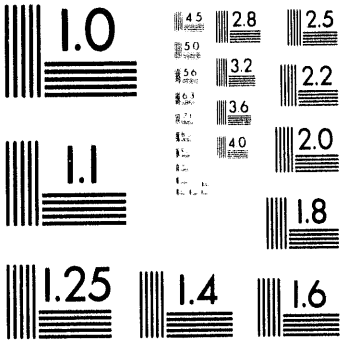
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Organic Chemical Aging Mechanisms: An Annotated Bibliography

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MASTER

Executive Summary

An annotated bibliography has been compiled of the potential chemical and radiological aging mechanisms of the organic constituents (non-ferrocyanide) that would likely be found in the UST at Hanford. The majority of the work that has been conducted on the aging of organic chemicals used for extraction and processing of nuclear materials has been in conjunction with the acid or PUREX type processes. At Hanford the waste being stored in the UST has been stabilized with caustic. The aging factors that were used in this work were radiolysis, hydrolysis, and nitrite/nitrate oxidation. The purpose of this work was two-fold: to determine whether or not research had been or is currently being conducted on the species associated with the Hanford UST waste, either as a mixture or as individual chemicals or chemical functionalities, and to determine what areas of chemical aging need to be addressed by further research.

The findings of this search are:

- Reported work has encompassed neither the chemical complexity of the tank waste nor the scope of issues necessary to determine the mechanisms or products from aging.

- The search revealed relevant studies on two major waste components: two studies on the alkaline hydrolysis (Burger 1955) of TBP and one example of radiolysis of NPH under alkaline (Shcherbak 1960) conditions.

- Pathways are enumerated for destruction of the organic compounds, but vagaries due to lack of detailed information about the actual past and present conditions in the tanks (i.e., NO_x and oxygen gas contents) and interfacial effects limit our ability to make conclusions about the degree of degradation and their on associated safety issues.

- Additional research directed towards elucidating the effects of radiolysis on organic constituents in alkaline conditions as well as the synergistic and inhibitory effects of oxide surface-interactions, metal ion catalysis, mass transfer, solid-liquid interfaces, high concentrations of nitrate and nitrite would be helpful in addressing safety concerns. Most useful would be knowledge of the current state of tank contents obtained by thorough sampling of the problematic tanks.

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1.0 Introduction and Approach

The chemical literature has been reviewed to obtain information on chemical mechanisms that operate to transform or age organic constituents of the underground storage tanks (UST) on the Hanford site (Gerber 1992). This effort is part of a larger program at PNL to assess issues concerning the safety of storage tank wastes. Knowledge of the fate of organics in the tanks will help determine the extent to which hazards such as waste deflagration have increased, decreased, or remained unchanged since additions of the organics to the tanks.

No examples were found of any groups or individuals attempting to determine the chemical aging of wastes similar to those in the Hanford UST. Some work has been specifically targeted at these compounds but it has been concerned almost exclusively with degradation pathways in 3 M or greater nitric acid. Aging mechanisms of ferro/ferrocynides and the EDTA type complexants were not addressed in this effort because other investigators have recently examined the fate of these constituents (Lilga 1992; Meisel 1991; Meisel 1991).

Initially, the report was to consist of only an annotated outline of the pertinent references for all of the major and minor chemicals/chemical functional groups thought to be present in the UST. The dearth of information on the radiolysis of NPH/alkanes under alkaline conditions and a request for description of aging mechanisms of NPH and hexone for presentation at a technical meeting (Babad 1993) led to a descriptive review of possible pathways for the oxidation of NPH under conditions expected in the waste tanks. Three types of chemical processes that can transform the organic components are considered: gamma-radiolysis, nitrate/nitrite oxidation, and alkaline hydrolysis. The reference data bases -- Nuclear Sciences covering 1948-1976, Engineering Science & Technology covering 1974 to present, and Chemical Abstract Service from 1967 to present -- were electronically searched for the primary chemical or chemical functionality versus the factor that was to cause chemical aging. Furthermore, Chemical Abstracts from Volume 41 to the current volume were searched manually.

In the subsequent sections of this document we review the literature and give details of the specific reaction mechanisms that are potentially at work in the UST. The information is provided in three sections. Section 2.0 is a review and discussion of possible reaction pathways for NPH. Section 3.0 is an outline of the relevant literature pertaining to the reactions of othermajor chemical constituents. Section 4.0 addresses the minor chemical constituents. This information is grouped according to the following compound classes: normal paraffin hydrocarbons, hexone, tributyl phosphate, and minor constituents including ethers, esters, alcohols, carbohydrates and chlorohydrocarbons. Subsequent sections provide a list of cited references with abstracts and a bibliography of all the literature reviewed in the preparation of this document.

2.0 Aging Mechanism of Normal Paraffin Hydrocarbons (NPH): Possible Reaction Pathways

Drawing upon limited experimental data and a diverse array of literature, a case can be made that organic chemicals of the types utilized in waste management and chemical processing are, in general, not stable in the alkali radioactive environment of the tanks with respect to degradation products or other adducts. In most cases these reactions appear to take place slowly, but certain reactions appear to be quite rapid.

Radiolysis of water and nitrates generates reactive radicals that attack organic and inorganic species making organic radicals, inorganic radicals, and higher valent metal ions (except for solvated electrons and H• which reduce many transition metals to lower valent states). (Daniels 1969; Broszkiewicz, Kozlowska-Milner et al. 1981; Neta and Huie 1986; Buxton, Greenstock et al. 1988) Some conditions reduce the role of e- and H•. Oxygen (Milosavljevic and Micic 1978; Buxton, Greenstock et al. 1988; Ross, Mallard et al. 1992) and gaseous nitrogen oxides scavenge e- and H• at near diffusion limits (Knight and Sutton 1967), so if tanks are air saturated or have dissolved NO_x gases of nitrogen oxides (Warman 1967; Seddon and Young 1970; Janata and Schuler 1982), then these reactions should be significant.

$$NO_x\bullet + e^- \rightarrow NO_x^-$$

$$O_2 + e^- \rightarrow O_2^-\bullet$$

$$NO_2\bullet + H\bullet \rightarrow NO\bullet + HO\bullet$$

$$O_2 + H\bullet \rightarrow HO_2\bullet$$

Products so produced are of intermediate oxidation states and may be further oxidized or reduced depending on their reactions with other constituents in the tanks. For example, species such as O_2^- and $HO_2\bullet$ both reduce and oxidize transition metals such as Fe(III) (Rush and Bielski 1985) while oxidizing many classes of organic compounds (Buxton, Greenstock et al. 1988; Ross, Mallard et al. 1992).

Numerous competing reaction pathways are feasible; which pathway will dominate depends on the conditions in the tanks. These pathways are illustrated in Figures 1 and 2. Initiation of redox chain oxidations and radical-induced crosslinking by direct radiolysis of organic phases will receive serious evaluation as part of the program to resolve the organic waste safety issue at the Hanford site (Babad and Johnson 1992).

The very low solubility of NPH in high ionic strength aqueous solutions suggests that they will not be oxidized much in the bulk aqueous phase. Instead, NPH are expected to degrade via radiolytic processes that occur in the organic phase and at interfaces between aqueous and organic phases. As much as 5-10% water is solubilized in the organic phase by polar organics such as alkyl phosphates. This will result in the generation of some HO and H radicals being generated in the organic phase by radiolysis. In their critical review of rate constants for reactions radicals derived from radiolysis of water, Buxton et al. point out that the amount of energy absorbed by any component of a solution exposed to high energy radiation is proportional to the electron fraction of the component. This means that direct radiolysis of the organic components will likely be a dominant pathway for initiating NPH aging pathways.

Radiolysis of neat saturated hydrocarbons produces saturated hydrocarbons of both higher and lower carbon numbers, hydrogen and olefins (Bugaenko, Kuzmin et al. 1993). The products show a dependence on hydrocarbon structure with branched hydrocarbons yielding greater amounts of light hydrocarbons and lesser yields of hydrogen. Unsaturated hydrocarbons give predominantly condensed and polymer products; yields of hydrogen are much smaller than from saturated hydrocarbons, as well. Initially-formed transient species ($< 10^{-16}$ sec) include electrons, NPH radical cations, and excited states (Abell and Funabashi 1973; Avdonina 1975; Bugaenko, Kuzmin et al. 1993). Subsequent reactions that ensue include charge recombination, fragmentation of ions, ion molecule reactions, degradation of excited states, radical molecule reactions, and radical combination reactions (Bugaenko, Kuzmin et al. 1993). The $NPH^{+\bullet}$ is initially formed with excess energy that facilitates fragmentation to radicals and ions (occurs within 10^{-12} sec after ionization) (Bugaenko, Kuzmin et al. 1993). Once $NPH^{+\bullet}$ has attained thermal equilibrium, deprotonation to NP• radical and protons is energetically feasible. For example, fragmentation of butane radical cation is substantially endothermic but hydrogen atom transfer as shown between ethane radical cation and ethane is thermoneutral (Lias, Bartmess et al. 1988).

$$C_4H_{10}^{+\bullet} \rightarrow C_2H_5^+ + C_2H_5\bullet \qquad \Delta H^\circ \approx 31 \text{ kcal/mol}$$

$$C_4H_{10}^{+\bullet} \rightarrow C_4H_9^+ + H\bullet \qquad \Delta H^\circ \approx 42 \text{ kcal/mol}$$

$$C_2H_6^{+\bullet} + C_2H_6 \rightarrow C_2H_7^+ + C_2H_5\bullet \qquad \Delta H^\circ \approx 0 \text{ kcal/mol}$$

In the absence of scavengers, NP• radicals combine to form branched or cross-linked NPH and disproportionate to form alkanes and olefins (Pancini 1970; Tanaka 1974; Avdonina 1975; Seguchi 1985; Tilquin 1988; Seguchi 1991; Tilquin and Baudson 1991). Cleavage of alkyl chains via β-scission of thermalized alkyl radicals does

not occur because these reactions are endothermic by 30 to 40 kcal/mol (Frey and Walsh 1969; Benson and O'Neil 1970; Kerr 1977). The relative yield of disproportionation to combination products from termination of radicals depends on structure, with recombination generally favored for primary and delocalized radicals (Trotman-Dickenson and Milne 1967; Ingold 1973).

Conventional pathways for production of hydrogen include combination of H atoms and abstraction of H from NPH by H•. However, hydride ion (H^-) may also be produced during the very early stages of the ionization process via Dissociative-Attachment processes. If so, their reaction with proton sources would also lead to H_2 production. Studies of the Dissociative-Attachment phenomenon show that low energy electrons (0 to 20 eV) impacting hydrocarbon films produce H^- and to lesser extent C_1-C_3 anions (Rowntree, Parenteau et al. 1991). The mechanism is thought to consist of an initial resonant electron capture by the neutral hydrocarbon molecule followed by direct dissociation of the anion into neutral and anionic fragments.

If oxygen is present in sufficient concentrations in the tanks, then scavenging of radicals and chemistry analogous to autoxidation reactions are possible. Autoxidation pathways produce hydroperoxides initially (Ingold 1968; Howard 1970), but continued reaction of these compounds yields alcohols, ketones, and acids (Ingold 1968; Hawkins 1973; Kochi 1973). Radical chain lengths are short for primary and secondary alkane C-H and therefore oxidation is inefficient because attack is slow on these inactivated C-H bonds. Attack on these bonds is a mildly endothermic process as evidenced by the bond strengths of alkanes, which typically range from 96 to 101 kcal/mol, compared to that of the product hydroperoxide, which has a bond strength (RO_2-H) of ~87 kcal/mol [Ingold 1968; Lias, Bartmess et al. 1988]. Self-termination reactions of peroxy radicals and cross termination of peroxy and alkyl radicals ought to dominate product formation depending on the amount of oxygen in the organic phase.

Organic layer saturated with O_2: $2R_2HCO\text{-}O\bullet \rightarrow R_2HCOH + O_2 + R_2C{=}O$

Organic layer depleted in O_2: $R_2HCO\text{-}O\bullet + R_2HC\bullet \rightarrow R_2HCO\text{-}OCHR_2$

C-H bonds tertiary, aldehydic, or α to activating groups such as OH, OR, C=C, Aryl, etc. are more susceptible to attack, but carbonyl groups deactivate neighboring C-H toward attack by electrophilic oxygen centered radicals. Therefore, ketones and acids are plausible products of aging via autoxidation mechanisms. Such products were observed during radiolysis of H-C (Tanaka 1974). In strongly basic media, these products will be in equilibrium with more reactive carboxylate anions and enols such that even these compounds may not be viewed as inert to the conditions in the tanks. Attack

on carboxylate anion by one-electron oxidants such as NO_3 leads to decarboxylation. Enols are susceptible to attack by both addition or electron transfer pathways.

$$RCO_2^- + NO_3\bullet \rightarrow NO_3^- + RCO_2\bullet \rightarrow R\bullet + CO_2$$

$$RCH{=}CHOH + O^{-\bullet} \rightarrow RC(OH)H{-}CH{=}O^{-\bullet}$$

$$RCH{=}CHOH + NO_3\bullet \rightarrow [RCH{=}CHOH]^{+\bullet} + NO_3^-$$

NO_x compounds also participate in autoxidation reactions. NO converts peroxides and peroxy radicals to alkoxy radicals and NO_x radicals react with alkyl, alkoxy and peroxy radicals, forming alkyl nitrates and carbonyl compounds (Carter, Darnall et al. 1976; Carter, Darnall et al. 1979).

