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The Chemistry of Radon

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We shall review the discovery of this element, studies of its chemical nature, and modern ideas on its chemical and physical properties. Possible chemical and nuclear-chemical methods of synthesising new radon compounds and of determining their properties and their identity will be discussed, using information published up to May 1980. 121 references.

CONTENTS

I.	Introduction	12
II.	Physical properties	13
III.	Clathrate compounds of radon	
IV.	Stability of radon compounds	14
V.	Special aspects of the study of radon chemistry	15
VI.	Radon compounds	15
VII.	Radiochemical synthesis of radon compounds	17
VIII.	Possible applications of radon compounds	18

I. INTRODUCTION

Only 20 years have passed since Bartlett[1] prepared the first compound of xenon. In this relatively short time many compounds of the elements of Group O of the Periodic Table have been synthesised and studied in detail. The large volume of work in this new branch of chemistry has already been reviewed several times[2-10], and the most recent work is discussed in a comprehensive review[11]. However, most of this work is aimed at the chemistry of xenon and krypton, and the chemistry of radon has attracted much less attention, no doubt as a result of the special problems presented by the chemistry of this element.

The aim of this review is to examine in detail the known chemical compounds of radon, and to discuss possible directions of further research into the chemistry of this element.

Radon, the last (and heaviest) member of Group VIIIa (the zero group) of the Periodic Table, is unique among the inert gases in not having stable isotopes. The presence of isotopes of the element "eka-xenon" in natural radioactive series was established shortly after the discovery of radioactivity. In 1899 Marie Curie found[12] that air which has been in contact with a radium compound acquires a substantial amount of radioactivity. Rutherford and Owens[13] measured the activity of thorium salts, and noted a lack of reproducibility in the electrometer readings which Rutherford ascribed to the diffusion of a gaseous radioactive substance evolved from the thorium samples. Somewhat later, Dorn observed a similar phenomenon in a study[14] of radium-containing barium salts.

Later work showed that these gaseous radioactive substances, which had been called radium and thorium emanations (from the Latin *emanatio*, discharge), are inert gases with a high molecular weight. A third radioactive gas, evolved from actinium preparations, was discovered (independently) by Giesel[15] and Debierne[16]. Shortly after the detection of the radioactive emanations from radium and thorium their chemical nature was studied in depth[17,18], using the strongest possible oxidising or reducing conditions in order to induce compound formation.

Thus, Rutherford and Soddy[17] passed thorium emanation over platinum or palladium black, lead chromate, zinc dust, and magnesium powder (at red heat) using air, hydrogen, or carbon dioxide as the carrier gas. They failed to detect any chemical reaction. Similar results were obtained by Ramsay and Soddy[18] after lengthy exposure of a mixture of radium emanation and oxygen to an electric discharge, and also after passing radium emanation over magnesium oxide at red heat.

Table 1. Radon isotopes[19].

