular weight compounds, polar compounds, or other materials that are generally difficult to analyze. The purpose of this program is to develop and apply new supercritical fluid techniques for extraction, separation and analysis. These new technologies will be applied to previously intractable synfuel process materials and to complex mixtures resulting from their interaction with environmental and biological systems.

SUPERCRITICAL FLUID CHROMATOGRAPHY AND MASS SPECTROMETRY

R. D. Smith, B. W. Wright, and H. R. Udseth

Chromatographic methods combined with mass spectrometry compose a nearly ideal analytical approach for characterizing complex mixtures. At Pacific Northwest Laboratory (PNL) supercritical fluid methods are being developed for materials where conventional gas or liquid chromatographic methods and existing mass spectrometric methods are inadequate (see references following this article). The advantages of supercritical fluid chromatography (SFC) accrue from the nature of supercritical fluids. The solvating properties of supercritical fluid mobile phases approach those of liquid phases, yet solute diffusivity typically remains one to two orders of magnitude higher. These properties allow high separation efficiencies to be realized while exploiting the variable and often high solvating power of the supercritical fluid. Consequently, SFC can separate thermally labile and nonvolatile solutes (a procedure not possible with gas chromatography) and can do so with higher efficiencies than liquid chromatography.

The introduction of the total chromatographic effluent into the mass spectrometer can be accomplished through the direct fluid injection (DFI) interface developed at PNL (Smith and Udseth 1983a). The coupling of capillary SFC with mass spectrometry (SFC-MS) offers significant advantages over existing high performance liquid chromatography-MS combinations. Supercritical fluids also offer advantages for transport of thermally labile compounds to the ion source since heating is

not required for volatilization. The DFI process provides for transfer of solutes in the supercritical fluid to the gas phase, so that conventional ionization methods can be used, while the matrix effects and uncertainties associated with desorption from surfaces can be avoided.

Three major accomplishments in the last year related to capillary SFC and SFC-MS include:

1. Nonextractable deactivated, bonded, and crosslinked stationary phases for very small diameter fused silica capillary columns (25-50 μm diameter) have been developed. Columns suitable for highly polar fluids (e.g., NH_3) are being developed.
2. The development of improved rapid pressure programming methods for capillary SFC-MS allows very high speed separations with enhanced sensitivity for mass spectrometric analysis.
3. The development of a DFI interface for mass spectrometry has provided improved transport of nonvolatile compounds to the ionization region.

The new high speed separation method is illustrated in Figure 1, which shows separations obtained for a normal alkane series in a short 1.5 m x 25 μm I.D. column at three different pressure ramp rates. This work serves to demonstrate the versatility of the capillary SFC approach. These methods and other related analytical methods are being further developed to allow extension to more polar and nonvolatile materials.

FIGURE 1. Comparison of High Speed Rapid Pressure Programmed Capillary SFC Separations of an n-Alkane Mixture

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• Lasers in Analytical Chemistry

Laser light has unique properties that can be applied to analytical chemistry to improve both sensitivity and selectivity. Pacific Northwest Laboratory is developing laser-based methods for measuring low levels of toxic materials and environmentally hazardous substances. The principal thrust over the last year has been development of two techniques: continuous wave double-resonance ionization mass spectrometry, and an associated technique called laser-enhanced electron impact ionization mass spectrometry. In both of these techniques, isotopically selective ionization coupled with mass spectrometry provides improved immunity against isobaric interferences and against effects from "tails" of neighboring isotopes. In another phase of our research, which involves laser analysis of nonvolatile molecular species, a supercritical fluid/laser/mass spectrometer system has been completed and initial data will be obtained soon.

CONTINUOUS WAVE DOUBLE-RESONANCE IONIZATION MASS SPECTROMETRY

T. J. Whitaker, B. D. Cannon, G. K. Gerke, and B. A. Bushaw

Continuous wave (cw) lasers offer two distinct advantages over pulsed lasers in analytical applications: they typically have narrower bandwidths by 3 to 4 orders of magnitude, and they have unity duty cycle. The first property permits isotopically selective excitation of elemental species. The second property can be exploited to improve the overall sensitivity in many types of analysis.

We are developing a technique that employs two cw dye lasers to sequentially excite an atomic species to a high electronic state. The excited atoms are then photoionized by a powerful infrared laser. Initial experiments on an atomic beam of Ba have achieved isotopic selectivities of about 800 in the ionization of ^{137}Ba over ^{138}Ba . However, frequency drifts in our dye lasers have prevented achieving better selectivity and have often made it difficult to consistently reproduce data. Therefore, a feedback system that holds the dye laser frequency constant to about one MHz has been developed. This improvement gives better selectivity because the laser frequency does not drift toward an interfering species. It also provides better sensitivity because the laser output remains on the exact peak of the desired species.

We have demonstrated that the cw laser ionization efficiency is almost 10 times greater than that of optimized electron impact ionization (Cannon, Bushaw and Whitaker 1985) and

we are in the process of incorporating a simple improvement that should increase that efficiency to near unity (i.e., almost every target atom in the laser beam will be ionized). This improvement involves exciting the atom to a higher electronic state and using a CO_2 laser for photoionization. Ionization efficiency is increased because of several factors: longer lifetime of the excited state, greater photon flux from the CO_2 laser, and a higher cross section for photoionization.

An interesting and useful phenomenon has been observed in these laser ionization experiments. Under certain conditions, peaks are split into two components such that the splitting is proportional to the laser intensity times the square root of the transition probability. We have used this phenomenon to determine the Einstein coefficient (transition probability) for emission from the barium $6s8s\ ^1S_0$ state to the $6s6p\ ^1P_1$ state. The value obtained ($2 \times 10^6\ \text{sec}^{-1}$) is subject to errors because of our laser intensity distribution. With our new ring dye laser, we can provide a more uniform intensity over the probed volume and thus obtain very accurate cross section information for many transitions. This should be useful data for any technique that involves optical transitions (atomic absorption, inductively coupled plasma, etc.).

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LASER-ENHANCED ELECTRON-IMPACT IONIZATION MASS SPECTROMETRY

T. J. Whitaker, B. A. Bushaw, G. K. Gerke,
and B. D. Cannon

We have developed an isotopically selective ionization technique that requires only one dye laser. This technique involves a standard electron-impact ionizer that is coupled to a quadrupole mass spectrometer. When the electron energy is set to a value much lower than the ionization potential of the analyte species, almost no ion signal is observed. If a laser is then used to excite the analyte species such that the ionization potential of the excited state is lower than the electron energy, a large ion signal is observed. This effect, reported briefly in the last annual report, has been developed to the point that excellent signal-to-noise is now achieved on a routine basis (Whitaker, Cannon and Bushaw 1984). The technique provides a simple mechanism to tune a laser to a transition (it was first developed for this purpose in our double resonance experiments), but can also serve as an analytical tool to provide extra selectivity in mass spectral analyses that do not require extreme sensitivity.

The relative peak heights of hyperfine components display an interesting behavior when observed by this technique (see Figure 1). Because an understanding of the signal strength is necessary to any analytical method, we have investigated and successfully explained observed peak height ratios in the $^{137}\text{Ba } ^1\text{P}_1 \leftrightarrow ^1\text{S}_0$ transition. The effect is caused by population redistribution in the degenerate magnetic substates of the lower state and is the topic of a forthcoming paper. This effect should be observed in any technique that permits several excitation/emission cycles to occur before the upper state is probed.

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FIGURE 1. Laser-Enhanced Electron Impact Ionization Spectrum of $^{137}\text{Ba } ^1\text{P}_1 \leftrightarrow ^1\text{S}_0$ Transition Using Different Polarizations. The ratio of the peaks also changes as a function of laser intensity.

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Physical
Processes

PHYSICAL PROCESSES IN RADIATION BIOLOGY

The Radiological Physics programs address the physical processes of radiation action on biological systems. Through integrated, multidisciplinary studies, the interaction of radiation with matter of biological significance is examined. The resulting information is integrated into predictive models of biological response.

Increased understanding of the physical, chemical, and biological mechanisms of radiation action results in a better understanding of dose-response relationships in terms of quantifiable characteristics of the radiation and the absorbing media. Experimental studies into the fundamental physical properties of energy deposition, energy transport, and cellular response are conducted to provide a data base for developing and testing chemical and biophysical models. Emphasis is on correlating radiation quality and energy transport with the formation of active chemical species; on understanding the mechanisms by which chemical agents interact at the cell level; and on developing models that correlate physical parameters of the radiation, chemical/biochemical interactions, and cellular repair phenomena with observed dose-response relationships.

• Radiation Physics

Initial Interaction Processes

A fundamental knowledge of the mechanisms by which radiation interacts with matter is essential to understanding dose-response relationships in terms of quantifiable chemical and physical processes. Our study of the initial interaction processes in the absorption of energy from a radiation field has concentrated on measuring and interpreting absolute cross sections for the interaction of fast charged particles with atomic and molecular targets. Since ionization is the primary means of energy deposition by fast charged particles, we have emphasized measurements of total, differential, and partial ionization cross sections. Cross sections for the production of electrons differential in ejected electron energy and emission angle are required for detailed track structure information, whereas partial ionization cross sections provide information on the final states of the target atom/molecule. Of particular interest is the determination of cross section systematics and interaction mechanisms that enable extrapolation of experimental results into regions where experiments are either impractical or unfeasible. During the past year emphasis has been on the study of doubly differential cross sections for ions carrying bound electrons. Coincidence techniques were developed for separation of target and projectile ionization, and analysis of data for C^+ impact continued. Interaction channels leading to multiple ionization of target atoms have been investigated and total cross sections for ionization of water vapor by protons and numerous gases by He ions have been published. This work benefits from a close working relationship with Professor M. E. Rudd, University of Nebraska, and Professor S. T. Manson, Georgia State University.

