Chemical Effects of Nuclear Decay

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Introudction

Radioactive decay imparts recoil energy to the daughter nucleus. Apart from this recoil energy, which varies from zero to several Mev depending upon the nature of nuclear disintegration, a nuclear decay is associated with thermodynamic effects due to change in the chemical nature of the daughter atom and charge effects due to shake off of electrons or internal conversion processes. If such a nucleus is a part of a molecule these effects can lead to large vibrational and rotational distortions and consequently bond rupture can occur. The extent to which bond rupture takes palce depends upon the available energy and hence on the mode of nuclear decay. Earlier accounts[1-7] and some recent review[8-15] are confined only to special areas related to the chemical effects of radioactive decay. The various radioactive decay processes and the related effects are briefly discussed in the following pages. Attempt has been made to include most of the important processes and their effects in this account.

Physical Effects

Beta Decay and Isomeric Transition

Beta decay is a process in which either a negative electron and an antineutrino, or a positron accompanied by a neutrino, are emitted from the nucleus of a radioactive atom. Isomeric transition occurs when as excited nucleus does not possess enough energy to overcome the binding energy of a nucleon and therefore decays to a state of lower energy either by the emission of a gamma ray, or ejection of an orbital electron by means of an internal conversion process. The factors on which the final chemical state of the daughter atom depends are as follows:

1. recoil of the decaying atom;
2. excitation and ionization of the daughter atom;
3. thermodynamic effects associated with the change in chemial nature of the decaying nucleus as a result of beta decay.

Recoil

Following the emission of a beta particle the daughter necleus recoils with an energy that depends upon the energy of the beta particle and upon the angular correlation between the beta particle and the neutrino. The maximum recoil energy is given by:

$$E_M^{\beta}(\max) = \frac{548\, E_{\beta}(\max)}{M} + \frac{536\, E_{\beta}^2(\max)}{M} \qquad (1)$$

(of $E_M^{\beta}(\max)$)

where E_{max}^{β} is the maximum recoil energy in electron volts and $E_{\beta}(\max)$ is the maximum beta particle energy in Mev and M is the mass of the recoiling atom in atomic mass units[16]. Recoil effect is likewise associated with the emission of gamma rays and conversion electrons. If E_M^{γ} is the recoil energy in electron volts, then, in the case of gamma ray emission, it is given by:

$$E_M^{\gamma} = \frac{536\, E_{\gamma}^2}{M} \qquad (2)$$

where E_{γ} is the energy of the emitted gamma ray in Mev and M is the mass of the recoiling atom in atomic mass units[16]. When the conversion electron is emitted the recoil energy[16] is given by:

$$E_M^{e} = \frac{541\, E_e}{M} \qquad (3)$$

Here E_e is the energy of the emitted conversion electron in Mev and E_M^e is the recoil energy in ev.

Excitation and Ionization

The processes responsible for excitation and ionization of atom in the event of beta decay and isomeric transition are listed below[2]:

(a) recoil of the daughter atoms
(b) shaking of the electron core of the atom as a result of sudden change in nuclear charge
(c) direct collision of the beta particle with an orbital electron
(d) vacancy cascades
(e) shaking of the electron cloud as a result of the rapid change of the electrostatic field when orbital electrons are ejected
(f) internal conversion of internal bremsstra hlung
(g) capture of the beta particle in an electron shell of the daughter nuclide
(h) internal pair conversion of internal bremsstra hlung.

The most important of these excitation processes are a, b, c, and d, and are now described in detail.

Recoil of Daughter Atoms

For most radioactive atoms undergoing beta decay or isomeric transition the recoil energy is insufficient to cause bond rupture.

Shaking Effect

Beta particles usually possess a high energy and hence leave the atoms in a time which is relatively short as compared to the period of orbital electron motion. Thus, a sudden change of nuclear charge is felt by surrounding cloud of electrons and their electrostatic environment of the electron cloud is disturbed. The electrons moving in a potential field created by the charge on the parent atom suddenly feel the field produced by the daughter nuclie whose positive charge is one unit more or less than that of the parent. Usually the cloud will contract adiabatically to accommodate itself to the increased nuclear charge in negatron decay, or conversely, expand adiabatically in positron emission, but sometimes ionization or excitation may result. A number[17-33] of theoretical discussions of the shaking effects have appeared in the literature so far. All of them are based on the concept that the sudden change in nuclear charge is responsible for the excitation and ionization of the daughter atom. According to these studies electronic excitation is more probable than ionization in the light nuclides and reverse is the case for the medium and heavy nuclides.