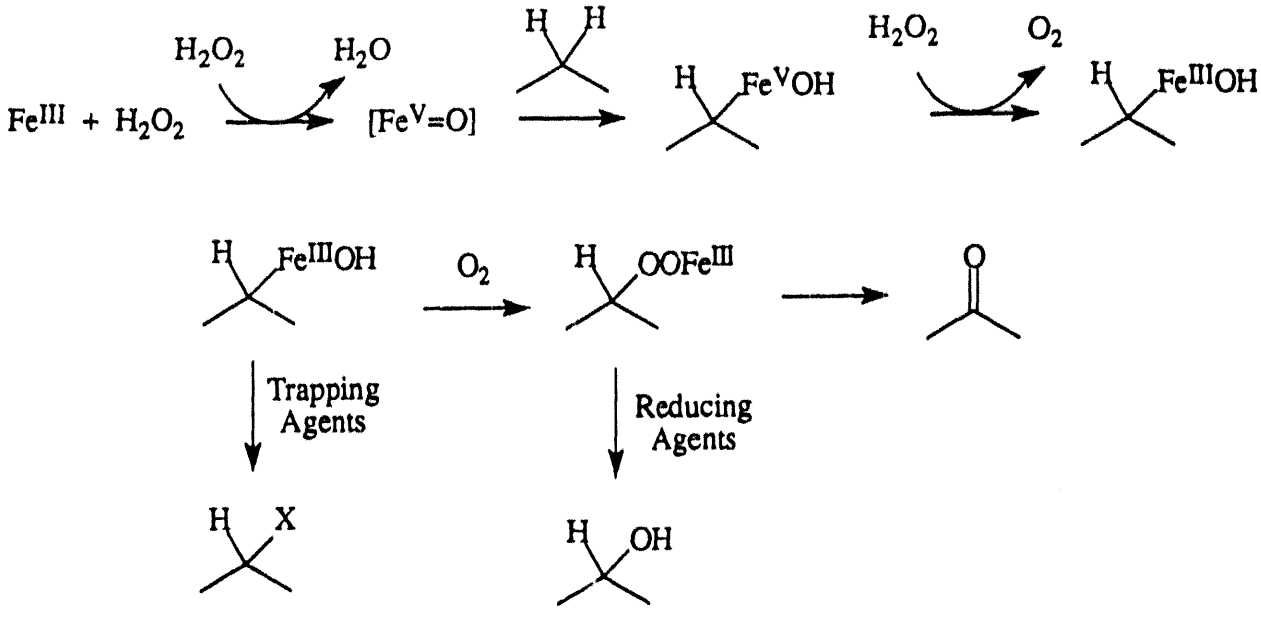
Trace organic-soluble transition metal complexes catalyze autoxidations. Conventional autoxidation are accelerated because hydroperoxides are reduced forming chain-propagating alkoxy radicals (Kochi 1973; Sheldon and Kochi 1981).

$$R_2CHO_2\bullet + M^{+n} \rightarrow R_2CHO_2^- + M^{+n-1}$$

$$R_2CHO_2^- + M^{+n} + H^+ \rightarrow R_2CHO\bullet + M^{+n-1} + OH^-$$

Alkoxy radical intermediates can yield fragmented products via β-scission (Kochi 1973) and long chain alkanes can be oxidized to tetrahydrofurans and bifunctionalized products due to facile 1,5-H atom shift reactions of alkoxy radicals (Kochi 1973; Carter, Darnall et al. 1976). Recent studies of metal-catalyzed oxidations suggest that reactions of metal ions with dioxygen or peroxidic compounds may form high-valent metal-oxo species that are capable of oxidizing even inactivated C–H bonds such as those in alkanes (Barton and Doller 1992), see Scheme 1.

Finally, certain transition metal ions [e.g. Cu(II), Fe(III), Mn(III), Co(III), Pb(IV)] can oxidize radicals such that even if oxygen concentrations are low, NPH could be oxidized via a redox chain reaction, instead of crosslinked (Kochi 1973; Sheldon and Kochi 1981). Oxygen, hydroperoxides and peroxides are needed to propagate these reactions by recycling lower valent metals to upper valent states while generating attacking radicals. Alternatively, in the tanks, if lower valent metals are transported to aqueous phase, then they would be oxidized by aqueous phase radiolysis intermediates (e.g. $O^-\bullet$, $NO_3\bullet$, H_2O_2). Transport back to the organic phase would propagate the redox chain reaction.


H_2O_2
H_2O
Fe^{III} + H_2O_2
$[Fe^{V}=O]$
H H
H $Fe^{V}OH$
H_2O_2
O_2
H $Fe^{III}OH$
H $Fe^{III}OH$
O_2
H $OOFe^{III}$
O
Trapping Agents
Reducing Agents
H X
H OH

Scheme 1.

3.0 Outline of the Literature Relevant to Aging of the Major and Minor Organic Constituents in USTs

3.1 NPH

3.1.1 Radiolysis

A. Radiolysis of neat saturated hydrocarbons produces saturated hydrocarbons of both higher and lower carbon numbers, hydrogen ($G=3.3$ at $55^{0}C$) and olefins (Bugaenko, 1993; Seguchi, 1991).

1. The products show a dependence on hydrocarbon structure; branched hydrocarbons yield greater amounts of light hydrocarbons and lesser yields of hydrogen.

2. Unsaturated hydrocarbons give predominately condensed and polymer products; yields of hydrogen are much smaller than from saturated hydrocarbons, as well.

B. Initially-formed transient species ($<10^{-16}$ sec) include electrons, NPH radical cations, and excited states (Abell, 1973; Avdonina, 1975; Bugaenko, 1993).

1. Subsequent reactions that ensue include charge recombination, fragmentation of ions, ion molecule reactions, degradation of excited states, radical molecule reactions, and radical combination reactions (Bugaenko, 1993).

2. The $NPH^{+}\bullet$ is initially formed with excess vibrational energy, which facilitates its fragmentation to radicals and ions (occurs within 10^{-12} sec after ionization) (Bugaenko, 1993).

$$C_4H_{10}^{+}\bullet \rightarrow C_2H_5^{+} + C_2H_5\bullet \quad \Delta H^{\circ} \approx 31 \text{ kcal/mol (Lias, 1988)}$$

$$C_4H_{10}^{+}\bullet \rightarrow C_4H_9^{+} + H\bullet \quad \Delta H^{\circ} \approx 42 \text{ kcal/mol (Lias, 1988)}$$

3. Once $NPH^{+}\bullet$ has attained thermal equilibrium, hydrogen atom transfer to $NP\bullet$ radical and protons is energetically feasible.

$$C_2H_6^{+}\bullet + C_2H_6 \rightarrow C_2H_7^{+} + C_2H_5\bullet \quad \Delta H^{\circ} \approx 0 \text{ kcal/mol (Lias, 1988)}$$

4. In absence of scavengers, NP• radicals combine to form branched or cross-linked NPH and disproportionate to form alkane, olefin or polymers (Avdonina, 1975; Pancini, 1970; Seguchi, 1985; Seguchi, 1991; Tanaka, 1974; Tilquin, 1988; Tilquin, 1991; Seguchi, 1991; Seguchi, 1991).

5. NP• radicals that contain small amounts of ketone groups (LDPE's) increase the efficiency of cross-linking up to 1% by weight and increased main chain scission by a factor of two (Bakh, 1972).

6. β-scission of alkyl radicals is endothermic by 30-40 kcal/mol (Kerr, 1970; Frey, 1969; Benson, 1970).

7. The relative yield of disproportionation to combination products from termination of radicals depends on structure; recombination is generally favored for primary and delocalized radicals (Trotman-Dickenson, 1967: Ingold, 1973).

8. Conventional pathways for production of hydrogen include combination of H atoms and abstraction of H from NPH by H•. However, hydride ion (H^-) may also be produced during the very early stages of the ionization process via Dissociative-Attachment processes (Rowntree, 1991).

9. For times up to 10^{-13} sec after ionization, electrons with energies in the 0 to 20 Ev range scattered in an ionization event may produce hydrogen and light hydrocarbons via electron Dissociative-Attachment processes (Rowntree, 1991).

   a. Studies of this phenomenon show that electrons of this energy impacting hydrocarbon films produce H^- and to lesser extent C_1-C_3 anions (Rowntree, 1991).

   b. The mechanism is thought to consist of an initial resonant electron capture by the neutral hydrocarbon molecule followed by direct dissociation of the anion into neutral and anionic fragments. (Rowntree, 1991)

C. Initiation of redox chain oxidations and radical-induced cross-linking by direct radiolysis of organic phases should receive serious consideration.

D. The very low solubility of NPH in aqueous solutions suggests that direct attack by aqueous-derived oxidizing radicals should not be favorable except for ionization events that occur at interfaces between aqueous and organic phases or organic and solid phases.

E. If oxygen is present in tanks, then scavenging of radicals and chemistry analogous to autoxidation reactions are possible.

$$RH + \gamma h\nu \rightarrow R^{\cdot}$$

$$R\bullet + O_2 \rightarrow RO_2\bullet$$

$$RO_2\bullet + RH \rightarrow RO_2H + R\bullet$$

1. Autoxidation pathways produce hydroperoxides initially (Howard, 1970; Ingold, 1968).

2. Introduction of NaOH up to 0.3% (Na) increased the rate of autoxidation in NPH. Above 0.3% NaOH the rate of reaction sharply decreases. An induction period (3-4 h) for the increase in the rate of oxidation is indicative of interfacial phenomena (Shcherbak, 1960).

3.1.2 Acidic Radiolysis

PUREX type mixtures of NPH-TBP were irradiated in the presence of Zr, Nb, U, etc. cations. The expected formation of monobutyl phosphate complexes with the cations was found along with the formation of nitro paraffins (Blake, 1963; Blake, 196; Moore, 1970). The products where C-N bonds are formed were hypothesized to arise from the reaction NP• with NO_2^- and the nitrate esters from the corresponding alcohol and nitric acid.

3.1.3 Hydrolysis

Alkanes do not hydrolyze in the common use of the term.

3.1.4 Oxidation by NO_2^-/NO_3^-

Nitration of organics, in the USTs requires the presence of carbon oxygen moieties, i.e., alcohols, carbonyls, esters, acids, and acid catalyst. The driving force for esterification is the removal of the water generated as the ester bond is formed. In the

UST, the medium is basic, reducing the likelihood of N, C, or P ester formation. The presence of amines, olefins and aromatic compounds, materials which undergo highly exothermic nitration reactions are either absent or in such low quantities in the USTs as not to be of a safety concern.

3.1.5 Conclusion

There are numerous possibilities for the radiolytic initiation of chemical conversion. Oxidation of the NPH fraction via radiological initiation and autoxidation are mechanistically similar in that each pathway involves the formation of the NP•. The problem of mass transfer, surface oxide catalysis and the effect of alkalinity need to be investigated before a conclusive oxidative pathway can be obtained.

3.2 Tributyl Phosphate (TBP)

3.2.1 Radiolysis

A. Not surprisingly, no references were found for the change in composition of TBP in an basic media under γ-irradiation (Burger, 1956; Burger, 1957,). Molecular hydrogen has a G value of 2.5 and butene at 0.08 (Burger, 1958).

B. Irradiation of neat TBP up to 500 rads produced increasing amounts of dibutyl hydrogen phosphate (DBHP) but increased radiation produced no effect (Azzouz, 1989). P-O bond cleavage was the predominant reaction. This is in contrast to reaction involving carboxylic esters where alkoxy radicals were formed. Speculation that the leveling off of P-O bond breakage is inhibited due to lack of water to form the alcohol and ester.

C. A kinetic model of the energy transfer has been developed from published experimental work (Wu, 1980)

D. The action of alpha irradiation in the TPB/DD(dodecane)/HNO_3 were measured with Pu(IV) species (Lloyd, 1985). The primary product is DBMHP, which complexes with Pu(IV).

E. The flammability of the hydrolysis products TBP/NPH/HNO_3 and the components of the hydrolysis have been of interest to researchers in the former Soviet Union (Kulikov, 1981; Kulikov, 1981).

3.2.2 Hydrolysis

A. Alkaline hydrolysis (Burger, 1955) stops at DBP (dibutyl phosphate) with concurrent formation of one equivalent of n-butyl alcohol, either neat or diluted with NPH. Rate of hydrolysis is increased with increasing temperature. No other discussion of alkaline hydrolysis was located in the open literature.

B. Reports of polymer formed during the hydrolysis of TBP may be due to the formation of butyl metaphosphate, which may possibly be equivalent to the meta phosphates formed from alkali metal dihydrogen phosphate.