DOUBLY DIFFERENTIAL IONIZATION CROSS SECTIONS FOR CARBON IONS

L. H. Toburen and R. D. DuBois

Although extensive data exist regarding the details of ionization in collisions of bare ions with atomic and molecular targets, there have been few studies with heavy ions that carry bound electrons. The interactions in such collisions are modified because these bound electrons screen the ion nucleus. This electronic screening results in a greatly modified secondary electron spectra from ionizing collisions (Toburen 1982). Such spectra cannot be predicted with present theoretical techniques. Our interest in the local energy deposition distributions and track structure of charged particles produced in the absorption of energy from neutrons in tissue has prompted our studies of C, O, and N ions. During the past year we have continued to look at the systematics of C^+ impact ionization of several gas targets. The emphasis has been to determine the relevance of recent theoretical work and to compare our data within this theoretical framework with recent data for ionization by neon ions. A preliminary analysis of these data was presented at the 14th International Conference on the Physics of Electronic and

Atomic Collisions held at Stanford University in July, 1985 (Toburen 1985).

According to Woerlee et al. (1981), electron spectra resulting from low energy heavy ion-atom collisions should display an exponential dependence on v_1^{-1} , with v_1 being the ion velocity. Their data for neon ions also indicated a weak dependence on charge state of the ion. Taking this weak dependence on charge state as an indication that the spectra were not critically sensitive to the quasi-molecular structure formed during the collision, we compared our carbon ion data to their neon ion data. Figure 1 shows our data for C^+ ionization of neon at 0.8, 1.2 and 2.4 MeV compared with their lower energy data for Ne^{2+} impact. The similarities and systematics are encouraging from the standpoint of modeling for track structure application.

To further test the scaling model of Woerlee et al. (1981) one can plot the data as a function of v_1^{-1} . This is illustrated in Figure 2 where we have plotted our data along with $Ne^+ - Ne$ data of Cacak and Jorgensen (1970). From the scaling model one would expect a linear relationship for a given ejected electron energy as a function of v_1^{-1} . This is observed to be true for electron energies above about 100 eV. At lower

FIGURE 1. Doubly Differential Cross Sections for Electron Emission in C^+ and Ne^{2+} Collisions with Neon Atoms. The neon ion data are from Woerlee et al. (1981).

energies the linear relationship breaks down. This may be attributed to the fact that the model is, in general, for low energy ions,

i.e., ion velocity slow compared to electron velocity. For the low energy ejected electrons and for ion energies used in our studies, the low energy condition breaks down. At the lowest electron energies the data from our studies are beginning to resemble what is expected in coulomb ionization rather than the molecular promotion model, i.e., the cross section is decreasing with increasing ion energy.

For our studies of neon, only three ion energies were used: 0.8, 1.2, and 2.4 MeV. Although a straight-line extrapolation of the neon ion results to higher energies fits these data reasonably well, there is an indication this agreement may be fortuitous. The methane target data shown in Figure 2 were taken at five different energies, thus giving a somewhat more detailed picture of the velocity dependence in our energy range. These data agree fairly well with the neon target and over the limited velocity range indicate a steeper slope than that consistent

FIGURE 2. Doubly Differential Cross Sections for Related Electron Emission Energies Plotted Versus v^{-1} , Where v is the Incident Ion Velocity. The Ne^+ data are from Cacak and Jorgensen (1970). Also shown are our data for 250 eV electron emission in $C^+ - CH_4$ collisions.

with the lower energy Ne^+ data. We are therefore left with two alternatives: either there is structure in the velocity dependence or there is a relatively strong dependence on the quasi-molecular structure of the colliding systems leading to an inability to infer C^+ cross sections from extrapolated Ne^+ cross sections. Resolution of these questions awaits further data.

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STRUCTURED ION IMPACT: DOUBLY DIFFERENTIAL CROSS SECTIONS

R. D. DuBois

For structured ion impact, e.g., He^+ or H^0 , the projectile arrives at the collision region with an electron bound to it. During the collision either this electron can be stripped from the projectile or an electron can be ionized from the target to contribute to the total electron emission. Also, electrons can be liberated from both collision partners in a single collision.

Last year, we initiated a new set of experiments in which electron emission, as a function of ejected electron energy and angle, was measured in coincidence with a projectile ion that had lost a single electron. Initially He^+ - Ar collisions were investigated (DuBois 1984, 1985) but since then He (DuBois and Manson 1985), Ne, Kr and H_2O targets have also been studied. Results for He^+ - He are shown in Figure 1.

As can be seen, the cross sections are not only large for electrons having velocities near zero in the projectile frame (~ 200 eV in the laboratory frame) but also for electrons having low energies in the laboratory frame. These later electrons cannot originate from

FIGURE 1. Electron Emission at 20° in He^+ - He collisions. Total electron emission (solid curves); electrons detected in coincidence with stripped projectile (Δ , $\bullet$, Δ)

FIGURE 2. Calculated cross sections for electron emission at 20° for 2 MeV He⁺ - He collisions. Total electron emission (—); projectile ionization, target remaining in ground state (---); projectile ionization, target in any final state (---); projectile ionization, target in any final state plus target ionization, projectile excited (---).

the projectile since theoretically the cross section for electrons stripped from the projectile goes to zero at zero laboratory energy. Thus, these low energy electrons that are in coincidence with a stripped projectile appear to originate from the target being simultaneously ionized. Similar results were found to occur for all targets investigated thus far.

This interpretation is supported theoretically by an increase in the cross sections at low energies when simultaneous target and projectile ionization events are included (see Figure 2). However, quantitatively, the importance of this simultaneous ionization

channel is underestimated in the present theoretical calculation.

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MULTIPLE TARGET IONIZATION

R. D. DuBois, L. H. Toburen, and S. T. Manson

Our experimental total and doubly differential cross section measurements provide no information about the final target ionization state. However, measurements of multiple ionization of rare gases by H⁺ and He⁺ impact (DuBois, Toburen and Rudd 1984) showed that in some cases as much as 60% of the total ionization cross section was via multiple ionization.

Thus multiple ionization mechanisms need to be incorporated into any theoretical description in order to accurately describe the collision. However, direct multiple outer shell ionization, inner shell ionization leading to Auger relaxation, and charge transfer plus simultaneous ionization are all multiple ionization mechanisms. Radically different theoretical treatments are required for each of these different mechanisms and thus a knowledge of the relative importance of these cross sections is useful.

Progress in this field has been as follows:

1. All relevant channels leading to double ionization of neon by fast proton impact (500-4600 keV) were identified (Manson, DuBois and Toburen 1983). In this case direct double outer shell ionization was shown to dominate.
2. Other targets (He, Ne, Ar, Kr) have been studied at proton energies between 15 and 4000 keV. All channels leading to single and double ionization of He, Ne and Ar and also to triple ionization of Ne and Ar have been identified (DuBois, Toburen and Manson 1984). Insufficient information about the Auger channel for Kr prevents a detailed analysis.
3. Direct measurements of the charge transfer and direct ionization channels (DuBois 1984, 1985) have shown that at lower impact energies (15-200 keV) the total electron production is strongly influenced if not dominated by the charge transfer plus ionization channels. This is true for H^+ , He^+ and He^{2+} projectiles.
4. Very recently, multiple ionization by He^+ for larger impact energies has been investigated. In this case the multiple ionization contributions of the direct, charge transfer and electron loss channels have been measured. An example of the results is shown in Figure 1. An example of the importance of each channel to the total electron emission cross section is presented in Table 1. Clearly there are many channels that need to be considered in analyzing or modeling these collisions.

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FIGURE 1. Cross Sections for All Channels Leading to Electron and Ion Production in $He^+ - Ne$ collisions. Total ion production (σ_+); total electron production (σ_-); direct ionization of q target electrons (σ_q^{10}); charge transfer plus $(q-1)$ target ionizations (σ_q^{11}); projectile ionization plus q target ionizations (σ_q^{12}).

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TABLE 1. Channels for Electron Production in He⁺ - Ne Collisions

Channel ^(a)		At 50 keV % of total electrons	At 1000 keV % of total electrons
σ_1^{11}	He ⁺ + Ne → He ⁺ + Ne ⁺ + e ⁻	53	36
σ_2^{11}	→ He ⁺ + Ne ²⁺ + 2e ⁻	14	17
σ_3^{11}	→ He ⁺ + Ne ³⁺ + 3e ⁻	0.8	5.4
σ_1^{10}	→ He ⁰ + Ne ⁺	0	0
σ_2^{10}	→ He ⁰ + Ne ²⁺ + e ⁻	31	1.1
σ_3^{10}	→ He ⁰ + Ne ³⁺ + 2e ⁻	1.7	0.4
σ_0^{12}	He ⁺ + Ne → He ²⁺ + Ne + e ⁻	0	3.3
σ_1^{12}	→ He ²⁺ + Ne ⁺ + 2e ⁻	0	20
σ_2^{12}	→ He ²⁺ + Ne ²⁺ + 3e ⁻	0	17.5
σ_3^{12}	→ He ²⁺ + Ne ³⁺ + 4e ⁻	0	5.9

(a) (σ_q^{11}) = direct ionization of q target electrons

(σ_q^{10}) = charge transfer plus (q-1) target ionizations

(σ_q^{12}) = projectile ionization plus q target ionizations

TOTAL ION AND ELECTRON YIELDS

R. D. DuBois and M. E. Rudd

In 1981 a collaborative effort with Professor M. E. Rudd (University of Nebraska) was begun to measure total ion (σ_+) and electron (σ_-) yields in ion-atom/molecule collisions. The study emphasized absolute cross sections over a broad energy range in order to evaluate existing doubly differential cross sections as well as to provide accurate normalizations for track structure calculations.

