

wrong in the work, do not depend on any human authority. It may sometimes take many years before one knows the solution of a problem, before one can distinguish between truth and error; but finally the questions will be decided, and the decisions are made not by any group of scientists but by nature itself.

3) Finally there is the implication of an opportunity—nay, necessity—to turn our attention to the structure and the foundations of the moral and ethical framework within which we seek assistance in deciding *why* we must do those things that science tells us *how* to do. What an unparalleled opportunity to synthesize advances toward an un-

derstanding of our physical universe with advances in our understanding of man's role in this universe! For, as Teilhard de Chardin has suggested with such simple eloquence in *The Phenomenon of Man*:

How can one fail to recognize this revealing association of technical mastery over environment and inward spiritual concentration as the work of the same great force—the very force that brought us into being.

References

1. V. J. Schaefer, *Science* **104**, 457 (1946).
2. NAS-NRC Panel on Weather and Climate Modification, *NAS-NRC Publ. 1350* (Washington, D.C., 1966).

3. Advisory Committee on Weather Control, *Final Report, I* (Government Printing Office, Washington, D.C., 1957).
4. NSF Special Commission on Weather Modification, *NSF Publ. 66-3* (1966).
5. U.S. Senate Committee on Commerce, *Rept. 1139* (27 April 1966); U.S. Senate, *Weather Modification Act of 1966* (14 Oct. 1966).
6. AAAS Committee on Science in the Promotion of Human Welfare, *Science* **132**, 68 (1960).
7. V. J. Schaefer, *ibid.* **154**, 1555 (1966).
8. J. von Neumann, *Fortune* **51**, 106 (1955).
9. E. A. Morris, in *Human Dimensions of Weather Modification*, W. R. Sewell, Ed. (Univ. of Chicago Press, Chicago, 1966), p. 283.
10. Anon., *New York Times*, 10 June 1965.
11. See, for example, P. H. Abelson, *Science* **154**, 1123 (1966).
12. H. Cleveland, in R. N. Gardner, *In Pursuit of World Order* (Praeger, New York, 1964), foreword.
13. W. Heisenberg, *Physics and Philosophy*, (Harper, New York, 1958).

Destruction of Molecules by Nuclear Transformations

A molecule may explode as a result of some kinds of nuclear transformation of a constituent atom.

S. Wexler

Picture a molecule isolated in space, one of whose atoms is radioactive, that is, its nucleus is unstable. What happens to the molecule when the particular atom undergoes a nuclear transformation, when it changes to another energy state of the same nucleus or to an entirely different nucleus? For a number of reasons we may expect rather unique molecular effects. First, nuclear transformation is an internal process; the change occurs within the nucleus of a particular atom. The consequences of the nuclear event for the specific atom, as well as for the whole molecule, may be quite different from the effects provoked by an exciting agent, such as an electron, proton, or alpha particle, initially external to the molecule. Since the process takes place in a particular atom, its position in the molecule may be a decisive factor in the effects produced. Then, there are numerous dif-

ferent nuclear processes, and each influences the molecule in a characteristic way.

I will describe some of the phenomena resulting from nuclear transformations occurring in polyatomic molecules (1). The primary molecular effects to be discussed are the properties of the product species at the moment of formation, before these properties are altered by collisions of the products with neighboring molecules. The observable properties are the chemical natures, charge states, and kinetic energies of the various fragments formed in the breakup of the molecule. From these measured quantities we may deduce the mechanisms of molecular disruption induced by the nuclear transition. Experimental observations in this scientific discipline are made on an atomic or molecular scale, and they differ from those performed in nuclear physics since the latter usually consider the properties of the lightweight particles and photons initiating the nuclear tran-

sitions or emitted in them. The primary molecular processes occurring in nuclear events are studied as part of a general area of scientific interest concerned with the chemical effects of nuclear transformations. Because of the unusual charge, the large electronic, vibrational and rotational energies, and the excessive recoil energies received in the nuclear transformation, the various species, though altered by subsequent collisions, may eventually react chemically with molecules in unique ways not open to the species in their normal states (2).

Experimental Techniques

Two requirements must be fulfilled for the decomposition of molecules by nuclear decay to be studied. There must be low pressures of radioactive gaseous molecules to insure that measurements are made on only the primary products arising from events in isolated molecules and not on those formed by secondary reactions, and there must be efficient detection and identification of the new species. Fortunately, most of the products become electrically charged as a result of the nuclear process, and, consequently, they can easily be deflected in electric and magnetic fields and detected by sensitive electronic devices. Radioactive species often produced in transformations can also be collected on charged electrodes, and the charge states of the species can be deduced from the manner in which the electric field affects the distribution of active material deposited on the various electrodes (3). In other experiments, the direct currents produced by the charged products are measured, and

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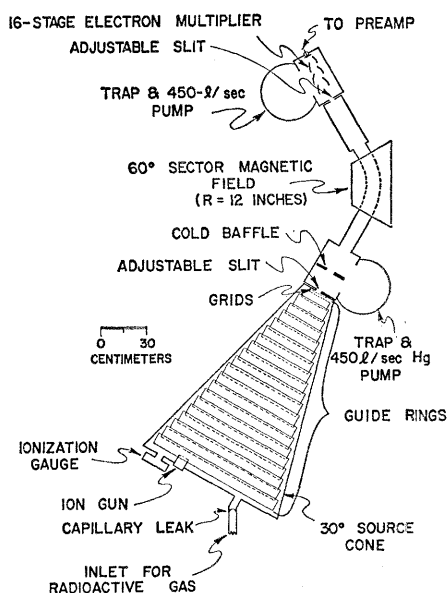


Fig. 1. Mass spectrometer for studies on fragmentation of molecules by nuclear transformation.

from such observations their mean charge can be determined (4).