C. The rate of hydrolysis of TPB to dibutylhydrogen phosphate (DBHP) and monobutyl dihydrogen phosphate (MBDHP) increases with acidity (Schuster, 1990; Vladimirova, 1991; Vladimirova, 1985).

D. Addition of lanthanides and actinides increased the rate of hydrolysis (Becker, 1983; Becker, 1984; Becker, 1984; Stieglitz, 1978; Stieglitz, 1984; Stieglitz, 1985; Lloyd, 1985; Clark, 1978; Vladimirova, 1985). The energetics of the conversion seems to be driven by the removal of the DBHP and/or MBDHP as complexents.

3.2.3 Oxidation-$NO_3^{\cdot}/NO_2^{\cdot}$

A. TBP was found to be stable to 0.2M dichromate in 1M HNO_3, $KMnO_4$, and $Ce(NO_3)4$ (Burger, 1955).

B. Oxidation of TPB with nitric acid (20 M) and at elevated temperatures was found. (Clark, 1978; Vladimirova, 1991).

3.2.4 Conclusion

The stability of TBP to only monoalkaline hydrolysis needs to be reevaluated in light of the limited open literature references. Work needs to be initiated to see if the radiological degradation of TPB is the same or similar in alkaline and acidic media. Again the lack of literature on alkaline conditions will require experimentation before mechanistic or safety issues can be solved.

3.3 Hexone

3.3.1 Radiolysis

No literature references were found that had useful information on the radiolysis of 4-methyl-2-Pentanone (aka:Methyl Isobutyl Ketone or Hexone). One article looked at very low levels of irradiation (107 rad) in the presence of thorium and uranium oxide fuels (Ringel, 1975), but no categorical change in hexone content was determined.

3.3.2 Radiolysis Reduction - other Ketones

A. Studies on the radiolysis of ketones have indicated several pathways for the generation of the radical species. The radical anion (II) of acetone, methyl ethyl ketone and diethyl ketone are formed from the ketone and a

$$RCH_2(R_2)C{=}O + e^{s} \rightarrow RCH_2(R_2)C^{\bullet}\text{-}O^{-} + ROH_{2+} \rightarrow RCH_2(R_2)C^{\bullet}OH$$

solvated electron. Protonation of the radical anion leads to an α hydroxy radical (III) (Akhtar, 1975). Abstraction of a hydrogen atom from another parent ketone leads to alcohol formation.

B. Excited states of irradiated ketones leads to formation of alkyl, hydrogen, and acyl radicals. All three radicals can either abstract hydrogen from a parent ketone, combine to give mixed alkyldimers and alpha diketones with a structure like IX or to a lesser extent disproportionate to an alkene (X) and an alkane (XI). (Akhtar, 1969; Matsui, 1974; Matsui, 1974; Hart, 1964).

$$RCH_2(R_2)C{=}O^{*} \rightarrow R_2\bullet\ (IV) \text{ or } RCH_2\bullet\ (V) + H\bullet + RCH_2CO\bullet(VI) \rightarrow$$

$$H_2 + R_2\text{-}CH_2R\ (VII) + RCH_2OH\ (VIII) + (RCH_2CO)_2\ (IX)\ (V)\ RCH_2CH_2\bullet + (V)$$

$$RCH_2CH_2\bullet \rightarrow RCH{=}CH_2\ (X) + RCH_2CH_3\ (XI)$$

C. Alkenes can also be generated by intermolecular rearrangements involving Norrish type III cleavages to give the aldehyde and the alkene [Akhtar, 1975; Dalton, 1970).

D. The "acetyl" type radical can further disproportionate to carbon monoxide and the $RAH_2\bullet$ radical (Torres, 1984)

E. The presence of base (KOH) in the radiolysis of non-enolizable cyclic ketones in small aliphatic alcohols reduced the formation of cyclic alcohols (Alipour, 1983) and increased the disproportionation of the cyclic radical anions to give alcohols and alkenes.

3.3.3 Radiological Oxidation

A. The hydroxy radical can be trapped by any number of electron accepting species, alkenes, N_2O, O_2, H_2O_2 or other electron rich molecules (Alipour, 1983; Habersbergerova, 1968).

B. Conversion of primary alcohols into aldehydes and diols (McDonell, 1954) was shown to be of extremely low conversion. Introduction of metal oxide surfaces, i.e. Al_2O_3, PbO, etc., was patented in the early 1950's (Battaerd, 1966).

$$R\text{-}CH_2\text{-}CH_2\text{-}OH \rightarrow R\text{-}CH_2\text{-}CH{=}O + (R\text{-}CH_2\text{-}CH\text{-}OH)_2$$

The presence of butanal in the waste tanks may directly come from the butyl alcohol derived from the hydrolysis of TBP. Conversion of alcohol to aldehyde was conducted in the 90 to 200°C and 0.1 Mrad to 10^8 Mrad ranges.

3.3.4 Hydrolysis

A. The carbonyl functionality does not hydrolyze in the classical sense into two or more components as an ester does.

B. The presence of concentrated HO^- (3 M or greater) is not sufficiently strong to cause enolization, the precursor to aldol condensations, by 4 orders of magnitude (House, 1972) p492. The presence of water in the UST precludes this mode of chemical aging as being a primary process.

C. There are two exceptions to this general rule:

a. The aldol product is continuously separated from the reaction mixture, i.e., in the preparation of the aldol condensation product of the acetone with Barium Hydroxide (Conant, 1944). Also Koevenagel reactions of formaldehyde and a

ketone will sometimes remove all of the alpha protons (House, 1972) p. 646-653.

b. Acid catalyzed enolization has been used to a limited extent to initiate aldol condensations (House, 1972), and the only concern to the UST would be the condensation of hexone prior to stabilization for disposal in the UST's.

D. The Cannizzaro reaction of the hydride catalyzed disproportionation of two aldehydes to the corresponding alcohol and carboxylic acid is the well know reaction of aldehydes with no alpha protons (Geissman, 1944). The only conceivable aldehyde with no alpha protons that can be generated in chemistry of the UST's is formaldehyde. No H_2 could be generated via this reaction mechanism. Recent work has indicated that the transient species, $H_2C(O^-)_2$, generated by the from hydrated formaldehyde, may react with water to give formate, H_2, and OH^- (Ashby, 1993).

3.3.5 Oxidation by NO_3^-/NO_2^-

A. No references were found for the oxidation of hexone by NO_3^-/NO_2^-.

B. Oxidations of ketones are usually carried out by peroxides under Bayer-Villiger conditions (House, 1972) p 629-30.

3.3.6 Conclusion

The potential for the radiologically initiated oxidation of hexone to an aldol intermediate exists. But, the lack of work under the conditions found in the UST precludes any definitive pathways or safety analysis.

4.0 Minor UST Components

4.1 Ethers

4.1.1 Radiolysis

Dilute solutions of alkylethers gave both the ether cation or the alkylradical (Symmons, 1984 #867).

$$\text{Me-O-Me} \ \gamma \rightarrow \text{Me-O}^{+}\text{-Me}$$

$$\text{Pr-O-Pr} \ \gamma \rightarrow CH_3CH_2CH_2\text{-OH}^{+} + \text{Pr}^{\bullet}$$

4.1.2 Oxidation by $NO_3^{\cdot}/NO_2^{\cdot}$

No references were found for the $NO_3^{\cdot}/NO_2^{\cdot}$ oxidation of ethers.

4.1.3 Hydrolysis

Hydrolysis of ethers is similar to the reverse reaction of the Williamson ether synthesis (Dermer, 1934).

4.1.4 Conclusion

There is no expectation that ethers will break down under hydrolytic conditions, but formation of radicals and alcohols may occur.

4.2 Esters

4.2.1 Radiolysis

The reaction of ethyl acetate with hydroxyl radical produced carbon dioxide, methanol, and ethanol (Campbell, 1978).

$$\text{ethyl acetate} + \bullet\text{OH} \rightarrow CO_2 + \text{Methanol} + \text{Ethanol}$$

Methyl and ethyl acetate were reduced by the radical abstraction of a proton from the alcoxy group. Formation of molecular hydrogen is a major component (Tabata, 1980; Nenadovic, 1978).

$$CH_3COOMe + \bullet OH \rightarrow CH_3COOCH_2\bullet + H_2O$$

$$CH_3COOCH_3 + \gamma \rightarrow \bullet CH_2COOMe + H\bullet$$

4.2.2 Hydrolysis

Saponification of fats (triglycerides) to carboxylic acids and alcohols is the second oldest known chemical reaction (March, 1968).

4.2.3 Oxidation by $NO_3^{\bullet}/NO_2^{\bullet}$

No data was found for the nitrate/ nitrite oxidation of esters.

4.2.3 Conclusion

The major aging pathway should be via saponification to an alcohol and the metal salt of the acid.

4.3 Alcohols, Including Carbohydrates

4.3.1 Radiolysis

A. The major products from the radiolysis of liquid 1-butanol is H_2 ($G \approx 4.2$), formaldehyde ($g \approx 1.44$), butylaldehyde ($G \approx 0.6$), and ethanol ($G \approx 1.4$) (Teply, 1969)

B. Methanol and ethanol also produce similar ratios of oxidation products (Seki, 1968; Basson, 1967; Feldman, 1984). Results indicated that the formation of formaldehyde was the result of the following reaction, $\bullet CH_2O\text{-}H \rightarrow CH_2O + \bullet H$, rather than disproportionation (Seki, 1968; Shiotani, 1976).

C. Larger molecular weight primary alcohols undergo additional intermolecular rearrangement of the radical (Harna, 1979; Hama, 1986; Basiladze, 1977).

D. The presence of molecular oxygen during the radiolysis can lead to the aldehyde, peroxide and hydrogen radical (Van Linde, 1977)

$$RH_2OH + O_2 + \gamma \rightarrow RCHO + H\bullet + H_2O_2$$

E. Formation of ethers has been shown to be possible under γ-irradiation (Niedzielski, 1980).

$$CH_4 + CH_3OH + \gamma \rightarrow CH_3\text{-}O\text{-}CH_3$$

F. The presence of copper(II) salts during the irradiation of isopropyl alcohols led to the formation of acetone (Freiberg, 1980). Other transition metal oxides could likewise catalyze the formation of ketones during irradiation.

$$CH_3CH(OH)CH_3 + Cu(II) \rightarrow CH_3COCH_3$$

G. Phillips has investigated at the radiological conversion of "sugars" since the late 1950s. The products are generally the oxalic acid, gluconic acid, 2, 3, and 4 carbon fragments as aldehydes and carboxylic acids. Sixteen papers were published between 1958 and 1966 by this author (Phillips, 1958; Phillips, 1958; Phillips, 1966). Zegota has also published the conversion of sucrose into other sugars (Zegota, 1977).

H. γ-Irradiation of sucrose is not as efficient as electron bombardment for the conversion into smaller carbon fragments (Rodriguez-Sanchez, 1975).

4.3.2 Hydrolysis

A. Alcohols do not hydrolyze into smaller molecules in a basic media. Bases stronger than OH^- are required to dehydrate alcohols at temperatures greater than is expected in the UST. Acid dehydration of alcohols occurs via carbonium ion formation, but is also not a candidate reaction in the UST.

B. Base catalyzed conversion of carbohydrates is a well documented process for sugars, cellulose, etc. (Theander, 1988; Greenfield, 1990).

sucrose + OH^- → saccharinic acids + pyruvaldehyde + acetol + glyceraldehyde + diacetyl + lactic acid + 2,4-dihydroxybutanoic acid + 1,2-dihydroxybenzene + 2,3-dihydroxytoluene + 2,4-dihydroxytoluene + 2,6-dihydroxytoluene + 2-hydroxy-3-methyl-2-

cyclopenten-1-one + 2-hydroxy-3,4-dimethyl-2-cyclopenten-1-one + dihydroxyacetone

4.3.3 Oxidation by $NO_3^{\cdot}/NO_2^{\cdot}$

A. No references were found for oxidation of alcohols or carbohydrates with $NO_3^{\cdot}/NO_2^{\cdot}$ under basic conditions and in temperature ranges under 200°C. The reaction of alcohols and carbohydrates with nitric acid is a well-known and explosive field of chemical investigation.

B. Denitration of the waste containing nitric acid was conducted by the addition of sugar and Vanadium oxide (Gerber, 1992). The nitric acid oxidizes the vanadium oxide to V^{+6}, the sugar is oxidized in to 2, 3, 6 carbon aldehydes and ketones. These materials will act as chelators for large cations.

C. The Oppeneaur Oxidation of alcohols to aldehydes and ketones by the use of aluminum alkoxides is a possibility due to the presence of alumina in the UST (Djerassi, 1951)

4.3.4 Conclusion

The sugar used for denitrification was converted into small polyalcohol and organic acids, under acidic conditions. These materials are similar to oxalic acid. Radiologically, alcohols may be converted to aldehydes, ketones, and hydrogen. The carbonyl compounds may be further converted via autoxidation to acids and Bayer-Villiger oxidation products. Again, the data present is insuficient for conclusions to be drawn on safety issues.