Direct Collision

The probability of excitation or ionization of an atom by direct collision between the emitted beta particle and an orbital electron has been described to be small by Migdal[18] and Feinberg[17]. However, Gerard[34] has shown that theoretically, excitation and ionization by direct collision can be comparable with shaking effect for the inner electrons of heavy atoms. Feinberg[35] in his later publication has shown that the direct collision between a beta perticle and an orbital electron plays a predominent role in the excitation of heavy atoms.

Vacancy Cascades

As a result of internal conversion and ejection of one orbital electron from K or L shell, a daughter atom is produced which is initially one unit more positive in charge than the parent. This creation of a vacancy or hole in the K or L shell of the daughter is very unstable and is filled by an electron from an outer less tightly bound shell. A smaller part of the excess energy is consumed in recoil of nucleus while major part is released as an x-ray or it is aquired by another electron through a non radiative process to cause the emission of an Auger[36] electron. This process produces an atom with a charge of 2 + and two holes in its electron shells. These vacancies in turn will be filled by electrons from shells which are less tightly bound, and again the excess energy will be compensated, either by x-ray emission, or Auger electron emission. Thus, a chain process is triggered off to yield vacancy cascades throughout the atom and a highly positively charged species is produced. A distribution of charge states is observed due to the possibility of may alternative electronic transitions for cascading the vacancies to the outermost shell of the atom. The net positive charge produced on the daughter is dependent upon the relative probabilities of Auger effect and x-ray emission. The Auger effect predominates for all the transitions in light atoms,[37-38] and in transitions outside the M shell for heavy atoms. The electronic transitions are not only possible in major shell but they similarly occur between the subshells and are known as Koster Kronig transitions[39]. In heavy atoms, the greater number of shells, and the possibility of Koster Kronig

transitions, provide many steps in which multiplication of the vacancies produced by the Auger effect is possible[40,41] and the daughter atom can accumulate a considerable positive charge in a time as short as 10^{-15} seconds[42].

Other Processes

The most important higher order processes in beta decay is the simultaneous emission of a beta particle with a photon and the neutrino. The emission of the photon known as internal bremsstra hlung is caused by the sudden change in the nuclear charge as a consequence of beta particle emission. The internal bremsstra hlung may be internally converted and transfer all or part of its energy to an electron, causing excitation or even ionization. Another nuclear porcess, which may be described as internal pair conversion of the internal bremsstra hlung, results in the simultaneous emission of four partilces, one neutrino and three electrons. However, both these effects of internal bremsstra hlung are of minor importance in connection with the excitation and ionization effects. Bound electron creation is said to occur in those nuclear transitions in which beta particles due to lower energy enter the bound energy levels of the daughter atoms.

Alpha Decay

Alpha decay is the emission of ^{4}He nuclei with energies characteristic of the decaying atom. As a result of the emission of aplha particles with different discrete energies, the daughter nucleus can be left in any one of several states of excitation. These excited states then lose their energy by emission of gamma rays, or by internal conversion. Accordingly, the atomic and moleucular effects following alpha decay may in some cases be partially masked by accompanying processes. If only the alpha emission is considered, the nuclear decay may be written as

$$ {}_{Z}X^{A} \rightarrow {}^{A-4}_{Z-2}(Y)^{2-} + {}^{4}_{2}He^{2+} $$

where X and Y are the parent and daughter atoms and Z and A are respectively the nuclear charge and mass.

Primary Effects

If the ejection of the alpha particle from the nucleus does not effect the electronic cloud, the daughter species should have a double negative charge. Whether or not the excess negative charge remains with the daughter will depend on the processes described below:

Electron Shake off

Alpha particle emission perturbs the electronic cloud around the nucleus and affects the electrons in the various shells to an extent depending upon their position with respect to the nucleus. The outer electrons are more likely to be excited or ionised than the inner ones as the velocity of alpha particles is more comparable to the electrons in the outer shells. A number of workers have tried to formulate the extent of ionization due to alpha decay. Their studies have been based on the following considerations[2]:

(1) Emitted alpha particle disturbs the surrounding electronic cloud and generates recoil momentum in the daughter nucleus. Both of these effects initiate ionization.
(2) decrease in the binding energy of the electrons, due to decrease of nuclear charge by two units will help ionization.
(3) the loss of two extra electrons possessed by the daughter atom will be resisted by the electron affinity of the daughter and thus electron affinity will be in a position to minimize the electron shake off effect.