The technique that has given the most detailed picture of the primary molecular processes associated with nuclear transformations has been that of mass spectrometry. A large number of different gaseous compounds have been synthesized with radioactive atoms incorporated in the molecules in high specific activity, that is, a high percentage of the molecules contained radioactive constituents. For the study of its fragmentation, each gas was introduced into a mass spectrometer (an instrument capable of separating ionic species according to mass and charge) with special constructional features. A sketch of the mass spectrometer at the Argonne National Laboratory is shown in Fig. 1. A spectrometer of considerably different design has been used for similar experiments at the Oak Ridge National Laboratory (5). While present at very low pressure in the large source chamber (Fig. 1), the radioactive gas is allowed to undergo spontaneous nuclear transformations, and the charged species formed in the nuclear decays are swept toward the apex of the cone by an axial electric field produced by the application of proper voltages to an electrode system within the conical housing. Just before the ions reach the adjustable source slit they are accelerated to high energies as they pass between two grids. The ions then leave the source chamber and drift into the magnetic field, where they are sorted out according to mass and charge. By varying the strength of the magnetic

field, we may focus in turn beams of each of the numerous fragments on the detector slit, through which the ions pass to be detected electronically by a very sensitive electron multiplier that can detect every ion that hits it. From the kinds of ionic fragments observed and their relative abundances (called the mass or charge spectrum), the mechanism of the dissociation of the molecule due to the nuclear event may be deduced.

Examples of molecular disruption observed in the mass spectrometer after three different kinds of nuclear transformations are presented below.

Negative Beta-Decay

When a nucleus of an atom undergoes negative beta-decay, it emits a very fast electron, and, if no electrons surrounding the nucleus (the so-called atomic or orbital electrons) are ejected at the same time, the neutral parent atom will be converted to an ion bearing a single positive charge. In addition, because the nuclear charge (the number of protons the nucleus contains) of the atom is increased by one unit, the chemical nature of the ion is that of the element one higher in the periodic table than that of the precursor atom, before the transformation took place. Note that ejection of an orbital electron is not necessary for the production of the charged daughter atom. The charging occurs as a natural consequence of the characteristics of this particular nuclear transformation. If the radioactive atom is initially bound in a molecule, the product of the negative beta-decay would be expected to be a positively charged molecular ion. However, several processes which accompany the emission of the beta particle may leave the molecular ion in a state of electronic, or vibrational-rotational excitation, or both, and as a result the ion may dissociate. By far the most important of these auxiliary processes is the so-called "shaking" of the cloud of electrons surrounding the transforming nucleus. Before the transformation, the electrons move about in their orbits under the influence of the positive charge of the parent nucleus. Suddenly a beta particle is eliminated from the nucleus, and the nuclear charge consequently increases by one unit. The electrons now suddenly feel the potential field of the increased nuclear charge, and a fair fraction of the daughter ions become electronically excited. Theoretic-

Table 1. Yields of initial fragment ions from beta decay of atoms in organic compounds.

Molecule	Initial fragment ion	Ion yield (%) abundance)
CH_3T	CH_3^+	83
$\text{C}_2\text{H}_5\text{T}$	C_2H_5^+	80
$n\text{-C}_3\text{H}_7\text{T}$	C_3H_7^+	56
$\text{sec-C}_3\text{H}_7\text{T}$	C_3H_7^+	41
$\text{C}_6\text{H}_5\text{T}$	C_6H_5^+	72
$o\text{-C}_6\text{H}_4\text{TCH}_3$	C_6H_7^+	78
$m\text{-C}_6\text{H}_4\text{TCH}_3$	C_7H_7^+	79
$p\text{-C}_6\text{H}_4\text{TCH}_3$	C_7H_7^+	76
$\text{C}_6\text{H}_5\text{CH}_2\text{T}$	C_7H_7^+	79
$\text{CH}_3\text{Br}^{82}$	CH_3^+	89
$1,2\text{-C}_2\text{H}_4\text{BrBr}^{82}$	$\text{C}_2\text{H}_4\text{Br}^+$	70
$\text{CCl}_3\text{Br}^{82}$	CCl_3^+	64
$\text{CH}_3\text{I}^{131}$	$(\text{CH}_3\text{Xe}^{131})^+$ and CH_3^+	69 2.0
$\text{CH}_3\text{I}^{130}$	$(\text{CH}_3\text{Xe}^{130})^+$ and CH_3^+	34 39
$\text{C}_2\text{H}_5\text{I}^{131}$	$(\text{C}_2\text{H}_5\text{Xe}^{131})^+$ and C_2H_5^+	1.4 89

ical estimates give a value of 30 percent as the percentage of $(\text{He}^3)^+$ that should be excited by beta decay of a tritium atom. Another contributing cause of molecular rupture is the recoil suffered by the daughter atom as a result of the ejection of the very energetic electron.

The rather typical spectrum of charged species formed by beta decay to He^3 of the tritium in monotrinitated benzene appears in Fig. 2 (6). Several unique characteristics of the mechanism of disruption of the benzene molecule following this transformation may be deduced from the histogram:

1) After the transition of $\text{C}_6\text{H}_5\text{T}$ to $(\text{C}_6\text{H}_5\text{He}^3)^+$, the carbon-helium bond almost always breaks, and the positive charge of the daughter molecule ion is taken by the organic fragment. The He^3 is apparently released as the neutral atom.