4.4 Chlorohydrocarbons

4.4.1 Radiolysis

A. Hydrogen and carbon tetrachloride were radiolyzed and found to form HCl, $CHCl_3$, C_2Cl_6, CH_2Cl_2, C_2Cl_4, and C_2HCl_5. The yields of HCl and chloroform increased with the molar ratio H_2/CCl_4 (Molinari, 1975).

$$CCl_4 \ \gamma \rightarrow HCl + CHCl_3 + C_2Cl_6 + CH_2Cl_2 + C_2Cl_4 + C_2HCl_5$$

B. Compounds containing trichloromethyl groups were exposed to γ-radiation at 77°K in solution or neat. CCl_4, $CHCl_3$, and several chlorinated ethanes were examined in this manner and their products were identified (Mishra, 1975).

$$HCCl_3 \ \gamma \rightarrow HCCl_3^+ + e^-$$

$$HCCl_3^+ \rightarrow (H^+) + \bullet CCl_3$$

$$HCCl_3 + e^- \rightarrow \bullet CHCl_2 + Cl^-$$

C. Neutral aqueous solutions of dichloromethane were irradiated in the presence of H-atom and hydroxyl radical scavengers. Hydrated electron reacts quantitatively with dichloromethane. Hydrolysis of $\bullet CH_2Cl$ radical produced by the initial reaction of the electrons with dichloromethane appears to be relatively insignificant (Balkas, 1972).

$$ClCH_2Cl \ \gamma \rightarrow \bullet CH_2Cl$$

D. Spin trapping experiments during the γ-radiolysis of dichloromethane gave the following radicals: $CH_2Cl\bullet$, $HCO\bullet$, and $Cl\bullet$. Other chlorinated hydrocarbons were also examined (Belevskii, 1981).

$$ClCH_2Cl \ \gamma \rightarrow CH_2Cl\bullet + HCO\bullet + Cl\bullet$$

E. Radiolysis of chloroform and dichloromethane in oxygenated solvent gave Cl_2CHOO and $ClCH_2OO\bullet$. The G-value for the formation of $CH_2Cl_4\bullet$ in the radiolysis of dichloromethane is about 4 (Emmi, 1982; Emmi, 1985).

$$ClCH_2Cl \ \gamma \rightarrow Cl_2CHOO\bullet + ClCH_2OO\bullet$$

F. Dependence of the yield of products in the γ-radiolysis of CCl_4 solutions in secondary alcohols was studied. Rate constants and activation energies were determined (Feldman, 1981).

$$CCl_4 \ \gamma \rightarrow \bullet CCl_3 + \bullet Cl$$

$$2 \bullet CCl_3 \rightarrow C_2Cl_6$$

G. The gamma radiolysis of carbon tetrachloride gave chlorine gas and C_2Cl_6, each with a radiochemical yield (G) of 0.70 mol/100 Ev of absorbed energy. Methanol is a receptor of chlorine atoms (Kovalev, 1984).

4.4.2 Oxidation by $NO_3^{\cdot}/NO_2^{\cdot}$

A. No references were found for the $NO_3^{\cdot}/NO_2^{\cdot}$ oxidation of chlorohydrocarbons.

B. Superoxide ($O_2^{\cdot -}$) reacted with carbon tetrachloride to form a peroxy intermediate which itself is a reactive oxidant. The $Cl_3C\bullet$ radical is formed first followed by the addition of O_2 to produce the corresponding hydroperoxide (Nagano, 1990). This reactive hydroperoxide may rearange to any number of reacive intermediates.

$$CCl_4 + O_2\bullet^- \rightarrow [CCl_4\text{-}O_2\bullet^-] \rightarrow \bullet CCl_3 + O_2 + Cl^-$$

$$\bullet CCl_3 + O_2 \rightarrow CCl_3\text{-}O\text{-}O\bullet$$

5.0 Conclusion

The literature does not contain the information necessary to gain an understanding of the mechanism of chemical aging in the Hanford UST. It does not provide a clear picture of the aging products, the mechanism, or the general safety considerations (flammability, explosiveness) of the contents.

The findings of this search are:

- Reported work has encompassed neither the chemical complexity of the tank waste nor the scope of issues necessary to determine the mechanisms or products from aging.

- The search revealed relevant studies on two major waste components: two studies on the alkaline hydrolysis (Burger 1955) of TBP and one example of radiolysis of NPH under alkaline (Shcherbak 1960) conditions.

- Pathways are enumerated for destruction of the organic compounds, but vagaries due to lack of detailed information about the actual past and present conditions in the tanks (i.e., NO_x and oxygen gas contents) and interfacial effects limit our ability to make conclusions about the degree of degradation and associated safety issues.

- Additional research directed towards elucidating the effects of radiolysis on organic constituents in alkaline conditions as well as the synergistic and inhibitory effects of oxide surface-interactions, metal ion catalysis, mass transfer, solid-liquid interfaces, high concentrations of nitrate and nitrite would be helpful in addressing safety concerns. Most useful would be knowledge of the current state of tank contents obtained by thorough sampling of the problematic tanks.

6.0 List and Abstracts of Cited References

Below in alphabetical order is a list of references for all the literature reviewed in preparing this document. The abstracts for primary references cover either the entire article or that portion that is pertinent to the subject being covered. The abstracts for referenced texts and review articles are necessarily short.

Abell, G. C. and K. Funabashi. 1973. "Simple exponential distribution for initial electron-positive ion separation distances as seen in the gamma-radiolysis of alkanes." J. Chem. Phys. 58:1079.

A numerical solution of the Smoluchowski equation for diffusion, reaction, and neutralization of ion pairs in a dielectric medium in the presence of a scavenger is applied for analysis of recent electron scavenging experiments in alkanes. These experiments can be quantitatively understood in terms of a simple exponential distribution of initial electron-positron ion separation distances. The credibility of this distribution is considerably enhanced when it is employed, in conjunction with Onsager's solution for diffusion of an ion pair in the presence of an external field, to calculate ion yields as a function of field strength. The agreement with experimental results is excellent. The attenuation parameter which characterizes our initial exponential distribution is related to the liquid structure factor and gas phase molecular scattering cross sections through the multiple scattering theory. The wave nature of electron motion is explicitly considered in a discussion of the physical meaning of ionization in liquids.

Akhtar, S. M. S. 1969. "Radiolysis of aliphatic ketones." Diss. Abstr. B 29(12): 69. No abstract available.

Akhtar, S. M. S., R. J. Woods, et al. 1975. "Gamma-radiolysis of liquid acetone, methyl ethyl ketone, and diethyl ketone." Int. J. Radiat. Phys. Chem. 7(5): 603.

The liquid phase radiolyses of acetone, mentyl ethyl ketone and diethyl ketone have been investigated and the detailed analysis of gaseous and liquid products carried out. The effect of absorbed dose on product yield has been examined. The results indicate that the formation of most of the products involve similar reactions for all three ketones, although an intramolecular rearrangement is suggested for ketones having beta-hydrogen atoms. ESR spectra are reported for the polycrystalline ketones at low temperature.

Alipour, E., D. Vidril, et al. 1983. "Effect of irradiation on the oxygen-containing compounds in the presence of olefins." Tetrahedron 39(17): 2807.

The gamma-radiolysis of 2-propanol solutions of cyclohexanone gives mainly hydrogen, acetone, pinacol, methane derived from 2-propanol, and cyclohexanol, 2-(2-cyclohexanonyl)-cyclohexanone, and 3-(2-hydroxy-2-propyl)cyclohexanone derived from cyclohexanone. The radiolytic yields of all these products were highly dependent on the initial cyclohexanone concentration. The formation of cyclic alcohols by radioreduction has been extended to various substituted cyclohexanones. Radiolytically generated solvated electrons are scavenged by cyclohexanone, leading to the corresponding radical anions. The protonation of these radical anions gives rise to cyclohexanol via the dismutation of the hydroxycyclohexyl radicals. Steady-state radiolysis measurements were complemented by pulse radiolysis in dilute solution. It was established that radical-anions and hydroxylated radicals decayed according to a second order rate law. When ketone concentration was lower than 0.1 M, radiolytic yields were in agreement with the mechanism mentioned above. However, in concentrated media the large increase in G(cyclohexanol) cannot be accounted for by only the involvement of radiolytically generated solvated electrons; probably it is due to an electron transfer from the cyclohexanone enolate to cyclohexanone itself, thus generating extra amounts of cyclohexanone radical anions.

Ashby, E. C. and et al. 1993. "Concerning the formation of hydrogen in nuclear waste. Quantitative generation of hydrogen via a cannizzaro intermediate." J. Am. Chem. Soc. 115(3):1171.

Formaldehyde and other aldehydes that do not possess alpha hydrogen atoms are very likely intermediates for the generation of hydrogen during the storage of alkaline nuclear waste. The reaction of water with the Cannizzaro intermediate (H)/sub2/C(O/sup-/)/sub2 yields formic acid, hydrogen gas and an equivalence of hydroxide in quantitative yields.

Avdonina, E. N. 1975. "Dose dependence of gamma-irradiated liquid n-alkanes fragmentation products." Radiat. Eff. 24(4):263.

The data on the dose dependence of light hydrocarbon radiation yields observed in liquid n-pentane gamma-radiolysis can be accounted for to a high degree by assuming that unsaturated compound, accumulating with dose, decrease light hydrocarbon yields in the same way as electron scavengers at very low doses; low molecular weight olefins content decreases due to the attachment of thermal H atoms, the radicals produced giving mainly the respective alkanes and thus increasing

fragment alkane yields; and the reactions of short radicals with unsaturates do not appear significant.

Azzouz, A. and M. Attou. 1989. "Effect of gamma radiation intensity on TBP radiolysis process." Centre de Developpement des Materiaux. Report No. HCR-CDM-06. Fanon, Algeria.

TBP was radiolyzed with a cobalt-60 source of irradiation at various doses and the products were analyzed by IR (Beckman-acculab 9) and UV spectrophotometry (UV-VIS Carl Zeiss-RFA) and by classical chemical methods such as acidimetry and TBP-perchloric acid complex volumetry. The results clearly show the effect of dose value upon the process and especially upon the nature of the products. The initial purity of TBP seems to play a certain role with respect to radiolysis rate. Moreover, it exists a certain correlation between degradation rate and the mixture acidity. This correlation could explain the acid products yield.

Babad, H., D. M. Camaioni, et al. 1993. "Tank Waste Chemistry - A new understanding of waste aging." Waste Management '93, Tucson, Arizona.

Ferricynide/ferrocynide destruction and NPH oxidative pathway presentation.

Babad, H. and D. J. G. 1992. Understanding the cyclic venting phenomena in Hanford liquid waste tanks: The evaluation of Tank 101-SY. Document No.1486-FP Westinghouse Hanford Company, Richland, Washington.

Summary of the work to date on the venting problems for the organic liquid tank 101-SY.

Balkas, T. I. (1972). "Radiolysis of aqueous solutions of methylene chloride"." Int. J. Radiat. Phys. Chem. 4:199.

Neutral aqueous solutions of dichloromethane were radiolyzed in the presence of H-atom and hydroxyl radical scavengers. Hydrated electron reacts quantitatively with dichloromethane. Hydrolysis of $\bullet CH_2Cl$ radical produced by the initial reaction of the electrons with dichloromethane appears to be relatively insignificant.

Barton, D. H. and D. Doller. 1992. "The selective functionalization of saturated hydrocarbons: GIF chemistry." Acc. Chem. Res. 25:504.

This paper is an account of Barton and collaborator's work on developing metal-based catalytic systems for selectively oxidizing saturated hydrocarbons. The pathways are thought to proceed via high valent metal-oxo ions that insert into C-H bonds.

Basiladze, T. M., A. D. Bichiashvili, et al. 1977. "Low-temperature radiolysis of some aliphatic alcohols and thiols"." Soobshch. Akad. Nauk Gruz. Ssr. 85:89.

Low-temperature radiolysis of alcohols showed that alcohols containing (radical) CHROH RO (radical) have a very high reactivity or undergo isomerization into alcohol radicals.

Basson, R. A., and H. J. Van der Linde. 1967. "Radical yield of propionaldehyde in the radiolysis of propanol." Chem. Commun. 2: 91.

Fe^{+3} was found to increase the yield of propionaldyde during the radiolysis of n-propanol. The Fe^{+3} is reduced to Fe^{+2} and the RHCOH radical is oxidized to the aldehyde.

Battaerd, H. A. J. 1970. "Oxidation of aliphatic alcohols to aldehydes." Canadian Pat # 405329.

The invention relates the manufacture of aldehydes from alcohols by the use of radiolytic processes in the presence of solid oxide catalyst from the group consisting of aluminum (III), lead(II), antimony(III), Vanadium (V), etc.