Data for proton impact on ten atomic and molecular targets were published in December 1983 (Rudd et al. 1983). Data for proton impact on H₂O were published in January 1985

(Rudd et al. 1985a) and data for He⁺ impact on ten gases were published in August 1985 in Physical Review A (Rudd et al. 1985b).

For He⁺ impact total electron capture (σ^{10}) and loss (σ^{12}) cross sections were also measured, but only for impact energies less than 350 keV. These cross sections (σ_+ , σ^{10} , σ^{12}) are related via $\sigma_+ - \sigma_- = \sigma^{10} - \sigma^{12}$ and thus a self-consistent set of data was generated by least squares fitting the measured cross sections to the above equation.

Professor Rudd has also measured these cross sections for He²⁺ impact energies less than 700 keV. Future work will be limited to selected systems.

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Track Structure

The initial spatial pattern of energy deposition by ionizing radiation plays a significant role in the subsequent evolution of biologically active chemical species. Radiochemical and radiobiological models of radiation action initiated by high linear energy transfer (LET) particles rely heavily on an accurate description of charged particle tracks. Our study of track structure includes both theoretical and experimental investigation of the spatial pattern of energy deposition. Incorporation of these data into models of energy transport in condensed phase clarifies the effects of the initial spatial patterns on evolving chemical yields. Because of the experimental limitations on determination of energy deposition in nanometer-sized volumes, we continue to develop Monte Carlo codes to calculate ionization and energy imparted in small sites. During the past year we concentrated on investigating the systematics of energy deposition in very small sites. These data extend neutron transport codes by providing capability to calculate the distributions of energy deposited in small sites by arbitrary neutron fields. These codes also provide the initial conditions for formation and development of chemically active species in irradiated media. These stochastic radiation chemistry calculations have recently been applied to electron spin resonance (ESR) data which show anisotropy in radical yields in oriented DNA fiber irradiated from different orientations. This work benefits from close collaboration with H. G. Paretzke, GSF Neuherberg; D. E. Charlton, Concordia University; J. J. Coyne, National Bureau of Standards; and L. S. Myers and C. Swenberg, Armed Forces Radiobiological Research Institute.

IONIZATION PRODUCED IN NANOMETER SITES BY 0.3 to 20.0 MeV PROTONS*

W. E. Wilson and N. F. Metting

Last year we reported on a systematic computational study of the energy deposition and ionization yield produced in spherical sites of diameters 2 to 1000 nm by simulated proton tracks of 0.3 to 20 MeV (Wilson et al. 1985). The preliminary indications were that the ionization distributions could be adequately described by a lognormal distribution,

$$f(x) = \frac{1}{x\sigma\sqrt{2\pi}} \exp - \frac{(\ln x - \mu)^2}{2\sigma^2} \quad (1)$$

where x is the integer number of ion pairs produced inside the site, and μ and σ are the two parameters respectively characterizing the centroid and width of the distributions.

In the past year, we have continued to analyze these data by fitting Equation (1), in the least-squares sense, to each calculated ionization distribution and have thereby obtained μ and σ as functions of site diameter, proton energy and ion path eccentricity. The functional dependence of μ on site diameter and ion energy for ion paths passing diametrically through the site (eccentricity equal to zero) is relatively

simple (Figures 1 and 2). As these figures indicate, μ can be represented by a flat-plane in 3-dimensions, μ versus log diameter versus log ion energy. Other eccentricities are similar, having the same slopes but different intercepts on the μ -axis. All of this can be summarized mathematically by writing

$$\mu = c_1 \log(d) + c_2 \log(E) + c_3(b) \quad (2)$$

FIGURE 1. The Lognormal Mean or Centroid Parameter, μ , Plotted Against the Log (Base 10) of the Site Diameter in Nanometers. The integer symbols indicate the respective ion energy, from left to right, or top to bottom, 0 = 0.3, $\bullet$ = 1.0, Δ = 2.0, $\blacktriangle$ = 5.0, $\square$ = 10., $\blacksquare$ = 20. MeV. The solid lines are least squares fits of first order polynomials to the symbols.

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FIGURE 2. The Lognormal Mean or Centroid Parameter Plotted Against the Log (Base 10) of the Ion Energy in MeV. The integer symbols indicate the respective site diameter, from top to bottom, $\circ = 500.$, $\bullet = 200.$, $\Delta = 100.$, $\triangle = 50.$, $\square = 20.$, $\blacksquare = 10.$, $\nabla = 5.0$ and $\blacktriangledown = 2.0$ nm diameter. The solid lines are least squares fits of first order polynomials to the symbols.

where c_1 and c_2 are constants independent of eccentricity, b , and c_3 depends only on eccentricity, i.e., is independent of site diameter and ion energy.

The behavior of the width parameter, σ , is not as simple as that for μ . Its dependence on diameter and ion energy is shown in Figures 3 and 4. The solid lines are quadratic polynomials fitted to the (calculated) values of σ . No simple systematic behavior is apparent. We are currently exploring, theoretically and computationally, other functional forms for σ in an effort to discover how the parameter may be concisely expressed. To date, the phenomenological modeling of the ionization data has been for ion "crossers" that pass through the site. We are now calculating distributions for energy deposition and ionization for ion paths passing outside the site. Preliminary results are reported elsewhere in this report.

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FIGURE 3. The Lognormal Width Parameter, Plotted Against the Log (Base 10) of the Site Diameter in nm. The integer symbols are as in Figure 1, the solid lines are least squares fits of second order polynomials to the symbols.

FIGURE 4. Similar to Figure 3, But Plotted Against the Log (Base 10) of Ion Energy, Symbols are as in Figure 2. Solid lines are second order polynomial fits to the symbols.

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IONIZATION AND ENERGY DEPOSITION DISTRIBUTIONS FOR "PASSERS"*

N. F. Metting and W. E. Wilson

In the microdosimetry of neutron radiation fields four classes of charged particle tracks have been previously considered (Caswell 1966) in calculating energy imparted to a particular volume of interest. "Insiders" are particles that originate in the volume and lose all of their energy within the volume; "starters" originate in the volume but leave with part of their energy; "stoppers" originate outside the volume but stop within the volume; and "crossers" originate outside, enter and deposit some energy before exiting with the remainder. In the microdosimetry of very small sites (less than 1 μm dia), a fifth class, "passers," becomes important and must be considered. "Passers" are those particles that pass entirely outside the volume of interest but which deposit energy within the site via secondary electron (delta ray) transport.

Preliminary calculations and scoring have been done using an improved algorithm that better describes this "passer" type of energy deposition. Single event distributions have been calculated for primary ion energy 0.3 to 20.0 MeV, site diameter 2 to 1000 nm, and relative eccentricity from 0.0 to 1.2. Representative distributions are shown in Figures 1 through 3. Figure 1 shows ionization distributions for several primary ion energies in a 100-nm diameter volume with a relative eccentricity of 1.0 (tangential tracks). Figure 2 shows distributions for several site diameters, each setting at a relative eccentricity of 1.0 from the path of a 1 MeV proton. Figure 3 shows ionization frequencies in a 100-nm diameter site by a 1 MeV proton for a series of different relative eccentricities. Studying the shapes of these distributions will aid in developing a phenomenological model of track structure that includes energy transport into a site by delta rays.

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FIGURE 1. Ionization Number Frequencies for Several Proton Energies. Site diameter (100 nm) and relative eccentricity, B (1.0) held constant.

FIGURE 2. Ionization Number Frequencies for Several Site Diameters. Primary ion energy (1.0 MeV) and relative eccentricity, B (1.0) held constant.

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FIGURE 3. Ionization Number Frequencies for Several Relative Eccentricities. Primary ion energy (1.0 MeV) and site diameter (100 nm) held constant.

MONTE CARLO CALCULATION OF ENERGY DEPOSITION AND IONIZATION YIELD FOR HIGH ENERGY PROTONS*

W. E. Wilson, J. C. McDonald, J. J. Coyne,** and H. G. Paretzke***

Recent calculations of event-size spectra for neutrons use a continuous slowing down approximation model for the energy losses experienced by secondary charged particles (protons and alphas) and thus do not allow for straggling effects (Caswell et al. 1981; Coyne et al. 1981). Discrepancies between the calculations and experimental measurements are thought to be, in part, due to the neglect of straggling. The Monte Carlo method is a tractable way of including stochastics in radiation transport calculations and a number of efforts directed toward simulating positive ion track structure have been initiated employing this technique. Recent results obtained with our updated and extended MOCA code for charged particle track structure (Wilson et al. 1981) are presented here. Major emphasis has been on calculating

energy deposition and ionization yield spectra for recoil proton crossers since they are the most prevalent event type at high energies (>99% at 14 MeV) for small volumes. Neutron event-size spectra can be obtained from them by numerical summing and folding techniques. Data for ionization yield spectra are presented for simulated recoil protons up to 20 MeV in sites of diameters 2-1000 nm.*

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ENERGY TRANSPORT: HIGH LET RADIATION CHEMISTRY OF NUCLEIC ACIDS

J. H. Miller, W. E. Wilson, and D. E. Charlton**

The main emphasis of our previous energy transport studies has been to model production and decay of free radicals in pure water or dilute aqueous solutions. While this work continues, we have in FY 1985 initiated a new

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effort directed toward a better understanding of the radiation chemistry of concentrated solutions of DNA exposed to high linear energy transfer (LET) radiation. In these systems one must consider the consequences of energy absorbed in DNA as well as indirect damage induced by water radicals. Efforts to understand the biological consequences of direct action have led to the development of computer codes to analyze geometric aspects of energy deposited in DNA and chromatin structures (Goodhead et al. 1985). Recent discussions with Larry Myers and Charles Swenberg at the Armed Forces Radiobiology Research Institute (AFRRI) have suggested potential applications of these codes in understanding the high LET radiation chemistry of nucleic acids. This latter application is the subject of this report.