2) The organic fragment, called the initial fragment ion, from this bond rupture (in this case the phenyl ion C_6H_5^+) dominates the pattern; 72 percent of the beta transformations produce only this one ionic species. In the remaining decays the initial fragment ions possess sufficient excitation to dissociate further, by ruptures of carbon-carbon and carbon-hydrogen bonds to a large variety of ionic species.

3) That there are low yields of doubly charged ions suggests that ejection of orbital electrons after the nuclear event is either of minor importance or that multiply charged polyatomic ions break apart in the source volume of the mass spectrometer.

Similar studies on a large number

of molecules tagged with tritium as well as with other radioactive atoms tend to corroborate the conclusions derived from the results on C_6H_5T (1). But isomeric effects (those due to the position of the radioactive atom in the molecule) and the influences that the recoil energy and the strength of the bond between the carbon and the rare gas atom have on the fragmentation pattern are sometimes observed. A very high fraction of the beta transformations in each organic compound usually gives only the initial fragment ion (Table 1). But smaller than expected yields of these species are found in the two isomeric propanes, because of the abnormally low energy required for dissociation of $C_3H_7^+$ into allyl ($C_3H_5^+$) ions by the elimination of H_2 . In addition, the abundance of the $C_3H_5^+$ ions from the propane in which tritium atoms had been attached to middle car-

bon atoms is lower than that from the propane with the tritium on end carbons. This observation is consistent with the fact that the structure of the secondary propyl ion has greater stability than its primary isomer has.

Almost identical ion patterns are found following transformations of tritium atoms attached to the four distinguishable positions (ortho, meta, para, and alpha) on the skeleton of the toluene molecule. This result can be accounted for by rearrangements, prior to dissociation, of the three tolyl and one benzyl structures formed by elimination of neutral He^3 atoms from the respective daughter species. The rearrangements all lead to the same seven-membered ring structure (the "tropylum" ion), in which the carbons and hydrogens are respectively indistinguishable.

The presence of the daughter molec-

ular ion (CH_3Xe^{131}) $^+$ in very high yield (Table 1) after decay of CH_3I^{131} , rather than its initial fragment ion CH_3^+ , has been accounted for by the exceptional strength of the carbon-xenon bond in this ion. Other differences found between the ionic abundances of species produced by decay of the iodine isotopes are the result of variations in amounts of recoil energy.

Not presented in Table 1 are the observations on the molecules $C^{14}O_2$ (5), TH , and T_2 (7). The preponderant product of the beta decay of carbon-14 is the $(NO_2)^+$ ion. Thus, the only thing that usually happens to the carbon dioxide molecule is the transformation of the carbon nucleus to a nitrogen nucleus, with accompanying charging of the molecule. In a minor fraction of the transitions the $(NO_2)^+$ is sufficiently excited that it dissociates into NO^+ , O^+ or N^+ , and unobserved neutral frag-