Becker, R., L. Stieglitz, et al. 1983. "Determination of the hydrolysis constants of TBP, HDBP, and H_2/MBP in 3-M nitric acid. Preliminary report No. DP-TR-28,

The rate constants k_1, k_2, and k_3 for the stepwise hydrolysis of TBP to H_3PO_4 were determined in 3-M HNO_3/at 50, 75, and 90°C. The temperature dependance of the hydrolysis constants can be described by the following functions: log k_1 = 13.855 - 5610.4 x 1/T, log k_2 = 13.574 - 5647.4 x 1/T,and log k_3 = 12.03 - 5246.5 x 1/T. A temperature increase of 10/sup 0/C leads to about a tripling of the individual hydrolysis constants. The activation energy of the hydrolysis reaction is 24 to 26 kcal/mol. Comparison of the constant k_1 with the corresponding constant measured in the two-phase system shows that the TBP hydrolysis in the aqueous phase is 30 to 40 times faster than the predominantly organic hydrolysis in the mixed-phase system. Nevertheless, in the two-phase system of the fuel rework process, more than 95% of

the TBP decomposition occurs in the organic phase because of the large difference in TBP concentrations.

Becker, R., L. Stieglitz, et al. 1984a. "Kinetic investigation of TBP-hydrolysis in the PUREX system." PWA-76/84. Kernforschugszent. Karlsruhe.

The experimental conditions were selected to allow a transfer of the results of the requirements of the PUREX process. Single phase experiments in aqueous systems were carried out at 50°C, 75°C and 90°C with nitric acid concentrations between 0,5 M/l and 6 M/l and with pure water. The temperature dependence of the hydrolysis rate constants k_1, k_2 and k_3 and the activation energy could be determined from the Arrhenius-plot. In addition, in the concentration range investigated, linear dependence of the rate constants on the acid concentration was found. The addition of uranium (VI) did not lead to a significant change of the rate constants. Mixed phase experiments were performed at 23.5°C, 50°C and 75°C with nitric acid concentrations of 0,5 M/l and 3 M/l. For the rate constant k_1 the temperature dependence and the activation energy were evaluated. The reaction seems to be a first-order reaction with respect to the acid concentration. After the addition of uranium (VI) [approx=35 g/l (org.)] or zirconium [1g/l (aq.)] the hydrolytic reaction rate was significantly increased. Under these conditions an influence of the nitric acid concentration could not be noticed. The formation of monobutyl phosphoric acid in mixed phase systems was also investigated. During the usual experimental time of approximately 14 days measurable H_2MBP concentrations were found in the aqueous phase only if there was a sufficient aqueous HDPB concentration.

Becker, R., L. Stieglitz, et al. 1984b. "Kinetics of the hydrolysis of tributyl phosphate in the PUREX system." Kernforschungszent. Karlsruhe. [Ber.] KfK. KfK-3836.

This investigation deals with the hydrolytic degradation of Bu_3PO_4 under acid conditions. Rate increases with concentration of nitric acid and is increased the presence of some lanthanide and actinide cations. The increase rate for specific cations is due to the removal of the DBP and MBP as a complex with the specific cation.

Belevskii, V. N., and U. Westfahl. 1981. "Identification of radicals in the radiolysis of perchloro- containing compounds"." Vestn. Mosk. Univ., Ser. 2 Khim. 22: 563.

Spin trapping experiments during the t-radiolysis of dichloromethane gave the following radicals: CH_2Cl, HCO, and Cl. Other chlorinated hydrocarbons were also examined.

Benson, S. W., and H. E. O'Neil. 1970. Kinetic Data on Gas Phase Unimolecular Reactions. National Bureau of Standards, Report Number NSRDS-NBS-21, Gaithersburg, Maryland.

Rate data provided in this book for beta-scission of alkyl radicals enable activation barriers to be estimated.

Blake, C. A., Jr., W. Davis Jr., et al. 1963a. "Properties and cleanup of degraded tributyl phosphate-Amsco solutions and the relative stabilities of alternative extractant-diluent systems." ORNL-3524, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Same abstract as below.

Blake, C. A., Jr., W. Davis Jr., et al. 1963b. "Properties of degraded tbp-amsco solutions and alternative extractant-diluent system." Nuc. Sci. Eng. 17:626.

The formation and chemical properties of the major degradation products of several diluents in contact with acidic nitrate solutions were studied, together with (1) methods for purifying degraded solvent, (2) methods of pretreating the solvent to minimize degradation, and (3) the relative stability of several solvent systems. Emphasis was on Amsco 125-52, a specially treated naphtha used extensively at ORNL in radiochemical processing. Degradation of the diluent, usually containing 1 M TBP, was induced by boiling with HNO_3 or by irradiating (^{60}Co gamma) while agitating the solvent phase with HNO_3. Nitroparaffins, RNO_2, were the major degradation products and were formed at twice the rate when TBP was present during degradation. Under the conditions of degradation, 100 watt-h/liter exposure was essentially equivalent to 11 h boiling with 2 M HNO_3 as shown by Zr-Nb extraction, total organic nitrogen analysis, and spectrophotometric analysis. Extraction from acid solution of certain fission products, e.g., Zr-Nb but not Ru, increased markedly after the degraded solvent was treated with alkaline reagents, and a test employing solid calcium hydroxide was developed to estimate the extent of diluent degradation. The alkaline treatment converted a significant fraction of the nitroparaffin to the enol form. There was synergistic extraction of Zr-Nb by the combination of TBP and degraded diluent. Cleanup of the degraded solvents can be attained by scrubbing with alkanolamines to remove nitroparaffin-amine salts or by contacting with activated alumina. Both methods have doubtful economic benefits, and the latter requires solids handling. Low pressure flash distillation is an effective, inexpensive method for partial or total repurification of degraded TBP-Amsco. With aliphatic diluents the configuration most stable against nitration is the simplest, e.g..

n-dodecane. Pretreating Amsco 125-82 by scrubbing with concentrated sulfuric acid before degrading with hot nitric acid improved its stability. Aromatic diluents increase the radiation stability and extraction selectivity of TBP. The stabilities of the simple alkylbenzenes against nitration vary widely with the number of alkyl groups, their type, and their position, but a mixture of diethylbenzenes low in the para isomer and some of the polymethylbenzenes did not form fission product extractants in routine tests. Nitration of the benzene ring does not produce Zr-Nb extractants.

Broszkiewicz, R. K., E. Kozlowska-Milner, et al. 1981. J. Phys. Chem. 85:2258.

This paper reports on pathways for generation of NO_3 radical from radiolysis of nitrate solutions.

Bugaenko, L. T., M. G. Kuzmin, et al. 1993. High Energy Chemistry. Prentice Hall, New York.

This book covers the fields of plasma chemistry, radiation chemistry, and photochemistry. The chapter on radiation chemistry provides a fundamental view. Its discussions of radiation processes of inorganic and organic compounds are brief but informative and well referenced. Especially good is a summary of the Russian literature. Tables show typical product distributions obtained from hydrocarbons and functionalized hydrocarbons. The perturbing effects of oxygen and mixed phases containing both organic and inorganic compounds lay beyond the scope of this chapter.

Burger, L. L. 1955. The Chemistry of Tributyl Phosphate A Review. Document No.40910, Hanford Works, Richland, Washington.

A general review of the chemistry of TPB was prepared, with the general emphasis on acid hydrolysis. Also, alkaline hydrolysis was found to stop unexpectedly at DBP.

Burger, L. L. 1956. The Flammability of Vapors Above the PUREX Process. General Electric-Hanford Company, Document Number 40820, Richland, Washington.

The flammability of the operating conditions for the PUREX solvent system was determined. For any of the NPH choices, as long as the temperature is kept below 45°C the system is safe.

Burger, L. L. 1957. The Esters of the Acids of Phosphorus As Solvents. Document No.44888, Hanford Works, Richland, Washington. Assessment of the TPB as a solvent for extracting uranium and plutonium was examined.

Burger, L. L., and E. D. McClanahan. 1958. "Tributyl Phosphate and Its Diluent Systems." Ind. Eng. Chem. 50(2):153.

TBP-diluent mixtures similar to the compositions used at Hanford were gamma irradiated; the presence of DBHP and some evidence of MBDHP were detected.

Buxton, G. V., C. L. Greenstock, et al. 1988. "Critical review of rate constants for reactions of hydrated electrons, hydrogen atoms, and hydroxyl radicals (HO•/O⁻•) in aqueous solution." J. Chem. Ref. Data 17(2):513.

A compilation of rate constants for reactions of radiolysis intermediates: HO•, H•, e- (aq), O⁻·

Campbell, I. M., and P. E. Parkinson. 1978. "Rate constants for reactions of hydroxyl radicals with ester vapors at 292°K"." Chem. Phys. Lett. 53:385.

Ethyl acetate and propionate were examined for CO_2 yields in gas phase hydroxyl radical reactions. Rate constants were determined.

Carter, W. P. L., K. R. Darnall, et al. 1979. "Reactions of C_2 and C_4 alpha-hydroxyl radicals with oxygen." J. Phys. Chem. 83:2305.

A gas phase study of photoxidation ethanol and butanol in NO_x-Air mixtures. Reaction of HO• with substrate produces alpha-hydroxyalkyl radicals that react with oxygen by H-transfer to produce carbonyl compounds.

Carter, W. P. L., K. R. Darnall, et al. 1976. Chem. Phys. Lett. 42:22.

This paper reports on the gas phase photo-oxidation of long chain alkanes in the presence of air and nitrogen oxides. A mechanism for production of alpha-hydroxy radicals from 1,5-shifts of alkoxy radicals produced by reaction of NO with alkyl peroxy radicals.

Clark, W. E., W. B. Howerton, et al. 1978. "The oxidation and degradation of short-chain aliphatic compounds by hyperazeotropic nitric acid." Nucl. Sci. Eng. 66(1):110.

Experiments were conducted to determine the ultimate fate of organic material present in nuclear fuel.

Conant, J. B. 1944. "Synthesis of 3-hydroxy-4-methyl-2-pentanone." Org. Syn.: Coll. V.1:199.

The aldol product was removed by separation Soxhlet Extraction. One of the very few examples of hydroxide anion aldol condensations of non-activated ketones.

Daniels, M. 1969. "NO_3 radicals by radiolysis." J. Phys. Chem. 73:3710.

This paper reports on the generation of NO_3• by radiolysis of nitrate solutions.

Dermer, O. C. 1934. "Williamson ether synthesis." Chem. Rev. 14:409.

The reaction of alkyl halides with sodium alkoxides for the formation of ethers is reviewed.

Djerassi, C. 1951. "The Oppenauer Reaction." Organic Reactions. J. Wiley and Sons, New York, 207.

The Oppenauer reaction is the reverse reaction to the Meerwein-Pondorff-Verlay reduction of aldehydes by the use of aluminum alkoxides. The majority of the work up to 1951 was to provide a selective oxidation of primary steroidal alcohols to aldehydes. The method has been found to be applicable to saturated alcohols [Davies and Hodgson, J. Soc. Chem. Ind., 62:109(1943)].

Emmi, S. S., G. Beggatio, et al. 1985. "Hydroxyl radical reaction with methylene chloride: direct spectrophotometric observation of $CHCl_3$ radical formation." J. Radioanal. Nucl. Chem. 93:189.

Hydrogen abstraction from dichloromethane by hydroxyl radical in aqueous solution was monitored by the direct observation of $CHCl_2$ radical.

Emmi, S. S., G. Beggiato, et al. 1982. "Haloalkyl radicals: formation and reactivity of mono- and dichloromethylperoxy radicals." Proc. Tihany Symp. Radiat. Chem. 5:677.

Radiolysis the chloroform and dichloromethane in oxygenated solvent gave Cl_2CHOO and $ClCH_2OO$. The G-value for the formation of CH_2Cl in the radiolysis of dichloromethane is about 4.

Feldman, L., and Z. B. Alfassi. 1981. "Kinetics of radiation-induced hydrogen abstraction of trichloromethyl radicals in the liquid phase. Secondary alcohols." J. Phys. Chem. 85:3060.

Dependence of the yield of products in the t-radiolysis of CCl_4 solutions in secondary alcohols was studied. Rate constants and activation energies were determined.

Feldman, V. I., V. N. Belevslii, et al. (1984). "Spin trap method. Mechanism of formation of hydroxylalkyl radicals during the radiolysis of alcohols." Vestn. Mosk. Univ. Ser. 2 Khim. 25:258.

t-Radiolysis was performed with methanol, ethanol, and isopropanol. The ratio of the quantum yield of hydroxylalkyl to alkoxy radicals was determined. Alkoxy radicals formed more readily in liquid phase.

Freiberg, M., W. A. Mulac, et al. 1980. "Reactions of aliphatic free radicals with copper cations in aqueous solutions. Part 3. Reactions with cuprous ions: a pulse radiolysis study." J. Chem. Soc., Faraday Trans. I 76:1838.

Deaerated solutions containing methanol, ethanol, and isopropanol in the presence of Cu(II) ions were studied under pulse radiolysis conditions. Upon removal of the hydrogen radical, all three carbon-center radicals reacted with Cu^{+2}(aq.) to give Cu+, H_3O^+ and a ketone except for methanol which gave a Cu-CH_2OH complex for which the formation complex is ca. $5x10^3$ dm^3/mol.