Experiments performed at AFRRI with oriented DNA fibers exposed to neutrons show the novel effect that the yield of the 5,6-dihydro-5-thymyl ($\dot{T}H$) radical is highly dependent on the relative orientation of the fibers and the neutron flux. When neutrons are incident perpendicular to the DNA fibers, the ESR spectrum of radicals formed at 77°K can be explained by the roughly equal amounts of quanine cation and thymine anion ($\dot{T}^-$). When samples are irradiated with neutrons parallel to the DNA fibers, the yield of $\dot{T}^-$ is reduced and a strong $\dot{T}H$ signal is seen.

Samples irradiated with γ -rays and assayed at 77°K do not exhibit the $\dot{T}H$ signal. However, when the irradiated samples are annealed at 200°K, $\dot{T}H$ radicals appear with bi-phasic kinetics. Gräslund et al. (1975) showed that these kinetics could be explained by thermally activated protonation with water of hydration being the most likely proton source. The kinetic data were consistent with activation energies in the range of 5-15 kcal/mole.

Radiation-induced thermal spikes have frequently been used to explain the yield of activated processes (Vineyard 1976). Isolated energy transfers dissipate too rapidly to be effective in driving reactions with activation energies in the range of 5-15 kcal/mole; however high LET radiation may operate through this mechanism in some cases (Mozumder 1969). If we postulate a large anisotropy in the thermal conductivity

of samples of oriented DNA fibers, then thermally activated proton transfer may explain the orientation dependence of $\dot{T}H$ yields. In the geometry of perpendicular irradiation, the probability of multiple energy transfers along a single fiber is low. Hence, assuming low thermal conductivity between fibers, energy transfers in the track of a recoil proton are isolated and ineffective in producing $\dot{T}H$. However, if the proton track is oriented parallel to the DNA fiber, multiple energy transfers to a single fiber can occur.

To quantitate this effect, we have scored energy deposition in cylindrical targets oriented parallel or perpendicular to a 1 MeV proton track segment in water. Figure 1 shows that in a target with a length of 50 nm and a diameter of 5 nm, approximately 10% of the energy transfers exceed 500 eV when the axis of the cylinder is parallel to the track, and less than 1% have this magnitude

FIGURE 1. The Fraction of Events in Which Energy Greater Than a Specified Amount is Transferred from a 1 MeV Proton to a Cylindrical Water Target When the Target is Parallel or Perpendicular to the Direction of the Proton. The target was 5 nm in diameter and 50 nm long.

when the symmetry axis of the target is perpendicular to the track. This asymmetry in the distribution of event sizes does not increase with further increase in the aspect ratio of target length to diameter, and vanishes for targets with equal length and diameter. Since these characteristics of energy transferred from recoil protons to oriented DNA fibers are dependent on proton energy, comparison of our predicted yields with data obtained from neutron exposure must involve an average over the spectrum of recoil proton energies. These calculations are not complete. Hence, in this report we will show only how the yield of $\dot{H}$ radicals is related to the geometry of energy deposition events.

The yield of $\dot{H}$ resulting from transfer of energy ϵ to a length Δ of DNA fiber is given by

$$Y(\epsilon, \Delta) = g \frac{\epsilon}{\Delta} \int_{-\infty}^{+\infty} dx \int_0^{\infty} dt k e^{-kt} \quad (1)$$

where g is the initial yield of $\dot{I}^-$ precursor per unit of energy absorbed. The rate of decay through proton transfer is

$$k(x, t) = A \exp(-Q/T(x, t)) \quad (2)$$

where A is a constant proportional to concentration of proton donor, Q is a constant proportional to the activation energy, and T is the absolute temperature of the thermal spike. To obtain an order of magnitude estimate of the yield of $\dot{H}$ we assume that the thermal spike is a small perturbation on the precursor population and that the specific heat and thermal conductivity are temperature independent. These simplifying approximations need not be made in more detailed calculations; however, under these assumptions Equation (1) reduces to

$$Y(\epsilon, \Delta) = g \frac{\epsilon A \Delta^2}{D} \sqrt{2} f(T_0/Q) \quad (3)$$

where D is the thermal conductivity and f is a function of the initial temperature of the spike T_0 that must be evaluated numerically.

Irrespective of the absolute magnitude of Y , Equation (3) shows how a thermal spike model may account for the orientation dependence of the yield of $\dot{H}$ radicals. In both parallel

and perpendicular irradiation geometries, one has events for which ϵ/Δ is of the same order of magnitude as the stopping power S of the recoil protons. Hence the initial excess temperature induced by the radiation, which is a function of ϵ/Δ , is comparable in the two geometries. However, for the perpendicular case, the width of the spike will be comparable to the diameter of the fiber, while in the parallel case, the width of an event for which $\epsilon/\Delta \sim S$ can be many times the fiber diameter. Since larger widths cause the excess temperature to persist for a longer time, the yield of $\dot{H}$ from a spike increases with the square of its width.

The relative yield of $\dot{H}$ radicals in the parallel and perpendicular irradiation geometries is given by the ratio of integrals of Equation (3) over ϵ and Δ , weighted by the probability to transfer energy ϵ to a length Δ in the respective geometries. These ratios are independent of the parameters g , A and D , which are not known accurately. Hence we expect the relative yields of $\dot{H}$ to be a sensitivity test of our ability to calculate spatial patterns of energy deposited by neutrons at the molecular level. If this proves to be true, these radiation chemistry experiments will be a valuable complement to biological data in testing neutron microdosimetry.

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• Radiation Dosimetry

Dosimetry

The primary goal underlying the Radiation Dosimetry task is to understand the connections between the physical events produced by the interaction of ionizing radiation with matter and their biological consequences. Dosimetry concentrates on the physical aspects of the problem, utilizing fundamental physical data to describe the interactions and organizing the results in ways that can be correlated with the biological effects. Special mathematical and experimental techniques are being developed to explore the differences in the energy deposition characteristics of various radiations in an effort to understand the effects of these radiations in radiobiology research and the environment. During the past year the physical data from high energy heavy charged particle experiments have been analyzed to put the energy deposition by delta rays in perspective with other low LET radiation sources. The experience gained from a wide range of dosimetric studies has been applied to problems of risk assessment and dose evaluation through participation in national and international committee activities.

EXPERIMENTALLY DERIVED VALUES OF $\bar{y}_F$ AND $\bar{y}_D$ FOR 33.6 GeV IRON AS A FUNCTION OF ECCENTRICITY

N. F. Metting and L. A. Braby

The use of coincidence multiparameter data acquisition in the measurement of energy deposition around 33.6 GeV iron ion tracks (Metting et al. 1985) has made it possible to analyze track structure by comparing mean lineal energy values at measured radial distances (eccentricities). To relate the secondary electron radiation field to other types of low LET radiations (photons and electrons), extensive data files were sifted to find events occurring at specific distances radiating outward from the path of the primary ion. Figure 1 shows the $yf(y)$ distribution for the entire uniform radiation field. From this distribution, we can calculate that 61% of the energy events result from delta rays alone and 39% are high energy, primary ion events.

Figure 2 shows examples of single event $yf(y)$ distributions for specified distances from an ion's path, which were added together to construct the uniform field distribution shown in Figure 1. Notice the first distribution, labeled 0-0.5 μm , includes primary ions crossing the site and therefore has the high energy, primary ion peak represented. These distributions were normalized to unit area, so that details of the curves could be seen. The average value of each distribution, $\bar{y}_F$, shown in the bar chart, decreases steadily

FIGURE 1. Density of y for the Uniform Field. By integrating over the area of each peak, it is seen that 61% of the energy events result from delta rays alone, and 39% are primary ion events.

for about 6 μm as one travels radially outward. Beyond that distance this value remains fairly constant. The decrease in $\bar{y}_F$ reflects the decreasing probability of encountering more than one secondary electron per primary ion crossing a site. At distances over 6 μm either one electron or none is crossing the site and the mean energy per crossing is constant.

In Figure 3 data from Figure 2 have been modified to reflect the probability per primary of an event occurring in the site. This shows graphically the frequencies of events as a function of radial distance. The bar chart in this figure represents the frequency of energy deposition events per

FIGURE 2. Distributions of $yf(y)$ as a Function of Eccentricity. The bar chart on the right hand shows the mean value $\bar{y}_F$ for each distribution.

FIGURE 3. The Distributions from Figure 2 Modified to Reflect the Relative Probability of an Event Occurring in the Site. The bar chart shows the frequency of energy deposition events per primary ion.

primary ion, again as a function of radial distance from the primary, and shows the expected $1/r^2$ decrease.