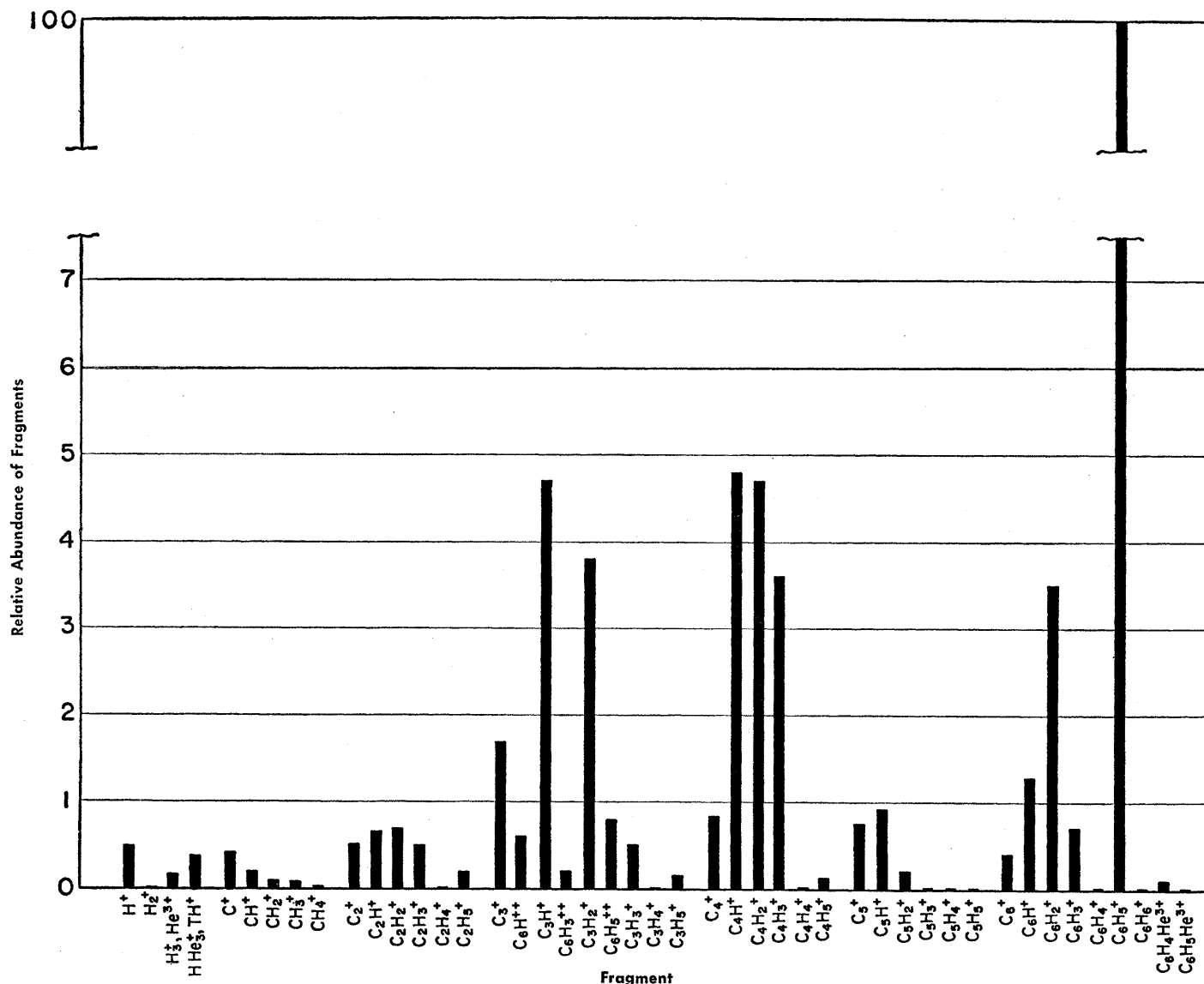


Fig. 2. Molecular fragmentation pattern following beta decay of monotritiated benzene (6).

ments. Similarly, nuclear transformations of tritium in the hydrogen-like molecules TH and T₂ lead to production of (HHe³)⁺ and (THe³)⁺, respectively, in approximately 90 percent of the decays. Small amounts of H⁺, (He³)⁺, and T⁺ are also formed.

In summary, nuclear transformation

of a constituent atom by emission of a beta particle causes only minor molecular disruption in a large fraction of the events. Because of the weakness of the bond, only a neutral rare-gas atom is usually eliminated from the daughter ion. In a smaller fraction of the transitions, the initial fragment ion dissociates

by a variety of modes to give a complex pattern of ionic fragments. This picture of molecular consequences of beta decay has been confirmed by the results of an entirely different experimental approach. A technique of double labeling has recently been used in which two tritium atoms were attached to the same molecule in order that the chemical reactions of the product ions from beta emission of one of the tritium atoms could be followed by means of the radioactivity of the second tritium (8). The yields of the stable products, as determined by radiogaschromatography, could be closely predicted from the abundances of the primary species obtained from the mass spectrometric experiments when these ionic species were assumed to react with the principal gaseous compounds by established ion-molecule reactions.

Isomeric Transition

In contrast to the rather mild effects of beta emission, a molecule may suffer disastrous consequences following the nuclear process of isomeric transition. This mode of transformation involves the de-excitation of a nucleus from an excited state to one of lower energy. It may be accomplished by emission of a photon of radiation (a γ -ray) from the nucleus of the attached radioactive atom. In this case, the molecule is almost invariably unperturbed. Instead of a γ -ray being emitted, however, the nuclear de-excitation may occur through the ejection of an electron from an electron shell deep within the atom (a shell near the nucleus). This process is called internal conversion. Its immediate effect is to cause the atom to be ionized to a species with a single positive charge. More importantly, the removal of the orbital electron from the deep-lying shell (usually the innermost K shell) leaves a hole there. This is a very unstable situation because the energy by which an electron is held in this shell is greater than twice the binding energy of an electron in the next outer shell (the L shell). Consequently, an L electron falls into the K shell, and a second L electron may be ejected from the atom (the process is called the Auger effect), producing now a doubly-charged ion. The two holes in the L shell may be filled in like manner by electrons from the next outer shell, giving temporarily a quadrupally charged ion. The concurrent movements of electrons then proceed