Isotope	Type of reaction	$T_{1/2}$	Nuclear reactions yielding the given nucleus
^{202}Rn	α	13 s	^{197}Au—^{14}N—9 *n*; Pt(^{16}O); Hg(^{12}C); ^{197}Au(^{16}O)
^{203}Rn	α	45 s	^{197}Au—^{14}N—8 *n*; Pt(^{16}O); Hg(^{12}C); Au(^{14}N)
^{203m}Rn	α	28 s	^{197}Au—^{14}N—8 *n*; Pt(^{16}O); Hg(^{12}C)
^{204}Rn	α	75 s	Hg(^{12}C); ^{197}Au(^{16}O); Pt(^{16}O); ^{197}Au—^{14}N—7 *n*
^{205}Rn	α	1.8 min	^{197}Au(^{16}O); Tl(^{12}C)
^{206}Rn	α(65%), ε(35%)	6.5 min	^{197}Au—^{14}N—5 *n*; Pb(^{12}C); Hg(^{12}C); Pt(^{16}O); Tl(^{12}C); ^{197}Au(^{16}O)
^{207}Rn	α(4%), ε(96%)	10 min	^{197}Au—^{14}N—4 *n*; Hg(^{12}C); Pt(^{16}O)
^{208}Rn	α(20%), ε(80%)	21 min	^{232}Th(*p*); ^{197}Au—^{14}N—3 *n*
^{209}Rn	α(17%), ε(83%)	30 min	^{232}Th(*p*); ^{213}Ra(2,7 min)α
^{210}Rn	α(96%), ε(4%)	2,42 h	^{232}Th(*p*)
^{211}Rn	α(26%), ε(74%)	15 h	^{232}Th(*p*)
^{212}Rn	α	25 min	^{232}Th(*p*); ^{212}Fr (19.3 min) ε
^{213}Rn	α	—	^{209}Bi(^{12}C); Pb(^{12}C)
^{215}Rn	α	10^{-6} s	^{227}U (1,3 min)α; ^{219}Ra (1 ms) α
^{216}Rn	α	$4{,}5\cdot10^{-5}$ s	^{238}U α..., ^{220}Ra (10^{-2} s)α; ^{228}U (9,1 min)α, ε
^{217}Rn	α	$5.4\cdot10^{-4}$ s	^{229}U (58 min)α; ^{221}Ra (31 s)α
^{218}Rn	α	0.035 s	^{222}Ra (38 s)α
^{219}Rn(An)	α(β⁻)	4.00 s	^{223}Ra (11,2 day)α; ^{219}At (0,9 min) β⁻; ^{227}Th (18,2 day)α...
^{220}Rn(Tn)	α	51 s	^{228}Th (1,9 year)α
^{221}Rn	α(20%), β⁻(80%)	25 min	^{232}Th(*p*)
^{222}Rn(Rn)	α	3.825 day	^{230}U (21) day)α; ^{238}U α... ^{226}Ra (1622 year)α
^{223}Rn	β⁻	43 min	^{222}Rn—*n*—γ; ^{232}Th(*p*)
^{224}Rn	β⁻	1.9 h	^{232}Th(*p*)

This degree of chemical inertness was known only in gases of the "argon family", discovered shortly before radioactivity. There were therefore strong reasons for assigning the radioactive emanations to the zero group of the Periodic Table.

The term "emanation" was used for a long time as a "combined" name for the elements of atomic number 86, though even in 1912 Ramsay had proposed the name "niton", by analogy with the names of the inert gases, in order to stress the position of element 86 in the Periodic Table. In 1923 this element was given the name "radon" in keeping with international chemical nomenclature (from its longest-lived natural isotope ^{222}Rn, $T_{\frac{1}{2}} = 3.825$ days, formed by α-disintegration of radium), and the other two natural isotopes thoron Tn and actinon An became ^{220}Rn and ^{219}Rn respectively.

The nuclear characteristics of the radon isotopes known at present are summarised in Table 1 (taken from Ref. 19).

II. PHYSICAL PROPERTIES

Under normal conditions radon is a colourless monatomic gas. The earliest measurements on radon samples prepared by gas diffusion gave seriously inconsistent values for the density and the atomic weight of this element: for example, atomic weights in the range 40–234 a.m.u. were reported. Later measurements by Debierne[21], using the diffusion method, gave an atomic weight of 220–221 a.m.u. for radon, in good agreement with the value obtained by Gray and Ramsay[22] by density measurements, using a specially designed buoyancy microbalance in which small volumes (0.1 mm^3) of radon could be weighed with an error not greater than 1%. The density of gaseous radon measured in this way was 9.727×10^{-3} g cm^{-3}, from which the atomic weight of radon was calculated as 222.4 a.m.u. Liquid radon is a colourless phosphorescent substance having a density[3,20] of 5.7 g cm^{-3}. The other physical properties of radon are listed in Table 2.

Table 2. Physical properties of radon.

Parameter	Value
Boiling point, K	208 [23]
Melting point, K	202 [24]; 160 [25]
Critical temperature, K	377.5 [24]
Critical pressure, Pa	$6.12 \cdot 10^6$ [24]
Molar heat of melting, kJ	2.717 [24]
Molar heat of evaporation, kJ	16.553* [27]; 18,338** [27] 19.228* [28]
Diameter of molecule, Å	3.64 [27]; 3,70 [29]
Viscosity at 273 K, Pa s	$2.13 \cdot 10^{-5}$ [30]
at T_{crit}, Pa s	$2.954 \cdot 10^{-5}$ [30]
Absorption cross-section for thermal neutrons (^{222}Rn), barn	0,72 [31]

*Calculated from the temperature dependence of the vapour pressure.
**Calculated by Trouton's rule.

Radon is sparingly soluble in water, its solubility being higher than that of the higher inert gases. The solubility of radon in water has been measured at low temperatures[32–35], and found to be lowered by the presence of various salts[36,37]. A study[36] of the distribution of radon between carbon tetrachloride and calcium chloride solutions of different concentration has shown that the decrease in solubility (salting-out effect) is due to the change in the activity of water in solution, which affects the hydration of the radon atoms.