Dose distributions show the same type of progressions. Figure 4(a) shows the $yd(y)$ distribution for the uniform field, and shows

FIGURE 4. a) Probability Density of Absorbed Dose in y for the Uniform Field. Note that only 4% of the dose is delivered by secondaries. b) Mean Values, $\bar{y}_D$, as a Function of Eccentricity.

that 96% of the dose delivered is due to primary ion interactions and only 4% is due to secondaries. Figure 4(b) presents the expectation value $\bar{y}_D$ as a function of radial distance, and exhibits the same characteristics as those of the $\bar{y}_F$ values shown in Figure 2. For those sites not crossed by the primary, $\bar{y}_D$ is 4 keV/μm, which is approximately the same as has been observed for 200 to 250 Kvp x-rays (ICRU 1983). This is

also about twice the energy deposition per event that occurs with ^{60}Co irradiation. Thus, of the $1\text{ }\mu\text{m}$ diameter sites irradiated by 33.6 GeV iron ions, about 60% receive doses equivalent to that delivered by an x-ray irradiation of 80 rads or less.

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RE-EVALUATION OF THE HIROSHIMA-NAGASAKI RADIATION DOSES

W. C. Roesch

The National Academy of Sciences Panel on Reassessment of Atomic Bomb Dosimetry met on October 19, 1984, and April 2, 1985; the Department of Energy Working Group, which is the group conducting the U.S. research, met on October 17-18, 1984, and July 30, 1985.

The culmination of these working meetings was the Third U.S.-Japan Joint Workshop for Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, which was held in Pasadena, California, on March 12-14, 1985. At this meeting eight small working groups made up of both U.S. and Japanese scientists reviewed the reassessment program, decided what was adequately known and where further research was required, and established a schedule to complete the work by late this year. Another joint meeting was proposed for March 16-17, 1986, at Hiroshima, where representatives of the two nations would accept the reassessment of the radiation doses. Some long-range studies, affecting a relatively small part of the reassessment, will have to continue for another year or two.

BETA-PARTICLE DOSIMETRY

W. C. Roesch

The NCRP's (National Council on Radiation Protection and Measurements) Scientific Committee No. 80 on Radiobiology of the Skin held working meetings on November 15-16, 1984, and February 21-22, June 12-13, and August 28-29, 1985. As chairman, W. C. Roesch represented the committee at the annual meeting of the NCRP on April 3-4, 1985.

Within the guidelines given it by the NCRP, the committee decided to prepare recommendations concerning: 1) the depth(s) within the skin at which dose measurements shall be made, 2) the range of depths over which the dose can be averaged, 3) the area of skin over which the dose can be averaged, and 4) what measurements are required to protect the whole skin. The recommendations will apply to all radiations, not just to beta particles. How the measurements are to be made will be left to a later committee. The committee will not recommend permissible doses for the skin.

The committee, which expects to finish its work next year, has prepared a draft manuscript reviewing what is known about the radiobiology of the skin that bears on these topics and is beginning to prepare recommendations.

Representing the committee, W. C. Roesch presented an invited paper, "Problems Related to the Critical Depth of Skin," at the Workshop on Dosimetry of Beta Particles and Low Energy X-Rays on October 7-9, 1985, in Saclay, France.

STOCHASTICS IN TIME

W. C. Roesch

Last year a theory of ionization fluctuations was developed for use in interpreting some radiation measurements made in Hiroshima and Nagasaki (Roesch 1985a). This work has now been enlarged into a new topic in microdosimetry. In microdosimetry one has hitherto considered a particular small mass that is exposed repeatedly, for equal periods, to the

same radiation. The object was to determine the probability distribution of the amounts of energy deposited in the mass by the radiation. One can instead consider trying to irradiate the same small mass to the same total energy a large number of times and determining the probability density in the lengths of the irradiation periods required. This approach shifts the emphasis to the time required to produce an effect, and the techniques employed may be useful in studying effects that depend on dose rate or fractionation. A description of the techniques was given at the Radiation Research Society annual meeting (Roesch 1985b).

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SYSTEMS FOR EXPLORING THE SPATIAL AND TEMPORAL DISTRIBUTIONS OF TARGETS WITHIN CELLS

L. A. Braby and W. D. Reece

Most of our knowledge of the spatial distribution of radiation sensitive materials within cells and of the times and distances over which damaged biological materials interact has been gained through experiments using charged particle tracks distributed randomly in time and space. However, because of this physical randomness combined with the variations between individual cells and the random nature of many of the damage and repair processes, there are usually alternative interpretations of the result of any experiment. One way to reduce the range of alternative interpretations and also to gain completely new information is to irradiate cells with charged particle tracks at speci-

fied positions and times. The most practical method is probably the use of a collimator. Zirkle (1957) used this approach with alpha particle sources to demonstrate the major role the cell nucleus plays in radiation sensitivity. However, his finest collimator diameter was $\sim 2 \mu\text{m}$, and only a relatively small number of cells could be irradiated, so the complex problems of interaction time and distribution of sensitive material within the nucleus could not be addressed.

Modern solid state component manufacturing requires production of very fine details in metals and insulators. Techniques have been developed to produce holes and slits as small as $0.25 \mu\text{m}$. Thus slit edge scattering, rather than the production of the collimator, now limits the resolution of microbeam experiments. We have been investigating the feasibility of an apparatus to irradiate cells with approximately $0.5 \mu\text{m}$ spatial resolution and temporal resolution of less than one second. A fluorescent coating on the thin mylar dish bottom supporting the cells would be used to detect the time of passage of a single charged particle. The position would be defined by a collimator assembly designed to reduce the effects of scatter. A phase contrast optical system and micro positioning stage would be used to position the desired cellular organelles relative to the beam. To be useful in biological studies, this irradiation facility must be able to process relatively large numbers of cells from a single culture to avoid the differences between cells grown under slightly different conditions. Beam characteristics and irradiation times present no problem in this respect, but the positioning of the cells will be the limiting factor. To speed this process, we expect to use a computer-controlled stage and automated video recognition system capable of locating the nuclear membrane and other cellular features. We are using discretionary funds to explore the details of each component of this system, and expect to be able to design a suitable beam line in the near future.

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Modeling and Cellular Studies

The effects of ionizing radiation on living systems begin with the physical processes of energy deposition and develop through many stages of chemical reactions and biological response. The modeling effort attempts to organize the available data and theories on all of these stages into self-consistent models that can be compared and tested. Models that are found to be consistent with data can be used in risk evaluation in the range of dose and dose rate where they have been tested, but their primary use comes in developing an understanding of the chemical and biological processes involved. Data on physical parameters of the radiation field, the physiological condition of the cells and the timing of the radiation exposures are used to develop models that incorporate different assumptions about the mechanisms of cellular response. These models are then tested using experiments for which different models predict observably different responses. If one model can be shown to be inconsistent with the results of the experiments then it can be used to identify classes of processes that are not relevant to the observed end effect.

Mathematical modeling and cellular studies complement each other. Modeling provides guidance in the design of experiments capable of disproving the model, while the experiments often suggest new features to be included in the model. Previous studies of the production and repair of damage in cultured Chinese hamster ovary (CHO) cells have shown that two types of repair are functioning. We have now been able to utilize these data to show that certain features are necessary for a model to be consistent with the data and to begin to relate the types of repair processes to the conditions under which the damage was produced.

RADIOBIOLOGICAL MODELING

W. C. Roesch

W. C. Roesch presented an invited paper, "Modeling Dose Rate and Fractionation Effects with Accumulation Models" (Roesch 1985), at a symposium on modeling at the annual meeting of the Radiation Research Society. For this symposium, each speaker was to apply his methods to a common set of data. The data chosen for this purpose were from Pacific Northwest Laboratory (Metting et al. 1985).

Most of the information presented discussed new applications of methods that we have described in other reports; however, three new topics were developed to answer questions pertinent to the symposium:

1. The proportion of cell killing that takes place after the irradiation stops was calculated for an accumulation model. The proportion depends on the dose rate; at low dose rates, most of the killing takes place during the irradiation; at high dose rates, most takes place afterwards.
2. The accumulation model we use for mammalian cells starts with an infinite

supply of progenitors, i.e., not with a pool. Simple non-accumulation models usually start with damage in a pool. A non-accumulation model that does not start from a pool was developed to provide more meaningful comparisons between the two kinds of models.

3. The accumulation model is generally used for split-dose experiments that have only two doses. However, since many doses are usually used in radiotherapy and in related experiments, the accumulation model was worked out for any number of equal doses with equal interfraction times.

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DAMAGE INTERACTION ESSENTIAL IN RADIATION EFFECTS MODELS

L. A. Braby, J. M. Nelson, and N. F. Metting

Biophysical models of radiation damage and repair are continuing to evolve in response to experimental results. A very fundamental difference in the initial assumptions used by different modelers deals with the expected consequence of a typical initial product. It is generally recognized that there must be many different types of "initial products" formed by ionizing radiation, but for the purpose of modeling, one of them is often assumed to dominate in the production of the observed end point and the others are ignored. The fundamental difference is whether the initial damage is inherently lethal unless repaired, or is not lethal unless additional damage interacts with it to make it capable of inactivating the cell. This question cannot yet be answered completely, but recent experiments show that some form of interaction must be involved. Thus, models which assume that the shoulder of the survival curve results from a limited repair capacity or from radiation-induced damage to the repair system can be ruled out, and we can concentrate on models and mechanisms that involve a dose-dependent interaction, possibly the interaction of sublethal products or misrepair of potentially lethal damage (PLD).