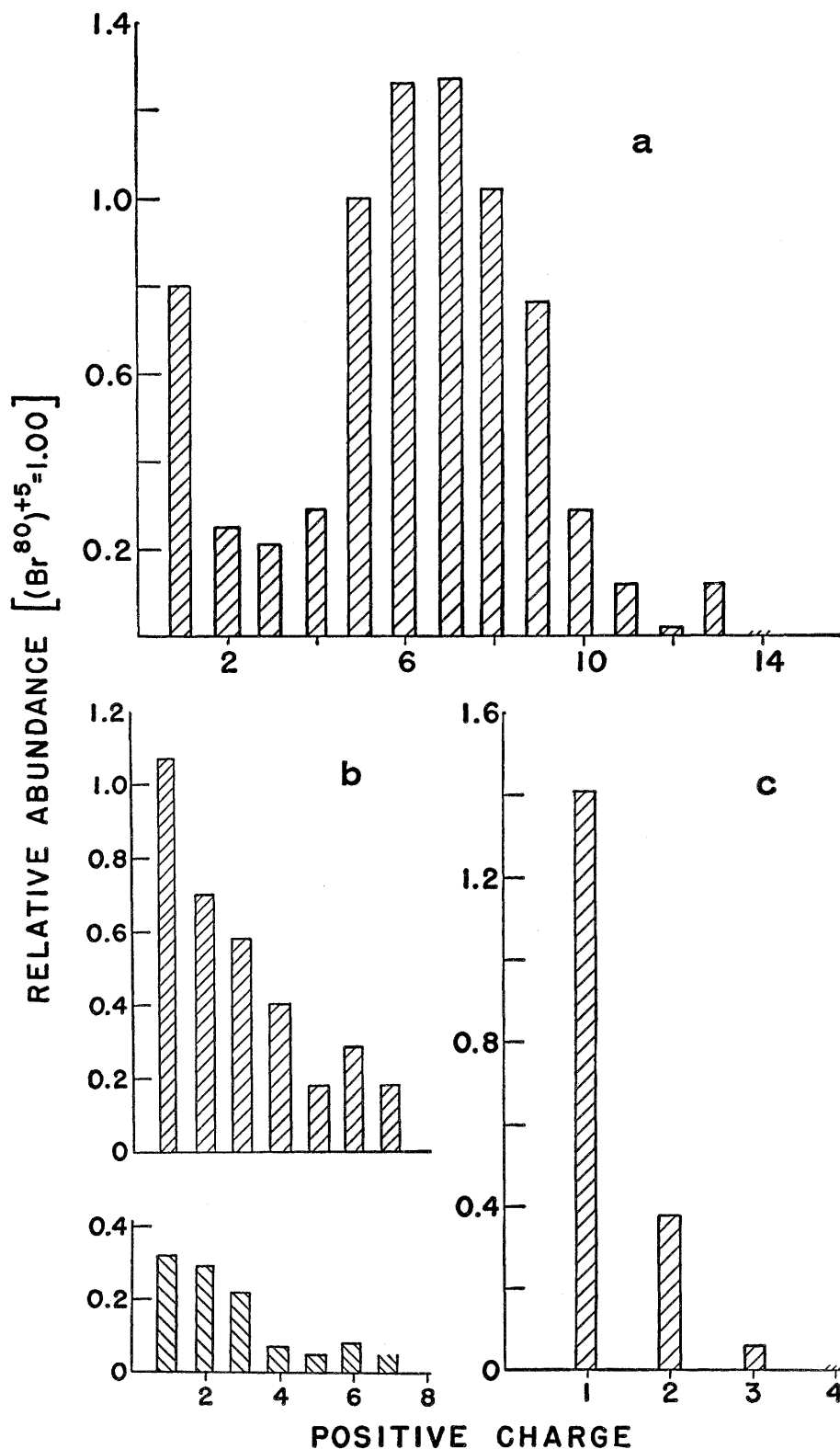


Fig. 3. Charge states of (a) Br⁸⁰ atoms, (b) chlorine isotopes (Cl³⁶, top; Cl³⁷, bottom), and (c) carbon fragments (C¹²) following isomeric transition CCl₃Br^{80m} (9).

through the remaining shells, so that a large number of holes are created in the outermost or valence shell of the atom. The entire charging process, called a vacancy cascade, takes place in a very short time, generally less than 10^{-15} second, which is much shorter than the periods of vibration of the radioactive and other atoms in the molecule. The large number of holes in the valence shell of the affected atom may be partially filled in a time of similar shortness by electrons from other atoms of the molecule, and these rapid transfers of electrons result in positive charges being distributed throughout the molecule. The charging and electron redistribution thus occur while the molecule remains intact. But since the positively charged atoms in the molecule strongly repel each other (coulombic repulsion), the molecule virtually explodes as a consequence.

That the foregoing mechanism of molecular destruction following isomeric transition is correct has been proved by the patterns of charged species observed following decay of the excited nuclear species $\text{Br}^{80\text{m}}$ incorporated in trichlorobromomethane (9) (Fig. 3). The charge distributions of bromine ions extending up to +13 electron units with a maximum having +7 electron units, of the two chlorine isotopes decreasing monotonically in abundance from +1 to +7 electron units, and of the carbon ions decreasing in yield from +1 to +3 electron units, are evidence for the violent disruption of the CCl_3Br molecule following this particular nuclear event. Molecular bromine and bromomethane containing $\text{Br}^{80\text{m}}$ have also been observed to be destroyed with the production of numerous ions of positive charge (1). The high charges found on the ionic fragments from these three compounds are concordant with the mean charge of $+(10 \pm 2)$ electron units (determined several years previously from direct-current measurements) on the fragments from isomeric transition of $\text{Br}^{80\text{m}}$ in ethyl bromide (4).

Other techniques have been employed to demonstrate that carbon-halogen and hydrogen-halogen bonds are ruptured in a very large fraction (usually greater than 90 percent) of the isomeric transitions of $\text{Br}^{80\text{m}}$ covalently bound in molecules (10). In these experiments the organic halide molecule, for example, is tagged with the radioactive isotope and then allowed to stand as a gas or in solution for a specified time, after which the

Table 2. Fractions of radiative neutron captures not leading to bond rupture of molecules containing halogen (14).

Molecule	Organic halogen* (average percentage)
CH_3Br	0.25 ± 0.01
CD_3Br	$.20 \pm .02$
CH_3Br_2	$.115 \pm .010$
CF_3Br	$.105 \pm .004$
CF_3Br_2	$.093 \pm .004$
CHClBr_2	$.087 \pm .005$
CCl_3Br	$.066 \pm .004$
CHBr_3	$.048 \pm .013$
CBr_4	$.031 \pm .006$
$\text{C}_2\text{H}_5\text{Br}$	$.33 \pm .03$
$1,1\text{-C}_2\text{H}_4\text{Br}_2$	$.173 \pm .009$
CH_3I	$1.09 \pm .03$
CD_3I	$0.68 \pm .06$
CF_3I	$.12 \pm .02$
CH_3I_2	$.068 \pm .004$
$\text{C}_2\text{H}_5\text{I}$	$.82 \pm .04$
$n\text{-C}_3\text{H}_7\text{I}$	$.66 \pm .05$
$i\text{-C}_3\text{H}_7\text{I}$	$.30 \pm .05$

*The uncertainty is the standard deviation of the mean.

compound is shaken with aqueous solutions to extract the unbound radioactive daughter species of the isomeric transition. The degree of radioactivity of the extracted daughter is compared with that previously in equilibrium with the parent, or the growth of the active daughter in the gas or solution is followed after the extraction. From such data the fraction of molecules destroyed by the bond rupture may be calculated.