Radon is appreciably more soluble in organic liquids than in water[35,37,38]. The solubility in alcohols and in fatty acids has been found[39,40] to increase with the number of the homologous group in the solvent molecule.

III. CLATHRATE COMPOUNDS OF RADON

The many unsuccessful attempts at preparing compounds of Group 0 elements led to the widespread belief (among chemists) in the absolute chemical inertness of these elements[21,41,42]. Theoretical calculations of the heats of formation of hypothetical halides and oxides of the inert gases, based on a Born cycle, confirmed that these compounds cannot be formed because all the reactions which might lead to them are strongly endothermic[43]. Only for radon fluoride was a heat of formation of ~41.8 kJ $mole^{-1}$ obtained. Thus it appeared useless, at the time, to search for compounds in which the inert gases are held by an ionic or covalent bond.

However, many compounds formed by van der Waals forces are known in chemistry. Compounds of this type involving the inert gases have been known for a long time: they include argon hydrate, discovered by Villard[44], and the hydrates of krypton[45] and xenon[46], discovered by Forcrand. All these compounds were obtained by compressing the inert gas (over water) to a pressure in excess of the dissociation pressure of the hydrate at the experimental temperature: thus, according to Forcrand, the dissociation pressure of argon hydrate at 273 K is 9.61×10^6 Pa, that of krypton hydrate is 1.42×10^6 Pa, and that of xenon hydrate is 1.13×10^6 Pa. The exact composition of these compounds was not determined at the time, but on general grounds it was assumed to be $Y_b \cdot 6H_2O$, where Y_b is an atom of the inert gas[47,48].

There can be no doubt about the existence of a similar compound of radon. However, its detection by the conventional method (change in vapour pressure) would require an impossibly large amount of radon.

Table 3. Stability of the hypothetical hexahydrates $A \cdot 6H_2O$.[42]

A	$10^{-25}\alpha$	$10^{-18}\mu$	I, kJ $mole^{-1}$	p, Pa
H_2S	37.8	0.93	100	95 980
Cl_2	46.1	0.23	127	33 730
Rn	54.2	0	103	39 990
SO_2	37.2	1.6	128	33 320

A new method of preparing and studying the molecular compounds of the inert gases, based on the phenomenon of isomorphous co-crystallisation, was proposed by Nikitin[42]. The method consists in co-precipitating the experimental substance (gas) with crystals of a known molecular compound (carrier). The method allows the formation of a molecular compound of the inert gas to be studied irrespectively of its absolute concentration. Nikitin showed that if two substances have similar van der Waals force components, and similar sizes, the molecular compounds which they form with a third substance should be similar, of comparable stability, and able to form mixed crystals. This rule can be used to define series of analogues among substances giving rise to molecular compounds.

Thus, by comparing the molecular characteristics of known hydrates with those of hydrates which had not yet been prepared (including that of radon) Nikitin concluded that the stability of radon hydrate should be of the same order as the stabilities of the hydrates of chlorine and sulphur dioxide, and significantly higher than the stability of the hydrate of hydrogen sulphide.

Indeed, in a study of the behaviour of radon during the deposition of SO_2 and H_2S hydrates Nikitin[42] demonstrated the formation of radon hydrate, co-precipitated isomorphously with the above hydrates. By using ultramicroscopic amounts of radon (~10^{-11} g) Nikitin was able to study the distribution of the element between the gas phase and the crystals of SO_2 and H_2S hydrates at a temperature (269.6 K) below the eutectic points of the SO_2–H_2O (270.5 K) and H_2S–H_2O (272.7 K) systems. The distribution coefficient of radon was found to be a constant irrespective of the direction from which equilibrium had been approached and of the relative volumes of solid and of gaseous phase. This demonstrated the isomorphous incorporation of radon into the lattice of SO_2 and H_2S hydrates as a radon hydrate of similar composition ($Rn \cdot 6H_2O$).

The relative values of the dissociation pressure of the inert gas hydrates show that the stability of these compounds increases with the atomic number of the inert gas (see Table 4): this suggests that polarisability effects are dominant in determining the stability of molecular hydrates. A method of separating the inert gases can be based on the difference between their dissociation pressures[42].

Table 4. Relative stability of the inert gas hydrates[42].