Frey, H. M., and R. Walsh. 1969. Chem. Rev. 103:103.

A review of alkyl radical decomposition reactions.

Geissman. 1944. "Canizzarro Reaction." Org. Reactions 2:94.

Review of the Cannizzaro reaction.

Gerber, M. A., L. L. Burger, et al. 1992. Assessment of Concentration Mechanisms for Organic Wastes in Underground Storage Tanks at Hanford. PNL-8339. Pacific Northwest Laboratory, Richland, Washington.

Evaluation of the constituents that may be in the UST at the Hanford site.

Greenfield, B. F., A. Rosever, et al. 1990. Review of the microbiological, chemical and radiolytic degradation of organic material likely to be present in intermediate level and low-level radioactive wastes. AEA Technology, Harwell Laboratory.

British Dept. of Ecology's reports on aging paths for solid organic materials contaminated with radioactive waste.

Gremlich, H. U., and R. E. Buehler. 1983. "Fate of the radiation-initiated charged species in liquid alkane-carbon tetrachloride systems." J. Phys. Chem. 87:3267.

Radiolysis of carbon tetrachloride, through a dissociative electron attachment, forms Cl_3C and Cl- with an alkane radical cation. Apparently, the radical and anion recombine to the an ion pair. The ion pair undergoes a charge transfer to give the alkane and Cl_3C^+.

Habersbergerova, A., P. Kourim, et al. 1968. "Radiolysis of liquid methanol in the presence of 1-octene: Mechanism of glycol and formaldehyde formation." Coll. Czech. Chem. Comm. 34: 3657.

The formation of glycol and formaldehyde in the radiolysis of liquid methanol has been investigated in the presence of 1-octene reacting efficiently with free radicals, but not so much with free electrons. Glycol formation is almost totally suppressed in the presence of 1-octene; this proves that glycol is formed from the combination of CH2OH radicals. The main products of the irradiation of the mixture lead to 1-nonyl alcohol and 2-methyl-1-octyl alcohol.

Hama, Y. 1986. "On intramolecular migration of alkyl radicals produced in rigid n-alkane diols by gamma-irradiation." Radiat. Phys. Chem. 27(5):411.

Thermally and photochemically induced radical conversion in the polycrystalline diols gamma-irradiated at 77 K is attributed to internal migrations. The above mutual radical conversion was observed at a temperature <173 K also in the glassy mixture. In this mixture, however, all the radicals disappeared on heat treatment at 200 K. The activation energy for the decay process of alkyl radical in the glassy mixture was 4.7 vs. 7 kcal/mol for the polycrystalline samples. Also, the results indicated that a photochemical reaction different from that in the polycrystalline sample occurred in the glassy one.

Hama, Y., K. Hamanaka, et al. 1979. "ESR studies of n-alkane diols irradiated by gamma-rays; identification of radicals and mechanism of radical migration." Radiat. Phys. Chem. 13(1-2):13.

Gamma-irradiation of alpha and omega-alkanediols produced predominantly terminal -HOHC• and internal -CH_2HC•CH_2- free radicals. The complex radical conformation of the terminal radicals was detected by computer simulation. Terminal radicals were converted to internal radicals by UV irradiation. Internal radicals were converted to terminal radicals by heating; this process is intramolecular and of 7 kcal/mol apparent activation energy. The kinetic expressions for the terminal-internal radical interconversion are discussed in terms of a potential model for the unpaired electron of the molecular chain.

Hart, E. J., S. Gordon, et al. 1964. "Rate constants of hydrated electron reactions with organic compounds." Physical Chemistry 68(6):1271.

Second-order rate constants of reactions of some aliphatic, aromatic, and heterocyclic compounds with hydrated electrons are reported. These constants were measured in unbuffered and in alkaline solutions by following the decay of the hydrated electron absorption band produced by a single 0.2 or 0.4 microsec pulse of 15 Mev electrons. The rate constant for acetone is 5.9×10^{1} M^{1} sec^{-1}.

Hawkins, E. G. E. 1973. "Reactions of hydroperoxides." Free Radical Reactions. London, Butterworths. Series One, ed. 95.

A review of the reactions of hydroperoxides with emphasis on the literature between 1971-73.

House, H. O. 1972. Modern Synthetic Reactions. The Benjamin/Cummins Publishing Company, Menlo Park, California.

Howard, J. A. 1970. "Homogeneous liquid-phase autooxidations." Free Radicals. Wiley-Interscience, New York.

A book chapter describing organic autooxidation chemistry with emphasis on understanding the theoretical aspects of autoxidation.

Ingold, K. U. 1968. "Peroxy radicals." Acc. Chem. Res. 2:1.

An account of research on the reactions of peroxy radicals, a key intermediate in autooxidation reactions.

Ingold, K. U. 1973. "Rate Constants for Free Radical Reactions in Solution." Free Radicals. Wiley-Interscience, New York.

A book chapter on the kinetics of free radical reactions in solution. Includes a section on the factors that influence disproportionation/combination product ratios for radical-radical termination reactions.

Janata, E., and R. H. Schuler. 1982. "Rate constant for scavenging e_{aq}- in N_2O saturated solutions." J. Phys. Chem. 86:2078.

A bimolecular solution-phase rate constant of 9.1×10^{9} is measured for reaction of N_2O with HO• (pH 6-6.5).

Kerr, J. A. 1970. "Rate Processes in the Gas Phase." Free Radicals Volume 1. Wiley-Interscience, New York : 1-36.

Gas-phase reactions of organic and inorganic radicals are described. Thermochemical kinetic data for reactions are compiled Theories explaining structure-reactivity relationships.

Knight, R. J., and H. C. Sutton. 1967. "Radiolysis of aqueous solutions of nitric oxide." Trans. Faraday Soc. 63:2628.

Bimolecular rate constant for reaction of H• with NO at pH 7 is $1.1e^{+10}$ L/mol•s) determined by competition kinetics. The reference reaction was the reaction of H• with nitrite ion. The molecular rate constant for reaction of solvated electron with NO at pH 7 is 1.1×10^{10}.

Kochi, J. K. 1973. Oxidation-Reduction Reactions of Free Radicals and Metal Complexes. Free Radicals. New York, Wiley-Interscience. 591-685.

Book chapter reviews mechanisms and rate data for reactions of metal ions with radicals. The roles of transition metal ions as catalysts in oxidation and autooxidation reactions are discussed.

Kochi, J. K. 1973. Oxygen Radicals. Free Radicals Volume 2. New York, Wiley-Interscience. 665-703.

Book chapter reviews the mechanisms and rate data for reaction of oxygen-centered radicals with organic compounds.

Kovalev, G. V., A. L. Karasev, et al. 1984. "Yields of radicals in the radiolysis of liquid carbon tetrachloride in the presence of methanol." Khim. Vys. Energ. 18:306.

The gamma radiolysis of carbon tetrachloride gave chlorine gas and C_2Cl_6, each with a radiochemical yield (G) of 0.70 mol/100 eV of absorbed energy. Methanol is a receptor of chlorine atoms.

Kulikov, I. A., N. V. Kermanova, et al. 1981. "Effect of alpha- and gamma-radiations on tributyl phosphate decomposition and Pu^{4+} and Zr^{4+} distribution coefficients." Radiokhimiya 23(6):825.

Radiation chemical decomposition of two-phase system of 30% TBP in paraffines-HNO_3 (3 mol/l) during cap alpha- and gamma-radiolysis under static conditions and during mixing in vacuum, in the atmosphere of oxygen and inert gases, is investigated. The yields of extractant radiation decomposition products and their distribution among aqueous, organic and gaseous phases are determined. The effect of radiation type on Pu^{4+} and Zr^{4+} distribution coefficients between organic and aqueous phases is studied. It is shown that with the increase of radiation dose the increase of distribution coefficients is similar for alpha and gamma-radiolysis. Pu^{4+} distribution coefficients increase from 1.9 to 75, and Zr^{4+} coefficients increase - from 0.04 to 1.7 with the dose increase from 0 to 1.6×10^5/Gy. Separation coefficients of Pu^{4+} and Zr^{4+} in the dose range studied practically do not change.

Kulikov, I. A., A. S. Milovanova, et al. 1981. "Influence of alpha-particle and gamma-rays on distribution coefficients of americium(3+) and cerium(3+) in the extraction system based on bis(2-ethylhexyl) phosphoric acid." Radiokhimiya 32(3):101.

The alpha-particle and gamma-ray induced radiolysis of a 20% solution.

Lias, S. G., J. E. Bartmess, et al. 1988. "Gas Phase Ion and Neutral Thermochemistry." J. Phys. Chem. Ref. Data 17, Sup. 1.

This reference is a comprehensive compilation of critically-evaluated heats of formation of organic and inorganic compounds and ions.

Lilga, M. A., M. R. Lumetta, et al. 1992. Ferrocynide Safety Project Subtask 3.4 Aging Studies FY 1992 Annual Report, PNL-8387, Pacific Northwest Laboratory, Richland, Washington.

At high caustic levels (pH 12-14) $Na_2NiFe(CN)_6$ is 95% dissolved in 0.5 h to give $Na_4Fe(CN)_6$ plus $Ni(OH)_4$, which is insoluble. Radiation did not affect the solubility. Hydrolysis of the CN^- will be conducted later.

Lloyd, M. H., and R. L. Fellows. 1985. "Alpha radiolysis and other factors affecting hydrolysis of tributyl phosphate." ORNL/TM-9565, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

The primary purpose of this study was to identify the principal degradations to TPB (30%) in dodecane solutions. Results indicated that DBP was the primary complexant for Pu below 50°C, MBP was the primary complexant above that temperature, and hydrolysis was dependent upon temperature.

March, J. 1968. Advanced Organic Chemistry: Reactions, Mechanisms, and Structure. McGraw-Hill Book Company, New York.

Top organic chemistry reference for chemical mechanisms and reactions.

Matsui, M., and M. Imamura. 1974. "Radiation chemical studies with cyclotron beams. III. Heavy-ion radiolysis of liquid aliphatic ketones." Bull. Chem. Soc. Jap. 47(5):1113.

McDonell, W. R., and A. S. Newton. 1954. "The radiation chemistr of aliphatic alcohols." J. Amer. Chem. Soc. 76:4651.

The products formed in the irradiation of the liquid, air-free alcohols -- methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, s-butyl, t-butyl, n-octyl, and n-decyl -- with 28 Mev helium ions have been determined. The reduced products were hydrogen and saturated hydrocarbons with a total G (reduced) of 7 to 8 equivalents per 100 eV in all cases. The oxidized products were aldehydes and glycols from primary alcohols; aldehydes, ketones and glycols from secondary alcohols; and ketones from tertiary alcohols. Carbon monoxide and water were formed in all cases. The products are consistent with the principal bond rupture at the carbinol carbon atom; the reactivity of such carbinol bonded groups exhibited the order $H > C_2H_5 > CH_3$ in the alcohols studied. The mechanism of formation of some of the products has been discussed.

Meisel, D., H. Diamond, et al. 1991a. Radiation chemistry of synthetic waste. ANL-9141, Argonne National Laboratory, Chicago, Illinois.

The yield of H_2 from radiolysis of aqueous solutions is substantially reduced by the presence of nitrate and nitrite in waste solutions. Nitrate is more efficient in scavenging the precursors to H_2 than nitrite. Nitrate is the major scavenger for e-aq and nitrate is the major scavenger for H atoms.

Meisel, D., H. Diamond, et al. 1991b. Radiolytic generation of gases from synthetic waste. ANL-9142, Argonne National Laboratory, Chicago, Illinois.

Yields of H_2, N_2O, O_2 and N_2 in simulated waste solutions, containing high nitrate, nitrite, hydroxide and aluminate, were experimentally measured in the presence and absence of moderate concentrations of organic chelators and some of their degradation products. These yields were measured from 30 to 60°. No effect of H_2 yield on dose rate was observed.

Milosavljevic, B. H., and O. I. Micic. 1978. "Solvated electron reactions in water-alcohol solutions." J. Phys. Chem. 82:1359.

Rate constants were measured for reactions of H atom with oxygen in water-alcohol solutions. The rate constant with O_2 is diffusion limited. Decay kinetics of the solvated electron were monitored at 600 nm.

Mishra, S. P. and M. C. R. Symmons. 1975. "Radiation processes for trichloromethyl compounds. Trichloromethyl radical." Int. J. Radiat. Phys. Chem. 7: 617.

Compounds containing trichloromethyl groups were exposed to t- radiation at 77K in solution or neat. CCl_4, $CHCl_3$, and several chlorinated ethanes were examined in this manner and their products were identified.

Molinari, M. A., N. R. Strehar, et al. 1975. "The reaction between carbon tetrachloride and hydrogen induced by gamma radiation." Argent. Com. Nac. Energ. At. 398: 11.

Hydrogen and carbon tetrachloride were radiolyzed and found to form HCl, $CHCl_3$, C_2Cl_6, CH_2Cl_2, C_2Cl_4, and C_2HCl_5. The yields of HCl and chloroform increased with the molar ratio H_2/CCl_4.