The amount of recoil energy received by the molecule when a γ -ray is emitted in an isomeric transition is insufficient to cause bond rupture. In illustration, zinc diethyl and zinc acetal acetate are not dissociated on isomeric transition of $\text{Zn}^{69\text{m}}$ (11), because the probability of internal conversion of this isotope is very small.

It should be noted that the entire chain of events ending in the violent destruction of a molecule was initiated by removal of an electron from a deep-lying shell of the radioactive atom during the course of the nuclear transition. But such creation of holes can also be performed by an external agent, such as an x-ray, and, indeed, molecules have been violently destroyed by this method (12), and the products have been observed to possess kinetic energies in the range of tens of electron volts.

Orbital Electron Capture

A very similar mode of destruction of a molecule occurs when one of its atoms is transformed by the nuclear process called orbital electron capture. As the name implies, the unstable nucleus absorbs an electron from usually the K or L shell of the atom, creating a hole in the shell, which is filled in a vacancy cascade similar to that following isomeric transition. Again, the production of a high electrical charge on the decaying atom and subsequent distribution of charges among the atoms of the molecule causes it to explode with the formation of a myriad of positive ions with rather high kinetic energies. In illustration, when $\text{CH}_3\text{I}^{125}$ and $\text{C}_2\text{H}_5\text{I}^{125}$ decay spontaneously by orbital electron capture, a spectrum of positively charged Te^{125} ions extending from +1 to +18 electron units with the greatest number having approximately +9 electron units is found (Fig. 4) (13). In addition to tellurium ions,

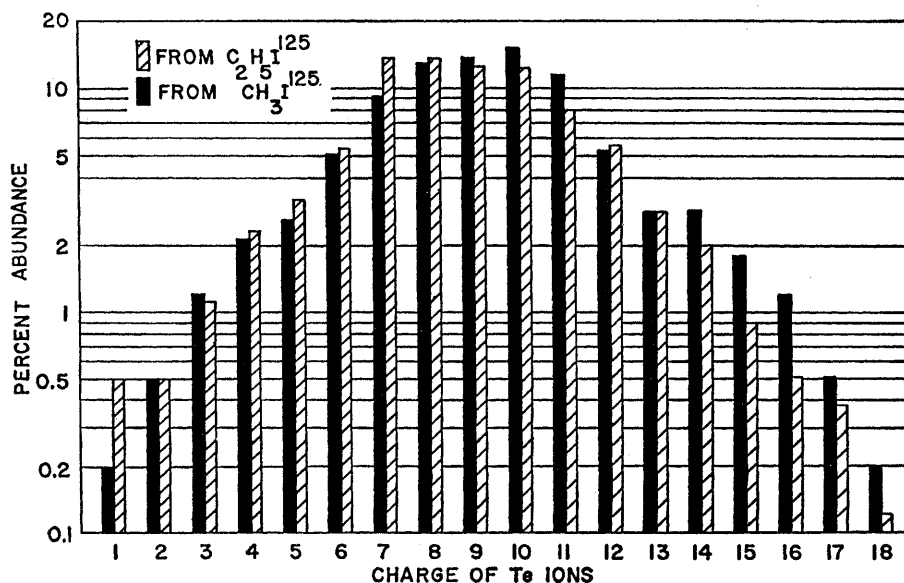


Fig. 4. Distributions of charge states of Te^{125} fragments from electron capture in $\text{CH}_3\text{I}^{125}$ and $\text{C}_2\text{H}_5\text{I}^{125}$ (13).

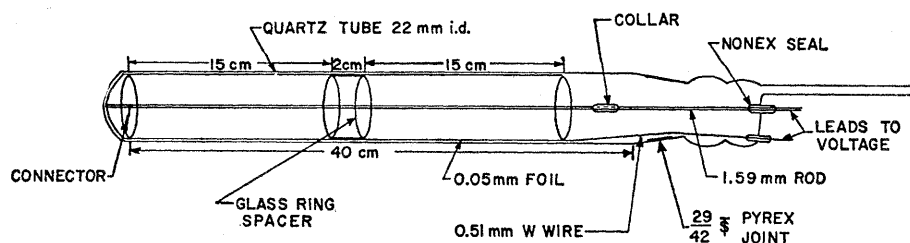


Fig. 5. Irradiation chamber for study of the charge states of recoil halogen atoms from neutron capture in molecules (16).

a complex pattern of singly and doubly charged hydrocarbons, carbon ions from +1 to +3 electron units, and H^+ is observed.

Although the mechanisms of molecular disruption are essentially the same following the nuclear transformations of isomeric transition and orbital electron capture, there are two fundamental differences. The latter event converts the radioactive species to an atom with chemical properties of the element one lower in the periodic table than that of the original atom. Thus, I^{125} became Te^{125} in the illustrations given above. In isomeric transition the element is unchanged. Second, the internal conversion that takes place in isomeric transition leaves the atom with a deep-lying hole and a charge of +1 electron unit, while orbital electron capture produces a neutral atom with a similar hole. The daughter of the capture process is initially uncharged because both the nuclear charge and the number of orbital electrons are each reduced by one.