Combining the effects of shaking and electron affinity (assuming both electrons to be bound) it has been theoretically predicted[43] that the daughter ^{218}Po species produced from alpha decay of ^{222}Rn will be left with a single negative charge in 40% of the atoms,neutral or positive in another 40% and with a double negative charge in the remainder of the decays. If one of the excess electrons is assumed to be bound, the charge spectrum should be 20% negative 1e, 40% positive 1e and 40% neutral.

Recoil

As a result of alpha particle emission the daughter

atom recoils. The recoil energy depends upon the energy of the emitted particle and the mass of the recoiling atom and is given by the equation[43].

$$E_M^{\alpha} = E_{\alpha} \frac{(4)}{M} \text{ Mev}$$

where E_M^{α} is the recoil energy, E_{α} the energy of the emitted alpha particle in Mev and M is the mass of the recoiling atom in atomic mass units; for ^{210}Po alpha decay, E_M^{α} = 103 Kev (E_{α} = 5.3 Mev) and for ^{241}Am decay E_M^{α} = 93 Kev (E_{α} = 5.477 Mev). Though the recoil velocities associated with these recoil energies will be high, they will be low as compared to the orbital electron velocity and will not be enough to eject the orbital electrons.

Charge Spectrum Studies of Nuclear Decay Products.

The average positive charge carried by the daughter species formed in the isomeric tranitions have been measured for a number of isotopes. Perlman and Miskell[44] obtained an average charge of + 8.5 ± 0.3 e for ^{131g}Xe Pleasonton and Snell[45] determined the charge spectrum of ^{131g}Xe using mass spectrometric methods and showed that the charge pattern had a symmetrical peak at + 8e and extended up to + 22e. Wexler[46] obtained an average charge of + 77 ± 0.4e for ^{80g}Kr produced by isomeric transition of ^{80m}Kr.

Snell and Pleasonton[47-49] have studied the charge spectra of the daughter atoms produced by the beta decay of the various isotopes of the inert gases, ^{6}He, ^{39}Ar, ^{85}Kr and ^{133}Xe. Their results showed that the singly charged ions, produced as a direct consequence of the emission of a beta particle, were the most abundant, and in 80 to 90% decay events the excitation energy was not enough to cause ionization. These results agreed with the theoretical predictions of Winter,[50,22] Carlson[24] and Green[21].

The maximum kinetic energies of recoil have been measured[51,52] for ^{88}Kr and ^{89}Kr. The experimental values of 115 ± 5 ev and 51.5 ± for ^{88}Kr and ^{89}Kr were not much different from the theoretical values of 120 ev respectively.

Rubinson[53] was the first to compile the results of various experiments which were carried out to estimate the electron shake off effect after alpha particle emission. The experimental results were based on the measurement of the characteristic X-rays of the daughter atom. Table 1 [54] shows the experimental and theoretical photon yields from ^{210}Po decay.

Table 1.

Experimental and TheoreticalPhoton Yields from ^{210}Po Decay

Shell	Experimental Yield	Theoretical Yield
K	1.5×10^{-6}	$0.2\text{-}0.3 \times 10^{-6}$
	2.0	0.136
	1.6	2.5
	1.5	2.5
L	2.2×10^{-4}	1.3×10^{-4}
	2.93	1.07×10^{-6}
	4	
M	1.5×10^{-3}	
	0.91	2.22×10^{-6}

It is evident from the table that the theory has underestimated the extent of photon yield as a result of alpha decay. Riou[55] also reviewed the experimental and theoretical data on internal ionization following alpha decay of ^{210}Po.

Mund[56] et al measured the charge distribution on the recoiling ^{218}Po produced from alpha decay of 3.8 day ^{222}Rn. The daugher ^{218}Po was collected at a reduced pressure (0.01 to 1 mm) on an aluminium or brass plate under an electric field of 3200 volts/cm. After the decay period was over the electrodes were analysed for the relative amount of active deposits in a comparative ionization chamber. The ratio of the number of recoil ^{218}Po atoms on the anode to the number on the cathode was compared with the ratio expected theoretically, from the field stregnth, distance between the electrodes, and diameter of collecting surfaces, for various assumed average charge states. The best fit was obtained when the mean charge on the recoil was taken as +2e.