Moore, J. G., and D. J. Crouse. 1970. "Solvent stability in nuclear fuel processing: cyclic irradiation studies of 15 vol percent TBP-n-dodecane." ORNL-4618, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Cyclic irradiation tests were conducted on a solution of 15%(Vol)TPB in n-Dodecane with 0.38 $M(U)_2(NO_3)_2$ and 3M HNO_3. Hydrolyzed TPB was removed by bicarbonate washes. Increasing levels of irradiation increased the ability of the solvent to extract zirconium and uranium cations.

Nagano, T., H. Yamamoto, et al. 1990. "Cooxidation reactions during oxidation of superoxide with polyhalides, carbon dioxide, phosphates, and acyl halides." J. Am. Chem. Soc. 112:3529.

Superoxide (O_2^-) reacted with carbon tetrachloride to form a peroxy intermediate which itself is a reactive oxidant. Apparently, CCl_3 is formed first followed by the addition of O_2 to form the corresponding hydroperoxide.

Nenadovic, M. T., and O. I. Micic. 1978. "Free radical reactions of aliphatic esters." Glas. Hem. Deus. Beograd 43:281.

Methyl and ethyl acetate were reduced using hydrated electrons. Pulse radiolysis indicated that the polaron added to the carbonyl group to give a ketyl. In acid the ketyl was protonated. Hydroxyl radical attack on the esters indicated that the radical abstracted a hydrogen from the alkoxy group.

Neta, P., and R. E. Huie. 1986. "Rate Constants for Reactions of NO3 Radicals in Aqueous Solutions." J. Phys. Chem. 90:4644.

This paper reports bimolecular rate constants for $NO_3\bullet$ radical with organic and inorganic compounds and metal ions. $NO_3\bullet$ was generated by pulse radiolysis of dilute nitric acid and 5 M sodium nitrate.

Niedzielski, J., and J. Gawlowski. 1980. "The formation of ethers in the gas phase gamma-radiolysis of simple alkanes in the presence of alcohols." Radiat. Eff. 52:35.

The formation of ethers in the radiolysis of methane, ethane, and propane in the presence of either methanol or ethanol was studied in the gas phase. The mechanism is apparently ionic and involves the condensation of ions originating from alkanes with alcohols, solvation of ionic complexes by alcohols, and finally supersession of ethers by alcohols in equilibrium exchanges.

Pancini, E., V. Santoro, et al. 1970. "Gamma-Radiolysis of alkanes I. Liquid n-pentane and n-hexane." Int. J. Radiat. Phys. Chem. 2(3):147.

The gamma-radiolyses of liquid n-pentane and n-hexane were investigated by means of gas-chromatographic analysis of all the products. The probability of fragmentation, dehydrogenation and substitution at any given position of the molecule is given. The results obtained with the 2 hydrocarbons are compared and analogies in their behavior are revealed by taking account of the particular condition of the central position of pentane and assigning to it a statistical weight of half that of the other C atoms. Some special aspects of the experimental results, namely the identification of pentane dimers and the overall H balance, are discussed in detail.

Phillips, G. O., and G. J. Moody. 1958. "Radiation chemistry of carbohydrates. Part II. Irradiation of aqueous solutions of dextran with gamma radiation." J. Chem. Soc. B 3534(9).

The degradation of dextran in aqueous solution by ^{60}Co gamma radiation is examined by measuring the amount of acid formed and the changes in optical rotation and reducing power. Chromatographic and isotope dilution methods reveal that the main products are glucose, isomaltose, isomaltotriose, gluconic acid, glucuronic acid, glyoxal, erythrose, and glyceraldehyde. These products are considered to arise from at least two independent degradation processes: (a) hydrolysis to glucose, isomaltose, and isomaltotriose, while secondary reactions involving glucose give erythrose, glyoxal and glyceraldehyde; (b) the formation of gluconic acid and glucuronic acid. The irradiated solution shows an absorption at 265 M μ due to dihydroxyacetone, which is also formed during the irradiation of glucose solutions.

Phillips, G. O., G. J. Moody, et al. 1958. "Radiation chemistry of carbohydrates. Part I. Action of ionizing radiation on aqueous solutions of d-glucose." J. Chem. Soc. B 34(1958):3522.

When irradiated in dilute aqueous solution, D-glucose is degraded to D-glucuronic acid, D-gluconic acid, glyoxal, D-arabinose, D-erythrose, formaldehyde, saccharic acid, and 1 : 3-dihydroxyacetone, which have been estimated quantitatively. Hydrogen peroxide is also formed. Under fully oxygenated and evacuated conditions, variation in glucose concentration and dose rate does not influence the yield of acid; the extent and pattern of degradation are the same for 1 Mev electrons and cobalt gamma radiation. The liberation of gas indicates a post-irradiation process, also shown by changes in ultraviolet absorption spectra and hydrogen

peroxide concentration. The absorption spectrum of the irradiated solutions shows a characteristic absorption maximum at 265 M μ sensitive to alkali; it is related to 1: 3-dihydroxyacetone. By following the yield-dose curves for the main products, the primary and secondary products are distinguished. The main types of degradation are thus identified.

Ringel, H. 1975. "Continuous production of thorium oxide and (thorium, uranium(IV)) oxide fuel kernels for high-temperature reactors." Ber. Kernforschungsanlage Juelich 1258(143).

The development of the KFA protonation process for producing ThO_2 and $(The,U)O_2$-fuel kernels for high-temperature reactors is described. Special considerations are given to the refabrication of kernels from highly radioactive fuel solutions of reprocessing plants. The process is based on gelation of ThO_2 and $(The,U)O_2$-sol droplets by NH_3 absorption in an orginal phase of Me iso-Bu ketone. The dried and fired fuel kernels are produced in a remote and continuous manner. Special studies were made on the following process steps: (1) generating drops of uniform size at high frequency within a liquid, (2) continuous washing of gel particles, and (3) continuous drying of gel particles. In an irradiation experiment, the ThO_2 and $(The,U)O_2$ sols and Me iso-Bu ketone were exposed stepwise to a total gamma-dose of 107 R. No damage which would impair the production process was detected.

Rodriguez-Sanchez, M. A. and J. Reyes Lujon. 1975. "Radiation effects on sucrose in aqueous solutions and in polycrystalline solid state." Rev. Soc. Quim. Mex. 19:48.

gamma-Radiolysis of aqueous solutions of sucrose produced glucose, gluconic acid, fructose, and glucosone. Electron radiolysis decomposes sucrose more efficiently than gamma-radiolysis.

Ross, A. B., W. G. Mallard, et al. 1992. NRDL/NIST Solution Kinetics Database, National Institute of Standards and Technology, Gaithersburg, Maryland. This reference is a computerized data base of rate constants for solution-phase reactions of radicals. It includes a comprehensive list of the reactions of radiolysis-generated radicals, HO, e-(aq), H, O^-, O_2^-, NO_3, SO_4^-, etc. Rate measurements are referenced, abstracted and evaluated for accuracy.

Rowntree, P., L. Parenteau, et al. 1991. "Anion yields produced by low-energy impact on condensed hydrocarbon films." J. Phys. Chem. 95:4902.

Low energy (0-20 eV) electron impact on thin condensed hydrocarbon films is observed to produce, via dissociative attachment and dipolar dissociation, significant yields of hydride ion and much lower yields of alkyl fragment (1-3 carbons) anions. Yields are measured as function of incident electron energy and film thickness for saturated hydrocarbons with 2-9 carbons and unsaturated hydrocarbons with 2-4 carbons. Each type of anion desorbing from the saturated hydrocarbon film exhibits a single dissociative attachment resonance at approximately 10 eV, which is found to vary only slightly with the hydrocarbon chain length, in contrast with trends observed in the gas phase for hydride formation from these systems. The thickness dependence of the anion yields suggests that the observed hydride ions are formed in the uppermost 3-5 layers of the film, while the larger desorbing anion fragments arise from the first 1 or 2 layers.

Rush, J. D., and B. H. J. Bielski. 1985. "Pulse radiolytic studies of the reactions of HO_2/O_2^- with ferric ions and its implications on the occurrence of the Haber-Weiss reaction." J. Phys. Chem. 89:5062.

This reference reports measurements of rate constants for redox reactions of HOO• and O_2. with Fe(III).

Schuster, J., R. Eichler, et al. 1990. "Hydrolysis of TBP in mixed phase experiments." Zentralinst. Kernforsch., Rossendorf Dresden. [Ber.] ZfK. ZfK:45.

The hydrolysis rate of TBP increases with temperature and HNO_3 concentration. The hydrolysis rate decreases in the absence of light. Rare earth nitrates have a synergistic effect on the hydrolysis in NO_3.

Seddon, W. A. and M. J. Young. 1970. "Pulse radiolysis of nitric oxide in aqueous solutions." Can. J. Chem. 48:393.

The rate constant for NO + solvated electron is measured directly in water at room temperature and pH 7 by decay kinetics. A value of $2.3e^{+10}$ L/(mol•s) is reported.

Seguchi, T. 1991. "Irradiation effects of paraffins as polymer model compounds." ACS Symp. Ser. 475 (Radiat. Eff. Polym):442.

Irradiation effects of n-paraffins and squalane as model compounds for polyethylene and ethylene-propylene copolymer, respectively, were investigated by product analysis $C_{20}H_{42}$, $C_{21}H_{44}$, $C_{23}H_{48}$, and $C_{24}H_{50}$, and squalane were irradiated by gamma rays under vacuum in crystalline, glassy, and liquid states. The evolved gases were

analyzed by gas chromatography and the molecular weight changes were analyzed by liquid chromatography and mass spectroscopy. The G-values of crosslinking of the n-paraffins were 1.2 in the crystalline and 1.7 in the liquid state, and showed no difference between odd and even carbon numbers. The G-value of squalane was 1.7 in liquid state and 1.3 in glassy state at -77 degree. Double bonds were plentiful in the crosslinked products, especially in the liquid state. The probability of chain scission was established as negligible, except at chain ends.

Seguchi, T., N. Hayakawa, et al. 1985. "Radiation-crosslinking of polyethylene model compounds." Radiat. Phys. Chem. 25(1-3):399.

Crosslinking of a series of n-paraffins with both odd and even C numbers was studied. From measurements of gel permeation chromatography and by mass spectroscopy, dimer, trimer and higher oligomers were detected. G-values of crosslinking, H and CH_4 evolution, and double bond formation were detected. Two different modes of crosslinking existed in the crystalline paraffins, one random and the another very selective at the edge of crystals.

Seguchi, T., Y. Katsumura, et al. 1991. "Irradiation effects on polymer-model compounds." Radiat. Phys. Chem. 37(1): 29.

Irradiation effects on n-paraffins and squalane, used as models of polymers, were investigated by product anal. Four n-paraffins, $C_{20}H_{42}$, $C_{21}H_{44}$, $C_{23}H_{48}$ and $C_{24}H_{50}$, and squalane ($C_{30}H_{62}$) were gamma-irradiated under vacuum in liquid, crystalline, and glassy states. The evolved gases were analyzed by gas chromatography and changes in molecular weight are analyzed by liquid chromatography and mass spectroscopy. G-values for crosslinking of n-paraffins were 1.2 for crystalline states (at 25°C) and 1.7 for liquid states (at 55°C), and showed no difference between odd and even C numbers. The G-value of liquid squalane was 1.7; it was 1.3 for the glassy state at low temp. (-77°). Double bonds were common in the crosslinked products, esp. after liquid-phase irradiation. The probability of chain scission was estimated as being negligible, though a small number of chain-scission products (which were products of scission at chain-ends or side chains) were observed by gas analysis.

Seki, H., R. Nagai, et al. 1968. "gamma-Radiolysis of a Binary Mixture of Methanol and Water. The Formation of Formaldehyde in the Radiolysis of Liquid Methanol." Bull. Chem. Soc. Japan 41(12):2877.

The gamma radiolysis of methanol and water over a wide range of concentrations was investigated. The observed yield of products are expressed as a linear function of the electron fraction yield of hydrogen and formaldehyde. Formaldehyde is likely produced through the decomposition of the CH_2OH radical and not by disproportionation.

Shcherbak, L. I., ard N. I. Mitskevich. 1960. "Oxidation of Liquid Paraffin Hydrocarbons in the Presence of Alkali." Sbornik Nauch. Rabot. Akad. Nauk Belorus. S.S.R., Inst. Fiz.-Org. Khim. (8):194.

The rate of autooxidation of NPH (I) (bp. 200-350, ave. mol. wt. 211) at 130°C was increased in the presence of small amounts of NaOH (greater than 0.11% Na). Above 0.3% Na the rate sharply decreased. Only after 3 to 4 h was the increase in the rate apparent: initially NPH containing NaOH was no different from pure NPH, due to NaOH insolubility in NPH. The induction factor (moles evolved of CO + CO_2/mole of O absorbed per mole of NPH) passed through a maximum of 0.13 at a NaOH concentration of 0.07% Na. Early in the oxidation mainly CO was evolved; when 0.15 mole of O was absorbed, the ratio of CO to CO_2 was 1:1; then the CO_2 increased until 0.8 mole of O_2 was absorbed and the ratio of CO_2 to CO was 1:3. Aldehyde and carboxylic acids were thought to be the sources of CO and CO_2, respectfully.