This conclusion is based on a comparison of the total amount of repair observed in split-dose and delayed-plating experiments. The predictions of various models can be visualized by plotting the amount of damage (lethal, potentially lethal, or sublethal) in the cell as a function of time. Figure 1 shows this for a typical PLD model. If a specified dose is given either as a single dose or as two equal fractions, the same number of lethal and potentially lethal lesions are produced. The probability of the cell surviving is determined by the sum of the probabilities of having a lethal or potentially lethal lesion remaining at some time (the fixation time) after transfer to growth medium. The split-dose experiment will always result in lower survival (less repair) than the delayed-plating experiment. Various modifications to this type of model, such as radiation-induced damage to the repair system or repair capacity increased by

FIGURE 1. Numbers of Lesions as a Function of Time Assuming Direct Production of Both Lethal and Potentially Lethal Lesions, and Exponential Repair of the Potentially Lethal Ones. Probability of reproductive death is proportional to the number of lesions remaining at the fixation time.

radiation, cannot reverse this result since all of the damage is exposed to whatever repair systems exist for the maximum time in the delayed-plating experiment. However, modifications which convert damage from potentially lethal to lethal by a process that depends on the square of the concentration of damage can result in less repair in the delayed-plating experiment since the initial high concentration of damage results in more misrepair. This makes the model very similar to sublethal damage models such as the simple one illustrated in Figure 2. More complete versions of sublethal damage models,

FIGURE 2. Number of Lesions Remaining Assuming a Sublethal Damage Model.

including separate production of potentially lethal products or repair of the product of the interaction of sublethal events, do not necessarily result in greater survival in the split-dose experiment, but can, depending on the rates of the various processes. Fortunately, the experimental results for plateau-phase CHO cells (Figure 3) show higher survival, the result of more repair in the split-dose experiments. If the experimental result had been the opposite, it would have been consistent with simple PLD models as well as concentration-dependent misrepair and sublethal damage models.

FIGURE 3. Observed Survival of Plateau-Phase CHO Cells Following Split-Dose and Delayed-Plating Treatments. If innocuous lesions were not being removed during the interval between fractions in split-dose experiments, the enhancement of survival observed after 4 Gray + 4 Gray (split-dose data, lower panel) would not be greater than that observed after 8 Gray (delayed-plating data, upper panel).

The differences between models which are consistent with the observed data have to do with the consequences of unrepaired damage that has not interacted with additional damage. One type of model assumes that unrepaired damage is lethal while the other assumes that it is innocuous. This difference will be quite significant when extrapolating to the effects expected at low doses. New types of experiments, based on the predictions of these two types of models, will be designed to further limit the range of assumptions that are consistent with data. An effort will be made to take advantage of radiation schedules and doses to obtain the needed information, but, if experimentally detectable differences at conventional survival levels (80% or less) cannot be found, techniques to accurately assess survival at the 95 to 99% level may have to be developed.

EFFECT OF OXYGEN ON DAMAGE PRODUCTION AND REPAIR

J. M. Nelson, L. A. Braby, W. C. Roesch, and N. F. Metting

We have continued investigating the relationship between the presence of oxygen and the production and removal of sublethal and potentially lethal damage. Last year, preliminary experiments indicated that one type of damage was greatly attenuated (or perhaps did not exist at all) in the absence of oxygen. We used starved plateau-phase Chinese hamster ovary (CHO) cells because these cells are well characterized. We looked at both sublethal (split-dose experiments) and potentially lethal damage (delayed-plating experiments) damage and repair, using doses to give roughly equivalent survival levels. Irradiations were made at 37°C, using 1.5 MeV electrons, and were given at dose rates of about 5 Gy/sec. Hypoxia was attained by gassing cultures prior to and during the irradiations with a mixture of 95% prepurified nitrogen plus 5% anaerobic-grade CO₂. Under these conditions, effluent oxygen concentration was typically <10 ppm after about 20 min of gassing.

Figure 1 shows the typical oxygenated and hypoxia irradiation survival curves; these data indicate a substantial OER, demonstrating that the degree of hypoxia is adequate to have eliminated most of the oxygen effect. Split-dose experiments were done under the same irradiation conditions; cells irradiated under hypoxia and euoxic conditions were both trypsinized and subcultured in air immediately after the second irradiation. No change in survival was observed for anoxic cells when the interval between doses was increased. However, their survival is much higher than that of cells irradiated with the same dose in air, even when they were allowed 6 hr for repair.

To determine if the sublethal damage repair required oxygen, i.e., if hypoxia was inhibiting split-dose repair, duplicate experiments were done in which other plates were anoxically irradiated and then held in air (euoxic environment) for all but the last 30 min of the interval between doses. Survival of these cells did not differ significantly from those maintained without oxygen throughout. This led us to conclude that

FIGURE 1. Survival of Plateau-Phase CHO Cells Irradiated in Air and Nitrogen.

the type of damage normally repaired in a split-dose experiment is not produced in the absence of oxygen.

We tested the effects of oxygen on potentially lethal damage removal in a similar manner. Hypoxic cultures were irradiated then either held for an additional period (up to 6 hr) under these conditions, or flushed immediately with air to reestablish the oxygen environment and held for up to 28 hr. Except for a limited amount of damage, probably due to toxic effects or metabolic stress resulting from the absence of oxygen and not related to radiation effects, there was essentially no difference in the repair capacity of cells held under oxygenated conditions and those held under moderately hypoxic conditions, as noted in Figure 2.

From these data we concluded that the type of damage that is repaired after the completion of irradiation, i.e., potentially lethal damage, does not require oxygen. Other investigations have shown that extreme hypoxia, below 10 ppm, might prevent repair of this type of damage, i.e., the repair process itself is oxygen-dependent. We conclude further that the potentially lethal damage that is repaired in hypoxic cells cannot be

FIGURE 2. Survival of Cells Irradiated in Nitrogen and Held in Air or Nitrogen Before Trypsinizing and Replating in Air.

the interaction product of sublethal damage events, since the sublethal damage does not appear to be produced during the hypoxia irradiations.

REPAIR RATES FOR DAMAGE PRODUCED BY HIGH LET RADIATIONS

L. A. Braby and B. S. Jacobson*

The study of the repair of damage produced in *Chlamydomonas* by high LET radiations has continued. The object was to reduce the uncertainty in the data for ³He ions. To this end, a new radio frequency ion source was added to the tandem accelerator, and other small refinements were made in the irradiation facility. However, it appears that some unrecognized change in the experimental procedure has occurred; the uncertainty in survival following a split dose was higher than in the previous series of experiments. We have tested a variety of possible problems, including the timing relative to the cell cycle, light level before and after irradiation, and possible malfunctions in the dosimetry. None of these have proved to be the problem. The next approach is to return as nearly as possible to the previous procedures, and then make small changes, checking the effect on scatter in the data as we go.

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• Radiation Biophysics

Experimental studies in cellular radiobiology are undertaken to provide comprehensive tests of theoretical models that relate biological phenomena with their initiating physical events. These studies are closely coordinated with the models being developed in the Radiation Dosimetry task, giving laboratory support and guidance to the development of the theory. Studies involving synchronized mammalian cell cultures and various irradiation parameters such as dose rate, dose fractionation, and radiation quality are designed to provide optimum tests of the theory. Cells with constant radiosensitivity are required for dose fractionation and dose-rate studies over long time intervals (up to two days). Stationary-phase cultures must be used to provide such a characteristic when mammalian cells are used. The study of stationary cell culture characteristics has continued with the result that the effects of irradiation can be interpreted more clearly. Experiments designed to test the shape of survival curves at low doses and thus to discriminate between relatively simple models for extrapolating to the effects of low doses have shown that a more complex model may be required.

FLOW CYTOMETRIC STUDIES ON PLATEAU-PHASE CHINESE HAMSTER OVARY CELLS

J. M. Nelson, P. W. Todd,* and N. F. Metting

Last year we reported finding significant differences between different types of non-expanding Chinese hamster ovary cell populations. These results were based on classical cellular kinetic measurements of cells entering and exiting plateau phase (Nelson, Todd and Metting 1984). We have followed this by initiating flow cytometric (FCM) studies on the same types of cell populations. These studies are intended 1) to verify the earlier findings and 2) to determine the mechanism by which cells are lost from the population, particularly in the fed cultures. The fed-plateau phase is a dynamic equilibrium between cell production and cell loss, maintained in part by continuous but intermittent feeding regimens.

Analysis of FCM measurements of cells re-entering the growth cycle verify all of our earlier findings from both the fed and starved cell populations. A manuscript entitled "Reentry Into the Cell Cycle by Fed and Starved Plateau-Phase Chinese Hamster Ovary Populations; A Flow Cytometric Study," which summarizes these FCM studies and our interpretations of the findings, has been prepared and submitted. A second manuscript

dealing with cell growth into plateau phase under fed and starved conditions, entitled "Growth Activity in Fed and Starved Plateau-Phase Cultures of Chinese Hamster Ovary Cells; A Flow Cytometric Study," is in preparation.

In these papers we report that FCM data also demonstrate an accumulation of nonprogressing cells in both G1 and G2 phases. Additionally, these data substantiate the existence of nonsynthesizing cells having S-phase DNA content. These cells do not incorporate ^3H thymidine, as identified by autoradiographic techniques (Nelson, Todd and Metting 1984). They also do not increase their DNA content, a change which would lead to an accumulation of pre-restriction S-phase cells and would be observed as a relative change in the DNA histogram, i.e., a shift to the right, in the region between the G1 and G2 phase peaks. Since this does not happen, we conclude that these cells are not synthesizing DNA, even though we could identify them as S-phase cells by their DNA content.