The charge distribution of recoils from ^{230}U alpha decay has been investigated by Gunter et al.[57] They collected the daughter ^{226}Th activity on a negatively charged platinum plate in air atmosphere and measured alpha particle energies, recoil energy of the daguhter They found a charge distribution from zero to +2. Results of some other studies have been complied in the Table 2[58].

Table 2.

Charge distribution in the daughter atoms produced as a result of alpha decay[58]

Decay	Z Daughter	Relative abundance of the final charge				
		-2	-1	0	+1	+2
$^{241}Am \rightarrow ^{237}Np$	93	0	0	1	4	1
$^{226}Th \rightarrow ^{222}Ra$	88	0.001	0.001	1	2	1
$^{223}Ra \rightarrow ^{219}Rn$	86	0	0	9	8	1
$^{220}Rn \rightarrow ^{216}Po$	84	0.05	1	30	1	1
$^{212}Pr \rightarrow ^{208}Pb$	82	0	0	1	2.2	0.8
$^{212}Bi \rightarrow ^{208}Te$	81	0	0	1	-1	0

If we look at the table we find that the mean recoil charge changes from +1 to zero in the change from radium to polonium and relative abundance of the negatively charged recoil ions goes from zero to 3%. This shift in the charge distribution may be ascribed to the closed shell configuration of Ra^{2+}, Rn^{O} and Po^{2-}. The charge distribution in the daughters produced by alpha decay lies between the O to +2 range provided that subsequent transitions, as a result of internal conversion processes, do not occur. However, if alpha decay is followed by internal conversion of gamma rays, the created vacancy cascades to the outer shells via X-ray or Auger electron emission. Subsequently when several internal conversion processes are carried through, the finanal oxidation state of heavy atoms reaches beyond +30. Perrin and Dewieclawic[59], employed a time of flight measurement technique to study the charge spectra of ^{208}Te and reported an average charge distribution of 12 ± 1 for ^{208}Te. In another study perrin and Wieclawik[60] studied the alpha decay of ^{227}Th to ^{223}Ra and found that after a number of internal conversions the charge on the daughter ^{223}Ra reaches up to + 25. Wieclawik[61] also investigated the alpha decay of ^{241}Am and found that the daughter ^{237}Np acquired a maximum charge of + 33 as a result of internal conversion. Meyer et al[62] studied the charge states of ^{208}Pb and ^{208}Te formed from alpha decay of a solid deposit of ^{212}Po and ^{212}Bi. In the absence of internal conversion the charge distribution was from -1 to + 6, the mean charge being less than one. Alpha decay followed by internal conversion produced a charge spectrum lying between +5 to +20 with a mean chages of +12.

The Molecular Effects of Nuclear Decay.

As a result of nuclear decay, the daughter atom produced may lie in a state of excitation and ionization and will be associated with some kinetic energy. If the decaying atom is a part of a molecule, the process of deexcitation and redistribution of positive charge in molecule leads to bond rupture[63]. Further, if the nuclear decay has occurred by means of alpha or beta decay, it will result in the transmutation of one element into another. At the moment of formation the chemical environment around the daughter will be the same as that of the parent and it is possible that this species will be thermodynamically unstable. It may stabilize itself by fragmentation or by reaction with the surrounding medium and thus cause molecular disruption. Therefore, in order to distinguish between the effects of nuclear decay and change in chemical property, consideration is to be given to the chemical behavior of the daughter nucleus arising from the presence of other molecules in close vicinity of the radioactive atoms in condensed media. Processes such as exchange reactions, collision, deactivation, cage effects and recombination reactions may

determine the fate of the daughter atoms in condensed media.

Experimental Methods

A variety of techniques are used for the study of the chemical effects of nuclear decay. In the case of studies on isolated molecules "Charge and Mass" spectrometry is useful. For nuclear decay studies in condensed media any one of the methods such as "Mosbauer spectroscopy", Angular correlation Techniques", or conventional radiochemical methods may be employed.

Conventional Radiochemical Techniques

In the conventional radiochemical method of study, the daughter atoms produced by nuclear transformation are radiochemically analysed in their stable chemical forms. The raidoactive assay is performed by means of normal counting techniques. Experiments are designed in such a manner that the only measurement of the chemical effects of nuclear decay is possible.