Inert gas	$10^{25}\alpha$	I, kJ mole^{-1}	$10^{8}r$, cm	$10^{-4}P_{diss}$, Pa
Rn	54.2	103	2.2	3.92
Xe	40.0	117	2.18	$1.13 \cdot 10^2$
Kr	24.6	134	1.98	$1.42 \cdot 10^2$
Ar	16.3	151	1.82	$9.66 \cdot 10^2$
Ne	3.9	207	1.6	—

Molecular compounds of the inert gases other than the hydrates have also been prepared by isomorphous co-crystallisation, including compounds of radon with phenol ($Y_b \cdot 3C_6H_5OH$), toluene ($Y_b \cdot 2C_6H_5CH_3$), and *para*-chlorophenol ($Y_b \cdot 3p\text{-}ClC_6H_4OH$).[42] Attempts to prepare molecular compounds of radon with aluminium halides, analogous to the hydrogen sulphide compounds $AlBr_3 \cdot H_2S$ and $AlCl_3 \cdot H_2S$, were unsuccessful[42].

According to current views on the structure of clathrate compounds[49–51] these materials should be treated as "intercalation" compounds, in which the included gas atoms or molecules are held by van der Waals forces in holes of the crystal lattice of the "host" substance.

IV. STABILITY OF RADON COMPOUNDS

The rapid development of the chemistry of the inert gases began with the discovery, in 1962, of the xenon compounds[1]. Within a relatively short time many compounds of different members of this group of elements (though mostly of xenon) had been prepared and studied in detail. Advances in radon chemistry were on a far more modest scale: only the lower fluoride of radon (RnF_2) and some of its complexes are known at present.

A comparison of the stabilities of compounds of the various inert gases suggests that the chemistry of radon should be at least as varied as the chemistry of the lighter inert gases (in particular, xenon). Thus, the heats of formation of argon, krypton, and xenon fluorides show a clear tendency for the stability of the fluoride to increase with the atomic number of the inert gas. This is ascribed to the increase in the ionisation potential from argon to krypton and xenon (see Table 5). Hence the radon fluorides should be even more stable than the corresponding xenon compounds.

Table 5. Ionisation potentials of the inert gases[62].

Element	He	Ne	Ar	Kr	Xe	Rn
$10^{-3}I$, kJ mole^{-1}	2.371	2.079	1.519	1.350	1.170	1.034

Quantum-mechanical calculations[52] of the bond ionicity and of the stability of the inert gas halides (Table 6) show an increase in the heat of formation with the atomic number of the inert gas and with the electronegativity of the ligands. The heats of formation of the radon compounds were not calculated because the ionisation potential of the element was not adequately known, but the existence of the compounds RnF_4, RnF_2, and $RnCl_2$ was thought to be possible[52].

Table 6. Bond ionicity (a), dissociation energy (E_{diss}), and heat of formation (ΔH_f°) of some inert gas halides[81,116].

Halide	a, %	E_{diss}, kJ mole^{-1}	ΔH_f^0, kJ mole^{-1}	Halide	a, %	E_{diss}, kJ mole^{-1}	ΔH_f^0, kJ mole^{-1}
XeF_4	58	208.0	−515	$KrCl_4$	37	−24.0	571
KrF_4	47	78.7	−2	XeF_2	63	101.3	−46
ArF_4	53	46.1	225	KrF_2	63	59.0	22
NeF_4	33	−452.0	2123	$XeCl_2$	54	29.7	180
$XeCl_4$	45	66.2	213				

Similar conclusions were reached[53] in a comparison of the electronegativities of the inert gas atoms and of the ligands, which led to the following stability sequences for the compounds of the group zero elements: radon fluoride > xenon fluoride > krypton fluoride; xenon fluoride > xenon oxide > xenon chloride.

Table 7. Electronegativity of the inert gases, halogens, and oxygen[8].

Element	κ	Element	κ
He	4.53	F	3.98
Ne	3.98	Cl	3.16
Ar	2.91	Br	2.96
Kr	2.58	I	2.66
Xe	2.24	O	3.44
Rn	1.98		

The electronegativities of the inert gases, halogens, and oxygen are listed in Table 7.