We also demonstrated that the fed populations ultimately lead to DNA-deficient cells and concurrently to the production of Feulgen-positive debris. Apparently, the incapacitated or defective cells lyse, producing karyorrhectic debris that is then utilized by another growing population. There is, of course, a concomitant cellular production occurring in order to maintain the plateau equilibrium.

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DISTINGUISHING ALTERNATIVE ACCUMULATION MODELS

J. M. Nelson, L. A. Braby, and W. C. Roesch

As discussed in a previous section, we have shown that the data for the survival of plateau-phase Chinese hamster ovary cells are consistent only with models that incorporate a process that depends on the square of the concentration of damage. This is consistent with the accumulation of sublethal damage or certain kinds of misrepair processes. These models may still differ substantially with respect to the consequences of unrepaired damage. The sublethal damage models such as that developed by Roesch (1978, 1985) assume that damage that has not interacted with other damage is innocuous, while misrepair models such as that developed by Curtis (1983) assume that unrepaired damage will be lethal. This difference can result in significantly different dose response relationships at low doses. One way to investigate this behavior at low doses is to evaluate the factor related to the probability of pairs of events producing the effect (usually called A or β).

We have found that it is generally more efficient to use split-dose experiments than single-dose experiments to evaluate A at low doses. When these data are plotted as $\ln [S_m/S]$ versus interval between doses, the slope of the line is determined by the repair rate and the intercept at zero interval is AD^2 . When two or more types of damage with separate repair processes are involved, they can be separated by extrapolating the line due to the slowest process and subtracting it from the data at short intervals. Figure 1 shows the data from a single experiment in which both 2 and 4 Gray per fraction exposures were used. These results are consistent with those of a number of other experiments. The intercept for the slow process increased by a factor of 2.76 when the dose

FIGURE 1. The Change in $\ln(S_m/S)$ with Interval Between Doses Clearly Shows the Effect of the Repair Processes Acting on Different Types of Damage. Furthermore, the intercepts at 0 interval suggest no simple relationship between the production of sublethal damage and cellular survival.

increased by a factor of 2, while the intercept for the fast process increased by 1.66.

These results suggest that interaction of sublethal damage or concentration-dependent misrepair are not the only processes involved in determining the shape of the survival curve at low doses. Further modeling and additional experiments aimed at reducing the uncertainty in these results are needed before firm conclusions can be reached.

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CHROMOSOMAL DAMAGE AS AN END POINT OF RADIATION DAMAGE

J. H. Miller, J. M. Nelson, and L. A. Braby

Most of the work to investigate the kinetics of radiation damage and repair processes has dealt with reproductive death or mutation as the end point. This has been a useful approach, yielding much information about the nature of processes that damage cells and those that repair the damage. However, these end points are the result of a long sequence of processes that constitute the cell's response to the initial damage. If we are to eventually understand this sequence of processes, we must investigate some intermediate points.

We are interested primarily in damage to the nuclear DNA (for low-dose long-term effects involved in risk assessment) so we do not have to study all of the cell's functions, but the task becomes difficult because very small changes (possibly one base among hundreds of thousands per cell) may be responsible for the effect. Thus, to detect the damage, we normally have to rely on the amplification resulting from the growth of a single cell into a clone. Using discre-

tionary funds, we are currently investigating one way to utilize this amplification and still look at damage to the DNA rather than the effect that damage has on the cell's proliferative capacity. If more than one kind of damage to the DNA at the gene or chromosome level can be detected, then each of these types can be investigated with respect to the effects of repair (or mis-repair) and the distribution of initial radiochemical products on its production. This would begin the process of filling in the steps from initial energy deposition to the final effect on the cell.

The technique relies on the use of cloned eukaryote genes (grown in bacterial cells and then labeled with a radioactive tracer) and the culturing of large numbers of cells known to have a radiation-induced defect related to the cloned gene. To evaluate the range of chromosome alterations, large numbers of cultures, each derived from a single irradiated cell, are needed.

The hypoxanthine guanine phosphoribosyl transferase (HGPRT) mutation system is one possible way to meet these requirements. This mutation occurs in a nonessential gene so that cells with inactive genes are easily detected and grown. The gene itself has been cloned and partially sequenced so it can easily be used in *in situ* hybridization and blot test assays. We are currently refining the techniques necessary to determine the nature of the chromosome changes being expressed as the HGPRT mutation in electron-irradiated Chinese hamster ovary cells.

• Modeling Cellular Response to Genetic Damage

This project is directed toward achieving a better understanding of the mechanisms by which damage induced in mammalian cells by physical and chemical carcinogens produces lethal and mutagenic effects. Our results continue to show the integrated nature of cellular response to radiation or chemical insult. Expression of DNA damage as either loss of reproductive capacity or mutation is modulated by intracellular pools of precursors used in DNA synthesis. This is probably due to the role of these precursors in regulating the initiation and/or rate of DNA replication which modifies the extent to which DNA damage is repaired. In some cases carcinogens appear to induce pool imbalance as well as DNA damage. The goal of this project is to elucidate and model the impact of perturbations in cellular metabolism on the cellular response to DNA damage.

REGULATION OF DNA SYNTHESIS BY DEOXYNUCLEOSIDE POOLS AND THE INDUCED MUTATION RESPONSE

C. N. Newman

A mutagen-induced enhancement of enzymes in Chinese hamster ovary cells associated with the synthesis/degradation of deoxynucleoside triphosphates (dNTP's, DNA precursor molecules) has been demonstrated (Newman 1985; Newman and Miller 1985). This altered enzyme response causes a marked imbalance in the concentrations of dCTP and dTTP: the former decreases while the latter increases. A dCTP/dTTP imbalance, effected by growing cells in the presence of 2 mM deoxycytidine (CdR), results in up to a tenfold reduction in mutations by ultraviolet radiation.

Studies of dNTP pool imbalances and mutation frequencies have been extended and expanded in the past year. Cells grown in the presence of CdR show a two- to threefold decrease in mutation from ionizing radiation (Figure 1). This result is qualitatively consistent with the earlier finding for UV radiation, suggesting an important role for dNTP imbalances and the cellular response to any premutagenic DNA damage.

A hypothesis that dCTP and dTTP each exert a regulatory role in the synthesis of cellular DNA was advanced in part, first by Nicander and Reichard (1981), then by Newman and Miller (1983). A correlary hypothesis proposed here is that an aberrant regulation of DNA synthesis may be caused by the abnormal levels of dCTP and dTTP that result from exposure of cells to mutagens or from growth in the presence of CdR. This, in turn, may mediate a

FIGURE 1. Induction of HGPRT⁻ Mutations by Ionizing Radiation. Exponential-phase CHO-K1 cells were exposed to 5 Gy 1.5 MeV electrons then, subsequently grown and periodically analyzed for resistance to 10⁻⁵ M 6-thioguanine. Growth was in the presence (●) or absence (O) of 2 mM deoxycytidine. Bars represent one standard deviation.

reduction in the frequency of induced mutation, possibly by interfering with the kinetics of mutation-fixation. Indeed, several experiments, discussed below, are consistent with this idea.

The inhibition of DNA replication by UV or ionizing radiations is greater and of longer duration when cells are grown in the presence of 2 mM CdR, as shown by [^3H] thymidine pulse incorporation data in Figures 2 and 3. Each response is normalized to an appropriate sham-irradiated control culture, and is therefore likely independent of differences in specific radioactivity caused by differential utilization and degradation of exogenous thymidine in the two media (Table 1).

Sucrose gradient profiles of radiolabeled DNA obtained from cells in each of the aforementioned media are presented in Figure 4. When cells are pulse-labeled with [Me^3H]-thymidine for 10 min, then "chased" (i.e., grown in the absence of label) for one hour, label from cells grown in CdR-containing medium sediments further down the gradient than label from cells grown without CdR. This result

FIGURE 2. Inhibition of DNA Synthesis by UV Radiation. Exponential-phase CHO-K1 cells grown in the presence of [$2\text{-}^{14}\text{C}$]thymidine were exposed to 10 J M^{-2} UV radiation, then periodically pulse-labeled with [^3H]thymidine for 10 min. Different symbols represent separate experiments. Cells were grown in Ham's F-12 with 10% fetal calf serum (□, ○, ■, ●) or Ham's F-12 with 10% fetal calf serum and 2 mM deoxycytidine (Δ).

FIGURE 3. Inhibition of DNA Synthesis by Ionizing Radiation. Exponential-phase CHO-K1 cells grown in the presence of [$2\text{-}^{14}\text{C}$]thymidine were exposed to 15 Gy 1.5 MeV electrons then periodically pulse-labeled for 10 min with [^3H]thymidine. Cells were grown in Ham's F-12 with 10% fetal calf serum (●) or in Ham's F-12 with 10% fetal calf serum and 2 mM deoxycytidine (○).

suggests that normal replication (chain elongation) is more rapid in the CdR medium, possibly because the positive effector of replication, dCTP, is present in a larger concentration. In contrast, when cells are UV-irradiated just prior to the "chase" period, DNA chain elongation is inhibited. This inhibition is greatest in cells grown in the presence of CdR.

Thus, the DNA synthetic rate is enhanced in the presence of CdR, a condition likely to result, in turn, in increased levels of the positive effector of replication, cellular dCTP. When these cells are exposed to UV radiation, which activates key enzymes of DNA precursor synthesis and degradation (Newman and Miller 1985), the dCTP is likely converted to the negative allosteric effector molecule, dTTP. This, then, probably exacerbates the radiation-induced inhibition of replication, affecting both its magnitude and duration. A working hypothesis, presently under investigation, is that the enhanced inhibition of replication in cells grown in CdR allows more time for repair of premutagenic lesions before damage is irreversibly "fixed" by subsequent DNA synthesis. This mechanism could account for the lower frequency of induced mutations in CdR-grown cells.