Charge and Mass Spectrometry

The use of this method is restricted to volatile compounds and the daughter atoms produced as a result of nuclear decay, need not to be radioactive. Wexler, Carlson and Snell and pleasonton[25,26,47] used specially designed mass spectrometers to study the charges on the molecular fragments produced by molecular disruption caused by nuclear decay. The raidoactive molecules in the gas phase are introduced into the spectrometers at low pressure in order to ensure that only the primary products are recorded. The time of flight of the ions in the spectrometer is of the order of 10^{-4} to 10^{-5} seconds, thus only the electronically and vibrationally stable species reach the detector of the spectrometer and the radiative transitions, and atomic virations which a time of the order of 10^{-8} and 10^{-14} seconds respectively, are not recorded.

Mossbauer Spectroscopy[64,67]

This method provides a techniques for studying the chemical effects within short time after the nuclear transformation. The charge distribution on the daughter atom is detected by measuring the chemical shifts and information about the life time of the valence states may also be obtained from broadeing of the resonance absorption peaks. The quardrupole and magnetic fine structures in the Mossbauer spectrum can give information about the enviromant of the daughter atoms, while the difference in the recoil free fraction in source and absorber of the compound helps to find the degree of molecular disruption. However, this technique can be used only in those cases where recoilless emission of gamma rays is possible.

Angular Correlation

When a nucleus decays be successive emission of two particles, such as a beta particle followed by a gamma ray, or two gamma rays one after another, there is no preferred angle between the radiations. This random emission of the particles is called as isotropic. Deviation from this isotropic behaviour is measured by the anisotropy, given as:

$$A = \frac{W(180^{o})}{W(90^{o})} - 1 \qquad (1)$$

where $W(180^{o})$ or $W(90^{o})$ are the number of events per unit solid angle which have an angle of 90^{o} or 180^{o} between the two radiations. If there exists an interaction between the quadrupole moment of the nucleus and the electric field around it, it will suppress the anisotropy peculiar to the nucleus. Thus the measurement of the decrease in anisotropy can be used to investigate the chemical environment of the daughter following the nuclear decay event.

Typical Experimental Results.

Isomeric Transition

In spite of the low recoil energies associated with Isomeric transition, the atoms produced as a result of this decay have been found to be separated from their parent molecules. Thus the decay of ^{80m}Br to ^{80g}Br state was found to break the C-Br bond in methyl bromide molecule by Hamill and Young[68]. Studies of De Vault and Libbey[69] showed that ^{80m}Br decay decomposed to the molecules of EtBr, (CH_2Br_2), MeCHBrCHBrMe, $(CH_3)_3$ C-Br, C_6H_5Br, BrO_3^- and Br_2 in the vapour phase.

A number of studies have been made in the chemical effects of the isomeric transition of ^{80m}Br to ^{80g}Br. A large variety of ions have been reported to exist as a result of ^{80m}Br to ^{80g}Br transition in CH_3Br[70] CCE_3Br[71]. The chemical effects of the ^{80m}Br in the hexabromo complexes of Re, Os, Ir and Pt were examined by Schmidt and Herr.[72] They could obtain very high (90%) yield of free bromide ^{80g}Br ions in acidic solution whereas the yields were very meagre in the solid state. The chemical effect of some other in-organic ^{80m}Br complexes[37.74] have also been studied. There have as well been some studies reported on the chemical effects of the isomeric transformation of ^{82m}Br to ^{82g}Br[75-78] and in a number of tellurium compounds[79-92] by using ^{127m}Te, ^{129m}Te and ^{131m}Te isomers. Nefedov and Sinotova[93] found that the amount of break up following an isomeric transition did closely correspond to the internal conversion coefficient of the decaying isomer and only 5% break out of the ^{69g}Zn-$(C_2H_5)_2$ was obtained, produced as a result of isomeric transition of ^{69m}Zn $(C_2H_5)_2$ in an ethyl acetate solution.