Radiative Neutron Capture

Molecules may also be decomposed by nuclear reactions, that is, reactions in which an elementary particle, such as a neutron or a proton, or a high-energy photon (γ -ray or x-ray) interacts with a stable nucleus to form a different nuclear product. Of the many types of nuclear reactions, only molecular decomposition by radiative neutron capture has been extensively studied. In this reaction the uncharged neutron enters a stable nucleus, raising it to an excited state of extremely high energy. This excitation is usually dissipated with the emission of cascades of γ -rays, and, because some of the γ -rays are often of very high energy, the capturing nucleus experiences a sharp recoil. For example, an iodine atom may receive a recoil energy of up to 182 electron volts as a result of capturing a neutron with only thermal kinetic energy. Such energies are clearly far in

excess of usual bond energies (2 to 6 electron volts), and, consequently, a molecule containing the reacting atom may be ruptured.

A second cause of molecular decomposition following absorption of a neutron may be expected to be the internal conversion of one or more of the γ -rays emitted. Generally, this becomes more probable with decreasing energy of the photon, and, since each cascade of γ -rays usually contains one or more low-energy γ -rays, a fair fraction of the capturing atoms may become highly charged as a result of the ensuing vacancy cascades. A molecule containing the atom would be violently shattered by coulombic repulsion of the separated positive charges. Thus, this mechanism of rupture is identical to that following the spontaneous nuclear transformations of isomeric transition and orbital electron capture.

As expected from the large kinetic energy of recoil from the gamma emission and the high positive charge following internal conversion, the activated atom (it often becomes radioactive in the nuclear process) is freed from molecular combination in a very large fraction of the neutron captures. As illustrated by the data in Table 2 (14), only very small fractions of the carbon-halogen bonds in a large number of compounds are not destroyed by neutron capture. These small percentages have been accounted for by partial cancellations of the momenta of the randomly emitted gamma rays in the cascades following absorption of the neutrons. Indeed, the original experiment (the so-called "Szilard-Chalmers effect") in the scientific discipline described in this article was the observation in 1934 that an appreciable fraction of the radioactive iodine formed by neutron irradiation of ethyl iodide could be obtained in the unbound chemical state (15).

That destruction of molecules on neutron capture is accomplished by the charging from vacancy cascades and the subsequent coulombic repulsion of

separated charges that follow internal conversion of emitted γ -rays has been demonstrated experimentally (16). Gaseous ethyl bromide and ethyl iodide were each irradiated between concentric cylindrical, chemically active collecting electrodes in the very high flux of a neutron chain reactor, or "pile." The gas was introduced into the quartz-pyrex irradiation chamber containing the electrodes (Fig. 5) at sufficiently low pressures that the radioactive bromine and iodine species dissociated from the gaseous halide and deposited on the electrodes could be examined under nearly collision-free conditions. The volume between the concentric cylinders on the left in the figure was kept free of electric fields by connection of the two, while a high voltage of either polarity was impressed on the outer electrode of the assembly on the right. The axial rod was grounded electrically. After irradiation of the sealed chamber, the gas was pumped out and examined for radioactive products, the electrodes were removed from the chamber, and the central rod was divided into the sections adjacent to the outer cylinders. The radio-halide was removed from each electrode, and then, after suitable radiochemical procedures, its yield was measured. Since less than 1 percent of the halogen activity after an irradiation was found in the gas phase (the bulk of the radioactive products was deposited on the electrodes), virtually every ethyl halide that captured a neutron was decomposed. From the ratios of the radioactivities of the halide on the outer electrodes to those on the appropriate sections of the central rod in each of two separate experiments (in which the direction of the electric field could be reversed), the distribution of recoil halogens with respect to the sign of their electric charge could be calculated. When gaseous ethyl iodide was the target gas, 44 percent of the iodine atoms undergoing neutron capture was found to be positively charged; the remaining iodines were neutral. Within experimental error, no negative iodine ions were formed by the nuclear process. On neutron capture by ethyl bromide, the percentages of positive bromines ranged from 12 to 25 percent, the yield being dependent on which radioactive bromine isotope was produced in the reaction. Again, the remaining percentages of bromines were uncharged, no negative species being detected.

Indirect evidence that positively charged species are produced in this

nuclear process come from the inhibiting effect of certain atoms and molecules on the reactivity of the recoil halogen products with methane (17). The additive substances possess ionization potentials (the energies required to remove electrons from them) nearly equal to or lower than those of the radioactive products formed in the neutron capture, and so exchanges of electrons between them can readily take place. Apparently, the formation of methyl halide may proceed through the reaction of positively charged recoil halogen ions with CH_4 molecules, and this mechanism is prevented by the neutralization of the halogens as a result of transfer of electrons.

A wide variety of phenomena are observed when the nucleus of a constituent atom of a molecule is transformed spontaneously or by nuclear reaction. The characteristics of the molecular dis-

ruption depend strongly on the type of nuclear transformation that occurs. In the illustrations described in this article, negative beta-decay generally causes only mild perturbation of the molecule, whereas violent molecular consequences follow the nuclear processes of isomeric transition, orbital electron capture, and radiative neutron capture.