TABLE 1. Distribution of Continuous Label in CHO-K1 Cultures

Medium	% Extracellular Continuous Label ^(a)		Incorporated ^(b) [¹⁴ C]dpm/10 ⁶ cells
	TdR	Thymine	
F-12 + 10% fetal calf serum	9.9	90.1	15038
F-12 + 10% fetal calf serum and 2mM CdR	99	1	61399

(a) 10⁵ cells were inoculated in respective media with 3 μ M TdR, and [6-³H]TdR, 0.1 μ Ci/mL. Growth was for 72 hr ($\sim 2 \times 10^6$ cells); TdR and thymine determined by thin layer chromatography analysis of spent growth media.

(b) Cells were grown for 72 hr as above but with [2-¹⁴C]TdR, 0.019 μ Ci/mL instead of [6-³H]TdR.

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FIGURE 4. Sucrose Gradient Profiles of [³H]thymidine Pulsed-Labeled DNA. Exponential-phase CHO-K1 cells grown in the presence (○, ●) or absence (□, ■) of 2 mM deoxycytidine were pulsed-labeled with [³H]thymidine (10 min) then UV irradiated, 10 JM⁻² (closed symbols) or not irradiated (open symbols). Subsequent growth in the absence of label for 1 hr was followed by exposure to 750R x-rays then lysis for 1 hr on 5-20% alkaline sucrose gradients at ambient temperature. Centrifugation was at 8000 rpm at 20°C for 17 hr ($w^2t = 4.28 \times 10^{10}$). Gradients were subsequently fractionated into 5-drop fractions. (Δ indicates cells continuously labeled with [2-¹⁴C]thymidine.)

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Fig. 1. Effect of temperature on the rate of polymerization. The reaction was carried out in a 100 ml. solution of 0.1 mole/l. of monomer and 0.01 mole/l. of catalyst. The concentrations of the reactants were: (a) 0.1 mole/l. of monomer and 0.01 mole/l. of catalyst; (b) 0.2 mole/l. of monomer and 0.02 mole/l. of catalyst; (c) 0.3 mole/l. of monomer and 0.03 mole/l. of catalyst; (d) 0.4 mole/l. of monomer and 0.04 mole/l. of catalyst.

The rate of polymerization was measured by the change in viscosity of the solution. The reaction was carried out in a 100 ml. solution of 0.1 mole/l. of monomer and 0.01 mole/l. of catalyst. The concentrations of the reactants were: (a) 0.1 mole/l. of monomer and 0.01 mole/l. of catalyst; (b) 0.2 mole/l. of monomer and 0.02 mole/l. of catalyst; (c) 0.3 mole/l. of monomer and 0.03 mole/l. of catalyst; (d) 0.4 mole/l. of monomer and 0.04 mole/l. of catalyst.

Table 1. Effect of temperature on the rate of polymerization.

Temperature (°C)	Rate of polymerization (mole/l-hr)
10	0.1
20	0.5
30	1.2

The rate of polymerization was measured by the change in viscosity of the solution. The reaction was carried out in a 100 ml. solution of 0.1 mole/l. of monomer and 0.01 mole/l. of catalyst. The concentrations of the reactants were: (a) 0.1 mole/l. of monomer and 0.01 mole/l. of catalyst; (b) 0.2 mole/l. of monomer and 0.02 mole/l. of catalyst; (c) 0.3 mole/l. of monomer and 0.03 mole/l. of catalyst; (d) 0.4 mole/l. of monomer and 0.04 mole/l. of catalyst.

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• Internal Microdosimetry

The purpose of this study is to develop practical methods for calculating microdosimetric distributions for plutonium or other alpha-emitting elements that are deposited internally, especially as particulates, in soft tissue and lung tissue. This study will aid the correlation and extrapolation of radiation effects measured at different levels of exposure, in different distributions in dose and in different species. Computational methods are developed and tested in the Radiological Physics Section. Concurrently, the Dosimetry Technology Section develops cell and tissue models in which those methods are applied.

MOMENTS OF MICRODOSIMETRIC QUANTITIES FOR PARTICULAR SOURCES*

W. C. Roesch

A. M. Kellerer and H. H. Rossi showed that the mean square of the specific energy, a microdosimetric quantity, was linear-quadratic in the absorbed dose. W. C. Roesch enlarged the theory of such moments by showing how some of them could be calculated for internally deposited sources. The latter work is here extended to more cases of internally deposited radioactive particulates and to some cases of externally incident radiation. The mean square specific energy for particulates contains the same terms as for external radiation plus another term that represents the effect of more than one particulate. This term can be linear, quadratic, or linear-quadratic in the dose depending on how the experiment is done.

ALPHA-PARTICLE DOSIMETRY

W. C. Roesch

Early this year W. C. Roesch was appointed to the National Academy of Sciences-National Research Council's committee "Biological Effects of Internally Deposited Alpha-Emitting Radionuclides and Their Decay Products." This title is so long that it was immediately shortened to BEIR-4, a name indicative of its position in the Academy's sequence of committees concerned with the biological effects of ionizing radiation. Dr. Jacob Fabrikant of the University of California-Berkeley is the chairman; there are currently eleven other members. The committee was asked to recommend ways to make

quantitative estimates of the risk to humans from internally deposited alpha emitters.

The committee has met five times since its formation: February 26-27, May 15-17, June 21, July 23-24, and September 18-20, 1985. A day of the second meeting was opened for testimony from the public. Otherwise, the early meetings were devoted to selecting and assigning topics and deciding what original work had to be done. The committee is now reviewing draft manuscripts and listening to invited speakers on special topics. W. C. Roesch prepared a draft general chapter on the physics and dosimetry of alpha-particle emitters and is now engaged in preparing material on the dosimetry of individual radionuclides.

SPECIFIC ENERGY DISTRIBUTIONS FOR ALPHA EMITTERS IN THE DOG LUNG

D. R. Fisher, R. T. Hadley, and R. Harty

The radiation dose to lung tissue from inhaled alpha-emitting materials can be described by either 1) the absorbed dose averaged over the entire mass of lung tissue, or by 2) a frequency distribution of doses in "specific energy" to small target sites within the lung. Each method has advantages and disadvantages. The absorbed dose is useful for record keeping, general comparisons, and approximations of the probable health risk. For a more detailed analysis of the importance of dose distribution, particle size effects, and specific activity of inhaled particles, a microdosimetric approach is needed.

Specific energy distributions are calculated using computer codes (Hadley, Wilson, and Fisher 1985). To do this, one must characterize the particle size distribution of the

*Abstract of a paper by this title published in 1985 in Radiat. Res. 102:392-398.

inhaled aerosol, consider deposition probabilities in the lung for various particle sizes, and relate the physical size of inhaled particulates with their aerodynamic diameters. Next, one must consider the distribution of target sites (such as individual epithelial or endothelial cell nuclei) in the lung, and the probability of an alpha particle intersecting one of these sites as it passes through tissue and air space. A lung model for microdosimetry of the dog lung was developed earlier for this purpose (Fisher and Hadley 1984). Finally, the range, energy loss, straggling characteristics, and delta-ray production of alpha particle tracks must be accounted for.

Specific energy distributions provide a graphic display of the stochastic variation in energy deposition and a means for comparing different exposure conditions involving inhaled alpha emitters. In the following example, we compare the microdosimetry of different particle size fractions in the lung of a beagle.

Dog number 753 received an inhalation exposure to $^{239}\text{PuO}_2$ and was followed for life-span effects. Original cascade-impactor sampling data were used to determine the particle size distribution of the plutonium oxide aerosol to which dog 753 was exposed. The mean particulate diameter of each of six aerosol fractions was calculated, and all particulates in that size range were assumed to be equivalent in size and activity.

A probability density in specific energy was calculated for each aerosol fraction (Figure 1), correcting for the deposition probability in the dog lung. This figure shows the characteristic distribution of doses to cell nuclei in the lung from each fraction. The mean physical diameter (of the particulates in each aerosol fraction) is the diameter of a sphere of equivalent activity with mass density of 10.6 g/cm^3 . The absorbed dose and percent of tissue irradiated by particulates of each fraction are also indicated. The combined probability density in specific energy for all six aerosol fractions was determined by convolution (Figure 2). The absorbed dose to the whole lung was 5141 rad (51.41 Gy).

FIGURE 1. Probability Densities in Specific Energy for Six Aerosol Fractions of Plutonium Oxide in a Beagle Dog.

FIGURE 2. The Probability Density in Specific Energy for the Convolution of Six Aerosol Fractions of $^{239}\text{PuO}_2$ Inhaled by Dog 753.

In a typical occupational inhalation incident, the deposited material may consist of particulates having a large range of physical sizes. Aerosol diameter influences deposition probability in the lung and also the

probability that cells near sites of deposition will be irradiated. Particulates of different size and specific activity irradiate lung tissue quite differently. By examining the microdosimetry of different aerosol fractions, we are able to observe their relative importance to the total dose distribution.

Probability densities in specific energy demonstrate the way radiation energy is distributed among cells at risk in the lung. Further study is needed to define the relationship between probability density in specific energy and the probability of lung tumor induction.

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W. E. Wilson
N. A. Wogman
Life Sciences Library (2)
Publishing Coordination (2)
Technical Information (5)