Nefedov and Sinotova have also reported[94] the disruption of the diethyl mercury compound following the isomeric transitions in ^{195m}Hg. Herr[95] reported the separation ^{104g}Rh following decay of ^{104m}Rh in rhodium phthalocyanine, and Simpson[96] observed the break out of ^{58g}Co from ^{58m}Co labelled tris (ethylendiamine) cobalt (III) nitrate following 88.7 ± 9.7% of the decays in the solid state and 51.8 ± 10.8% in solution. The chemical effects of isomeric transformation of Br^{80m} in Br^{80m} tagged $KBrO_3$ crystals have been reported by Serano.[97]

Glentworth and Wright and Abernethy[98,99] have studied the chemical effect of isomeric transition of ^{114m}In, ^{69m}Zn and ^{87m}Y when these isomers were complexed with a series of polyaminocarboxylic acid ligands in aqueous solutions. They found a close relationship between the internal conversion coefficients and the percentage break up values. Chemical effects of isomeric transition $^{81m}Sc \rightarrow ^{81g}Se$ in potassiumselenate have also been reported[100]. The fragmentation was found to be low (19%) in crystals as compared to solutions (52%). The fragmentation of solid cobalt phenyl phosphine complex has been found to be low following the transitions of the type $^{57m}Co \rightarrow ^{57g}Co$ and $^{60m}Co \rightarrow ^{60g}Co$[101]. Recently isomeric transtion of Bromine (80m) in H-^{80m}Br-CH_4 system has been published[102].

4.2 Beta Decay Effect.

Tritium

The chemical effects of beta decay of tritium in the hydrocarbons[103-108] has been extensively studied. It has been found that the nuclear decay almost invariably breaks the carbon-helium bond in the compounds like CH_3T and C_2H_5T, the positive charge is transferred to the organic fragment and ^3He is released as a netural atom. The organic fragment remains intact in 80% of the cases otherwise further fragmentation takes place to yield organic segments with fewer hydrogens.

Carbon 14

Snell and Pleasonton[109] found that the nitrogen atom produced as a result of beta decay of the ^{14}C in $^{14}Co_2$ remained bound as $(^{14}No_2)^+$ following 81.4% of the decays, whereas Wolfgnag et al[110] studied the chemical effects of beta decay of ^{14}C in ethane and in 47 ± 2% of the events they found that C-N bond in CH_3NH_2 was preserved. Manning and Monk[111] showed that beta decay of ^{14}C in the methyl group of toluene failed to produce any bond rupture.

Halogens

Chemical effects of beta decay in a number of halogen compounds have been reported[112-116]. Molecules like C_2H_4 Br ^{82}Br and ^{82}Br Cl_3 have been studied. As in the case of the beta decay of tritium, the initial fragment ions like $C_2H_5Br^+$ predominanted the spectra of the organic derivative. Carlson and White[112] found different break up values for beta decay in CH_3 ^{130}I, CH_3 ^{131}I and CH_3 ^{82}Br. They ascribed the high survival rate of CH_3 ^{130}I, CH_3 ^{131}I and CH_3 ^{82}Br. They ascribed the high survival rate of CH_3 ^{131}Xe (96%), compared with CH_3 ^{130}Xe (34%) and CH_3 ^{82}Br 0.43% to its lower recoil energy, 0.22ev compared with 0.78ev and 2.45ev respectively.

Organometallic Systems

As a consequence of beta decay, bond cleavage has been reported in a number of organometallic compounds of ^{210}Pb, ^{210}Bi, ^{125}Te and ^{125}Sb. For the general review see the references[117-131].

Inorganic Systems

The consequences of beta decay inorganic systems have been extensively studied. The first paper of any significance was that of Goryell et al[132] who showed that 60% of the ^{143}Ce produced as a beta decay product of ^{143}La, was in tetravalent oxidation state. However the result was not of much importance because a rapid electron exchange between $^{143}Ce^{4+}$ ions and Ce^{3+} ions a common impurity in La^{3+}, would give $^{143}Ce^{3+}$ Davies[133] found that the yield of bromine as bromate ion produced by the beta decay of ^{83}Se and ^{84}Se was about 35% and did not depend significantly upon whether the parent ions were selenite or selenate and the breaking of the parent selenate or selenite ions was suggested. However Davies found that the chemical form of tellurium affected the yield of ^{132}I formed by the beta decay of ^{132}Te in tellurium and tellurite ions and it was suggested that the breakdown of ions might be caused by internal conversion effect. cumminsky et al[134] found that 90% of beta decays from ^{132}Te produced bond rupture but the nature of the iodine species formed, such as periodate or reduced forms of iodine, depended upon the nature of the parent ion and the pressnce of oxidising and reducing agents in the solution. Gordon[135] reported that ^{132}Te when decaying from tellurite form produces very excited ^{132}I positive ions. Halpern[136] showed that the decay of ^{132}Te in toluene produced more iodine as methyl iodide, iodobenzene and polymers and compared to ^{131}Te decay. The reason for this was attributed to the difference in electronic excitation and ionization between the daughter products. Adroff and Lladabor[137] found that about 40% of the ^{132}I formed in butyl alcohol solutions of $^{132}TeCl_4$ reacted with orgnic fragments to give alkyl iodides. They also found that the yield of alkyl iodide formed by the beta decay of ^{132}Te in solutions of Ph_2 ^{132}Te increased in the solid phase.