References and Notes

1. A comprehensive review of this field of research is given by S. Wexler, in *Actions Chimiques et Biologiques des Radications*, M. Hassinsky, Ed. (Masson, Paris, 1965), vol. 8, pp. 103-241.
2. For illustrations, see the reviews of R. Wolfgang, *Annu. Rev. Phys. Chem.* **16**, 15 (1965) and I. G. Campbell, *Advance. Inorg. Chem. Radiochem.* **5**, 135 (1963); and the *Proc. Symp. Chem. Eff. Nucl. Transformations Prague* (1961); *ibid. Vienna* (1965).
3. S. Wexler and T. H. Davies, *J. Chem. Phys.* **18**, 376 (1950); *ibid.* **20**, 1688 (1952); S. Yosim and T. H. Davies, *J. Phys. Chem.* **56**, 599 (1952); J. L. Thompson and W. W. Miller, *J. Chem. Phys.* **38**, 2477 (1963).
4. S. Wexler, *Phys. Rev.* **93**, 182 (1954); M. L. Perlman and J. A. Miskel, *ibid.* **91**, 899 (1953).
5. A. H. Snell and F. Pleasonton, *J. Phys. Chem.* **62**, 1377 (1958).
6. T. A. Carlson, *J. Chem. Phys.* **32**, 1234 (1960).
7. A. H. Snell, F. Pleasonton, H. E. Leming, *J. Inorg. Nucl. Chem.* **5**, 112 (1957); S. Wexler, *ibid.* **10**, 8 (1959).
8. F. Cacace, G. Ciranni, R. Guarino, *J. Amer. Chem. Soc.* **88**, 2903 (1966); B. Aliprandi, F. Cacace, A. Guarino, *Proc. Symp. Chem. Eff. Nucl. Transformations Vienna*, 471 (1965).
9. S. Wexler, *J. Chem. Phys.* **36**, 1992 (1962).
10. R. H. Luebbe and J. E. Willard, *ibid.* **29**, 124 (1958); W. H. Hamill and J. A. Young, *ibid.* **20**, 888 (1952); A. A. Gordus and J. E. Willard, *J. Amer. Chem. Soc.* **79**, 4609 (1957); R. S. H. Chiang and J. E. Willard, *ibid.* **74**, 6213 (1952).
11. G. T. Seaborg, G. Friedlander, J. W. Kennedy, *J. Amer. Chem. Soc.* **62**, 1309 (1940); J. W. Kennedy, G. T. Seaborg, E. Segre, *Phys. Rev.* **56**, 1095 (1939).
12. T. A. Carlson and R. M. White, *Proc. Symp. Chem. Eff. Nucl. Transformations Vienna*, 23 (1965); *J. Chem. Phys.* **44**, 4510 (1966).
13. T. A. Carlson and R. M. White, *J. Chem. Phys.* **38**, 2930 (1963).
14. A. A. Gordus and C. Hsiung, *ibid.* **36**, 954 (1962).
15. L. Szilard and T. A. Chalmers, *Nature* **134**, 462 (1934).
16. S. Wexler and T. H. Davies, *J. Chem. Phys.* **20**, 1688 (1952).
17. G. Levey and J. E. Willard, *ibid.* **25**, 904 (1956); E. P. Rack and A. A. Gordus, *ibid.* **34**, 1855 (1961); *ibid.* **36**, 287 (1962); *J. Phys. Chem.* **65**, 944 (1961).

Ultrastructure and Cytochemistry of the Synaptic Region

The macromolecular components involved in nerve transmission are being studied.

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Some of the most outstanding findings of modern biology have been achieved through use of physical and chemical techniques in the study of macromolecular structure and function. Within this realm, now called molecular biology, form and function, which for centuries were considered separate entities, are inseparable. In the study of the nervous system this stage of analysis has been reached more slowly, and progress has been more difficult because of the extraordinary morphological and physiological complexity of this system. In recent years, with the use of fine microelectrodes for intracellular recording, it has been demonstrated that neurons have specialized loci of activity at which a variety of local and propagated potentials occur.

Studies in which pharmacological agents and labeled ions are used have also revealed the existence of chemical receptors and of ionic mechanisms in specialized portions of a single nerve cell (1). Until recently, this wide variety of activities could only be correlated with the structures observable with light microscopy. Investigations of this sort showed the complexity of the fine neuronal processes, of the axons and dendrites, and of the intercellular connections, but they could not give any information on the subcellular components involved in such mechanisms. It is only since 1953 that, with the use of the electron microscope, some of the subcellular structures that underlie nerve activity could be demonstrated. This instrument, associated after 1959

with methods of cell fractionation, has led to the recognition of some important features in the submicroscopic and chemical organization of the synapse, defined here as the region at which the excitatory or inhibitory influences from one neuron are transmitted to another neuron or to an effector in an irreciprocal direction.

In 1877, Du Bois Raymond (2) first suggested that synaptic transmission could be either chemical or electrical, and both mechanisms have been found to occur in the peripheral and central nervous systems. However, chemical synapses are by far the most common and are the only ones that will be considered here from the point of view of ultrastructure and cytochemical organization. Chemical synapses are also endowed with other important but still little-known functions generally called trophic and plastic properties, which may be the neurological basis of conditioning and learning (1).

Hypotheses concerning chemical transmission are based on the assumption that a specific transmitter is synthesized and stored in nerve endings and that it is liberated when the nerve impulse arrives at the terminal. It is also postulated that the transmitter reacts with a chemical receptor situated in the postsynaptic element and that from this reaction a change in ionic permeability takes place, this change

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