Sheldon, R. A., and J. K. Kochi. 1981. Metal-catalyzed oxidation of organic compounds. Academic Press, New York.

This book reviews the literature and expounds on the subject of metal catalyzed reactions of organic compounds.

Shiotani, M., S. Murabayashi, et al. 1976. "Spin trapping of the short-lived free radicals formed in t- irradiated alcohols." Int. J. Radiat. Phys. Chem. 8:483.

Gamma-radiation was used for radical initiation with methanol, ethanol, and isopropanol in the presence of nitroso spin-traps. The hydrogen radical adduct was observed, as well as alkoxy radicals. Methoxyl and hydrogen radicals were the primary products of radiolysis of liquid phase methanol.

Stieglitz, L. and R. Becker. 1978. "Formation of dibutyl phosphate by radiolysis and hydrolysis in the PUREX process." Conference Title: Lecture meeting on principles and applications of nuclear, radio, and radiation chemistry 16.

Stieglitz, L., and R. Becker. 1984. "Chemical and radiolytic degradation in the Purex process." Fuel Reprocess. Waste Manage., Proc. Am. Nucl. Soc. Int. Top. Meet. 1:451.

New data on the hydrolysis and radiolytic formation of HDBP, H_2MBP.

Symmons, M. C. R., and B. W. Wren. 1984. "Electron spin resonance spectra of ether radical cations generated by radiolysis." J. Chem. Soc., Perkin Trans. 2 (1984):511.

Radiolysis of dimethyl and diethylether gave primary cations; the unpaired electrons were largely confined to the oxygen. Propyl ethers provide hydrogen shifts to give alkyl radicals.

Tabata, M., J. Soma, et al. 1980. "Spin trapping of carboxylic esters s-irradiated in solid and liquid phases." Radiat. Phys. Chem. 16:369.

Spin trapping was used to examine radiolysis products from a series of acetates. The products were H-radical, RO_2CCH_2-radical and R(OAc)CH-radical, where R is H or Me.

Tanaka, T., and I. Okuzumi. 1974. "Alcohols, aldehydes, and carboxylic acids from paraffins by gamma-ray irradiation." Japan. Kokai 3.

A mixture of saturated C1-8 hydrocarbons, NO_2, and O was irradiated by gamma-rays to give alcohols, aldehydes, and carboxylic acids. Thus, in a 200-ml autoclave, 6.9 g NO2 and 3.2 g ethane were irradiated 3 h by gamma-rays at 1.8 x 10^5 rad/h to give EtOH 2.0, AcH 1.1, and AcOH 0.7 g. Similarly, 1.6 g CH4 gave 0.8 MeOH, 0.9 g HCO2H, and HCHO in approximately 4 wt.% CH_4 in the gas phase. Also, propane, butane, C_5H_{12}, C_6H_{14}, C_7H_{16}, and C_8H_{18} each gave the corresponding alcohols, aldehydes, and carboxylic acids.

Teply, J. 1969. "Radiation chemistry of alcohols, ethers, and ketones in condensed states." Radiat. Res. Rev. 1:361.

Theander, O., and D. A. Nelson. 1988. "Aqueous, high temperature transformation of carbohydrates relative to biomass utilization." Adv. Carbohydr. Chem. 46:273.

This 53 page review covers alkaline hydrolysis of most carbohydrates, including mechanisms and products. The hydrolysis of sucrose in aqueous alkaline solution leads to a number of products, many of which are not anticipated.

Tilquin, B., and T. Baudson. 1988. "Irradiation temperature effect upon crosslinks induced by gamma-irradiations of polycrystalline n-alkanes, n-undecane to n-heptadecane." Radiat. Phys. Chem. 32(3):379.

Even-odd alternation of dimer isomer distribution of a series of irradiated normal alkanes starting from undecane is due to prominent crosslinks at the ends of monomers in odd members of the series irradiated at 77 K. However, penultimate sites are also favored in opposition to ESR results. By increasing the temperature of the irradiation to 195 K, crosslinks in the central part of chains (H-links) are enhanced particularly for even homologies. The temperature dependence explains why none of the free radicals are found at the terminal position after irradiation of eicosane at room temperature.

Tilquin, B., and T. Baudson. 1991. "A chromatographic study on crosslinks induced by gamma-irradiation of alkane." Radiat. Phys. Chem. 37(1):23.

Radical formation as shown by chromatography analysis of dimer products and post-irradiation radical scavenging methods is in good agreement with ESR spectral assignments. Both methods are complementary. The chromatography technique gives more information about radicals at low concentration, such as allylic or fragment radicals, which are not detected by ESR measurements. Analysis coupled with radical scavenging is also able to detect the relatively low yield of primary radicals produced only at low temperatures in gamma-irradiated polycrystalline n-alkanes from hexane to hexadecane. The temperature dependence of crosslink age explains why crosslinks in the central part of chains (H-links) are enhanced at room temperature. It is also noted that for long-chain alkyl radicals, a disproportionation radical reaction is the main radical reaction.

Torres, L., M. El Desouky, et al. 1984. "Gamma-radiolysis of furylketones in 2-propanol solution." Radiat. Phys. Chem. 23(1-2):121.

Gamma-radiolysis of furyl ketones in 2-propanol is studied. The irradiated solutes 1-(2-furyl)-1-pentanone(I) and (2-furyl)-phenylmethanone(II) were synthesized in the laboratory. Apart from gaseous products (e.g H_2, CH_4), irradiation leads, in both cases, to the formation of homologous substitution, radioreduction and dimerization derivatives. The radiochemical yields of hydrogen and methane are lower than those obtained with pure 2-propanol. The yields of the main products formed during radiolysis of ketones (I) and (II) decrease then become stabilized as the dose absorbed increases. Concerning the mechanism, electron capture and radical processes are probably at the origin of the alcohols and pinacols, which are produced

by radioreduction and dimerization respectively; substitution products indicate radical processes.

Trotman-Dickenson, A. F., and G. S. Milne. 1967. Tables of Bimolecular Gas Reactions. National Bureau of Standards, Gaithersburg, Maryland.

This survey of literature through 1965 covers the kinetics of bimolecular and thermomolecular gas reactions that do not involve atoms or molecules in electronically excited states. Bimolecular reactions are here defined as reactions in which two molecules are involved as reactants, that yield two or more molecules as products. Reactions of oxygen and nitrogen atoms have been omitted.

Van Linde, H. J., and T. A. Du Plessis. 1977. "The chain formation of aldehydes in the radiolytic oxidation of aliphatic alcohols." Tetrahedron 33: 2099.

Oxygen saturated alcohols were radiolyzed and the yields of RCHO, H, and H_2O_2 were determined. With the exception of methanol, the yields of aldehyde and peroxide were dependent on radiation intensity.

Vladimirova, M. V., D. A. Fedoseev, et al. 1985. "Radiation-chemical behavior of actinoids in extraction systems. III. Precipitates of plutonium(IV) in the system of 30% tributyl phosphate -dodecane on gamma-radiolysis." Radiokhimiya 27(6):804.

The compounds, structure, conditions, and behavior in various polar and nonpolar solvents were examined for the ability to extract plutonium. The extraction seems to work best in a system of 30% TBP-dodecane under gamma irradiation.

Vladimirova, M. V., I. A. Kulikov, et al. (1991). "Thermal oxidation and nitration of systems of tri-n-butyl phosphate + diluent." Atomnaya Energiya 71(4):333.

Study of the acid hydrolysis of tri-n-butyl phosphate (TBP) has shown that above 383 K the nitric acid concentration in the extractant decreases because there is interest in investigating thermal oxidation and thermal nitration of extracting systems based on TBP. There is an indication that, above 403 K, liquid-phase TBP + HNO_3 forms carboxidation and nitration products; NO_2 N_2O...study the kinetics of gas evolution in the thermal oxidation and nitration of single-phase (TBP + HNO_3 and TBP + diluent + HNO_3) and dual-phase (TBP-HNO_3, TBP + diluent-HNO_3, and diluent-HNO_3) systems. Here n-paraffin (a mixture...aqueous and organic phases (v_{aq}/v_{org}). Before heating, some solutions were irradiated with gamma rays.

Vladimirova, M. V., I. A. Kulikov, et al. 1991. "Thermooxidation and nitration of the TBP + diluent system." At. Energ. 71(4):333.

The influence was studied of the concentration of HNO3 and the nature of the diluent on the gas evolution process during thermal oxidation and nitration. In single-phase systems of 30% Bu_3PO_4 + diluent + HNO_3, the rate of gas evolution increases with an increase in the HNO_3 concentration and temperature. Under very similar conditions, it is higher in solutions with a diluent of a mixture of naphthenes and n-alkanes than with a diluent of the alkane alone. In 2-phase systems of Bu_3PO_4 + HNO_3, the gas evolution rate increases with an increase in the HNO_3 concentration in the aqueous phase and with an increase in the vol. ratio nu.aq./nu.org and the dose of preliminary gamma-radiation. An equation is proposed for calculating the mean gas evolution rate and the composition of the gas mixture for a 2-phase system. For practically all the studied conditions in the gas phase there occurs an accumulation of C oxide and its concentration can reach a dangerously explosive level of 12%.

Warman, J. M. 1967. "Electron capture by nitrous oxide in irradiated alkane and alkene gases. Subsequent reactions of the oxygen(1-) ion." J. Phys. Chem. 71(12):4066.

The yields of nitrogen formed on irradiation of alkane (C2-C4) and alkene(C3, C4) gases containing small amounts (<5%) of added nitrous oxide have been measured. The yield of nitrogen, $G(N_2)$ for a given hydrocarbon increased with increasing concentration of nitrous oxide up to ~2% above which $G(N_2)$ is approximately constant. This limiting yield depends on the hydrocarbon used. The differences are attributed to subsequent reactions of the O- ion, formed on electron capture by the nitrous oxide. Rate constant ratios for these reactions have been determined by studying mixed-hydrocarbon + nitrous oxide systems and by the use of carbon dioxide and acetone as competitive scavengers of the O-ion.

Wu, J., R. Yuan, et al. 1980. "Gamma radiation effect of the systems TBP-DBBP-C_6H_6, TBP-DBPP and its energy transfer mechanism." Heh Hua Hsueh Yu Fang She Hua Hsueh 2(1):17.

Radiolysis of the system TBP-DBBP-C_6H_6 and the dependence of $G_{DBP+MBP}$/ on the composition of the system were investigated. Three solutions of $TB^{12}P$-C_6H_6 were prepared. The mole fractions of benzene in these solutions were 0.826, 0.785, and 0.916, and the mole ratios of C_6N_6 to TBP were 4.8, 3.72, 10.98 respectively. Different amounts of DBBP were added to these solutions, and then they were irradiated. The results give evidence that the apparent action of DBBP is dilution,

and the protective ability of benzene is depressed correspondingly. While treating DBBP as a diluting agent to the system C_6H_6-TBP, G value of acid products from the excited state of the second type has been found to be 0.95 ± 0.11. This state contributes about 43% of the excited states. It probably comes from the excitation of = = P = = 0 bond to higher singlet state. The protection of phenyl group on this state shows a linear relationship between the G value of DBP-MBP (G_1) versus mol fraction of benzene, i.e., ${}^2G_1 = {}^2G_0(1-x)$. The kinetic study shows that the collision mechanism predominates in the process of energy transfer, especially at lower concentration. (2) DBPP has a twofold protective effect on excited TBP, i.e., the effect of phosphoryl group and that of phenyl group. The inhibition effect of the phosphoryl group is similar to that of DBBP and that of the phenyl group is the same as benzene. It also shows a linear relationship between ${}^2G_1$ and mol fraction of DBPP, i.e., ${}^2G_1 = {}^2G_0(1-x)$. (3) Under gamma radiation, TBP may form three excited and ionic species at least, i.e., triplet state 29 ± 3%, singlet state 43 ± 4% and TBP^+ 28 ± 3%.;

Zegota, H., and C. Von Sonntag. 1977. "Radiation chemistry of carbohydrates. XV. Hydroxyl radical induced scission of the glycosidic bond in disaccharides." Anorg. Chem., Org. Chem., 32b:1060.

t-Radiolysis of a number of disaccharides in deoxygenated, N_2O saturated aqueous systems produced 5-6-carbon monosaccharides. This was due to the hydroxide radical induced scission of the glycosidic bond of the disaccharide.

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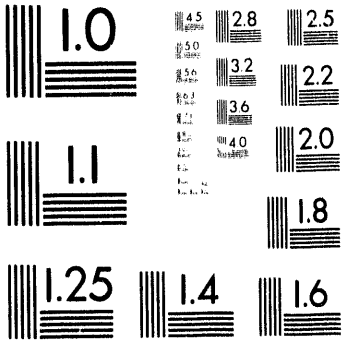
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