Petrukhin et al[138] observed the charge distribution of ^{239}Np produced from beta decay of ^{239}U in $^{239}UO_2$. 90% of the daughter was found ^{239}Np (IV) showing that the majority of the beta decay events had resulted in the Neptunium oxygen bond rupture. This was thought to be the result of excitation and ionization produced by the associated gamma rays and the shaking effects due to the sudden change in nuclear charge. Recoil was not to play any part in the disruption.

The chemical effects of beta decay of ^{144}Ce acetylacetonate were observed by Coryell and Edwards[139]. They found that the daughter ^{144}Pr could only be extracted into water form the carbon tetrachloride solution of the complex if the aqueous phase contained 10% acetylacetone. The authors suggested that the contact with acetylacetone caused the disruption of the activated that the contact with acetylacetone caused the disruption of the activateded complex species otherwise it got stabilized by charge exchange.

The chemical effects following the ^{144}Ce beta decay in the ^{144}Ce EDTA complex in solution were followed by Cendales et al.[140] They found that most of the ^{144}Pr daughter was in free ion form and suggested that the recoil energy was responsible for the molecular break up. Studies of the chemical effects of the beta decay of ^{144}Ce complexes with a number of polyamino carboxylic acid ligands have been carried out by Glentworth, Wiseall and Wright[141,142]. They found that the percentage break up of the daughter ^{144}Pr complex was highly dependent upon pH and thermodynamic stability of the complex. The authors suggested that the nuclear decay produced an excited species which could disrupt by hydrogen ion attack.

The chemical effects of the beta decay of ^{144}Ce in ^{144}Ce EDTA, ^{144}Ce DCTA in solution have also been investigated by Shiokawa et al.[143-146] According to them exhaustive bond rupture produces ^{144}Pr (III) in 25% decay events, as a result of internal conversion and electron shake off processes without any formation of an excited intermediate. In the cases where multiple charging does not occur, an excited intermediate complex is formed and is consequently attacked by hydroxide ions to form free $^{144}Pr^{3+}$ ions. Shiokawa et al[147] have also studied the chemical effects of the ^{144}Ce beta decay in solid crystals of ^{144}Ce EDTA. They found that the yield of $^{144}Pr^{3+}$ ions depended upon the amount of hydration in the crystal. The authors proposed that the internal conversion and electron shake off was responsible for multiple charging of the daughter atoms and hence for the molecular disruption.

Glentworth and Wright[142] have also studied the chemical effects of beta decay in ^{143}Ce in ^{143}Ce DTPA. They found that the two isotopes behaved differently. ^{143}Ce complex decayed to give 89% free ions of the daughter ^{143}Pr whereas ^{144}Ce DTPA decay resulted in a pH dependent break up which varied between 30.9 to

66.3%. The lower percentage break up values observed for ^{144}Pr in the same chemical environment was attributed to the lower probability of formation of multiply charged daughter atoms in ^{144}Ce decay. The marked dependence of the ^{144}Ce system on pH was thought to be caused by the presence of a high proportion of reactive daughter species which do not immediately decompose and have a finite life time.

Glentworth and Abernethy[99] studied the chemical effects of beta decay of ^{115g}Cd in ^{115g}Cd DCTA. The They found that only 18.8 ± 2% of the produced ^{115m}in atoms were in free ion form and concluded that only the multiply charged atoms produced by internal conversion process could result in break up and that the recoil energy was insufficient to cause the molecular diaruption.

Asano et al[148] have published their results of the studies on chemical effects of beta decay of ^{171}Er and ^{177}Yb in the Er and Yb, DCTA complexes. They found the occurrence of 70% bond rupture in ^{177}Yb decay and 100% in ^{171}Er decay at pH 6.5. The increase of pH favoured the break up in the ^{177}Yb DCTA system and the addition of sodium thiosulphate resulted in the decrease of percentage break up in the both systems. The authors have interpreted their results on the concepts of multiple ionization, recoil of the daughter and the proton attack mechanism. The decrease in the percentage break up by the addition of sodium thiosulphate has been attributed to its reducing effect.

Ikuta et al[149] have estimated the theoretical yields of beta decay in tritium labelled alkanes. Beta decay effects in C_5H_5 $^{99}Mo(Co_3)$ have been reported by Dejong and Wiles[150]. They found that in 74% of the decay events the daughter ^{99m}Tc remains bound to the parent molecule and a minimum of 10ev are required to break a molecule in solid state following the nuclear event. Tenorio et al[151] found that the beta decay of ^{83}Se in potassium slenate produces Br 48%, BrO_3 28% and BrO_4 24%.

Ikuta[152] published his theoretical estimates of beta decay effects in CH_3 ^{18}F molecule. He suggested that the final products will be formed as a consequence of recoil and electron shake off effects.

The rupture of metal-DCTA bond associated with beta decay of ^{171}Er and ^{177}Yb-DCTA complexes has been reported by Asano and Taniguchi[153]. This has been studied as a function of three variables mainly concentration of free ligand ions i.e. DCTA itslef, the glycerol content and the temperature. In this study, which is the only in non-aqueous medium, the low break up values have been reported and they are interpreted in terms of increased viscosity of the solution which seems to lessen the distance of separation of the geminate fragments and as a consequence recombination is favoured.

Asano and Taniguchi[154] have also reported the beta decay effects of ^{144}Ce-DTPA in frozen solutions. They proposed that the recombination of geminate fragments, produced following beta decay of the ^{144}Ce-DTPA complex, stay in excited states until the solution is forzen and the fates of these excited species are effected by the possible successive reactions as the liquefaction of the frozen solution proceeds. On the basis of chemical effect studies of five different Ce isotopes complexed with polyaminocarboxylic acid ligands, Beyer et al[155,156] discussed the mechanism of bond cleavage following nuclear transformations in these complexes. They report that almost 100% ruputre of the chemical bonds is obtained as a result of electron capture decay and gamma ray conversion processes. In 20% of the gamma ray disintegration and beta decay processes the bond rupture is by means of shake off effects. The process is independent of the nature of complexing ligand. In remaining 80% the break up of the bonds is not immediate, but firstly the daughter changes its oxidation state from +4 to +3 and then these reduced daughter ions leave the complex by means of metal exchange processes which depend upon the solution conditions and the kind of ligand involved.

4.3 Alpha Decay.

The chemical consequences of alpha decay have not been extensively studied. Haissinsky[157] and Cottin found only 50 to 80% of ^{234}Th as free ions from uranyl bensoyl acetonate following alpha decay of ^{238}U and the yield was even reduced to 10% in solid form. For this transformation the recoil energy is 71 Kev. They also studied the chemical effects of ^{238}U decay, in uranyl benzoylmethane solution and could find only 44% of the produced daughter in the free ion form, the yield was further reduced to 18% in the solid phase. These authors suggest that cage recombination is responsible for the less than expected molecular disruption. P. Glentworth and Naqvi, the author,[158] have recently published their studies on the chemical consequences of

alpha decay in ^{235}U ions complexed with ligands, EDTA. DCTA and DTPA. They found low yields of daughter ^{234}Th fragments and have proposed that recombination of fragments in solvent cage is responsible for less break up of the Th ligand bond.

Summary: The experimental and theoretical results discussed in this review indicate that the major causes of the molecular break up due to the nuclear decay are recoil, the changing effects caused by shake off and internal conversion events, and thermodynamic instability of daughter in the parents chemical environment. However. the efficiencies of these various modes of excitation in producing molecular break up are still open to some speculation. The chemical effects of nuclear transformation not only vary with the mode of decay but they also depend upon the chemical environment and chemical form of the decaying isotope.

Alpha particle emission accompanies a complex decay scheme. It not only produces highly energentic recoiling daughter atoms but it may also be associated with electron shake off and internal conversion processes. Unlike beta decay the chemical consequences of alphas decay have not been extensively studied.

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