These qualitative estimates of the stability of the radon halides were confirmed by thermodynamic calculations of the heat of formation of the compounds. Thus, by using

Pitzer's method[54] Vasilesku[55] was able to calculate the heats of formation of the radon fluorides and chlorides, and he concluded that the chlorides as well as the fluorides of radon are thermodynamically stable. The calculation was based on the iso-electronicity principle, according to which compounds with the same number of valence electrons have similar structures and bond dissociation energies (examples of iso-electronic pairs are ZeF_2 and IF_2^-, XeF_4 and IF_4^-, XeO_3 and IO_3^-, etc.). By using this principle we can treat the halogen monofluorides as pseudo-inert gas atoms. The enthalpy of formation of the inert gas halide XY_bX (where Y_b is an inert gas atom and X is a halogen) can therefore be related to the heats of formation of the halides of other group zero elements or to the heats of formation of their iso-electronic inter-halogen compounds by the equation

$$\Delta H_{ab}=\Delta H_a+\Delta I_{ab},$$

where ΔH_{ab} is the heat of formation of Y_bX_2, ΔH_a is the heat of the reaction $Y_a + X_2 = Y_aX_2$, and ΔI_{ab} is the difference between the ionisation potentials of the central atoms in Y_bX_2 and Y_aX_2. The heats of formation of the radon halides calculated[55] by this equation are given in Table 8.

Table 8. Heat of formation of some radon halides[8].

Standard compound	$10^{-3}\,\Delta I_{ab}$, kJ mole^{-1}	$10^{-3}\,\Delta H_a$, kJ mole^{-1}	Radon compound	ΔH_f^0, kJ mole^{-1}
ClF_3	2.21	−1.06	RnF_2	−314
BrF_3	1.05	−2.03	RnF_2	−297
XeF_2	1.39	−1.54	RnF_2	−314
XeF_2	1.39	−2.17	RnF_4	−360
ICl_3	−0.29	−0.61	$RnCl_2$	−31.8

More accurate values of the heat of formation of the radon fluorides, calculated by the same method but using more reliable values of the heats of formation of the xenon fluorides and of the ionisation potential of radon, are given in Ref.11: $\Delta H_f^\circ(RnF_2)_g = -240$ kJ mole^{-1}, $\Delta H_f^\circ(RnF_4)_g = -340$ kJ mole^{-1}, and $\Delta H_f^\circ(RnF_6)_g = -412$ kJ mole^{-1}.

Radon octafluoride should be much less stable than the hexafluoride[56], because the transition from the fifth to the sixth period of elements is accompanied by a sharp fall in the stability of the higher-valency compounds, due to the "inert pair" effect. This tendency is clearly displaced by all the elements of the sixth period from thallium to polonium[57].

As regards oxygen compounds of radon, the tendency for the stability of the compound to increase with the atomic number of the inert gas suggest that oxides of six- or eight-valent radon (iso-electronic with the known astatine forms AtO_3^- and AtO_4^-) could exist[58]. The existence of radon dioxide, which should be more stable than the (unstable) xenon dioxide, is also a possibility.

Thus there are grounds for the belief that the chemistry of radon is rich and complex: however, its study so far has been complicated by many special problems.

V. SPECIAL ASPECTS OF THE STUDY OF RADON CHEMISTRY

Experimental studies of the chemistry of radon are complicated by the unfavourable nuclear characteristics of the known isotopes of this element. The longest-lived isotope (^{222}Rn) has a half-life of only 3.825 days. Working with relatively large amounts of radon (in order to make use of the standard methods of physicochemical analysis) causes difficulties, due to the high activity and to the resulting decomposition of the compounds by auto-radiolysis under the influence of the emitted α-particles. Chemical identification of the compounds in experiments with submicroscopic amounts of radon is possible only by comparing the properties and the behaviour of the radon compounds obtained with those of known compounds (of the same type) of neighbouring elements in the Periodic Table. However, we should note that the filling of the 4*f* sub-shell in elements of the sixth period could magnify the difference in properties between the compounds of elements in periods 6 and 5 (as compared with the same differences for elements of the higher periods). Thus we can expect radon to be more "metallic" than xenon[59–61], and this expectation is borne out by the ionic form of radon difluoride (in contrast to the molecular form of the corresponding fluoride of xenon)[56,60]. Quantum-mechanical calculations[60] have confirmed that an increase in the ionicity of the fluoride should occur also between radon and element 118.

These problems in the synthesis and identification of radon compounds add further complication to the study of the chemistry of radon. Adloff, in a special publication[62] on the radiochemistry of the inert gas compounds, wrote that "the chemistry of radon will never reach the same level of development as that of xenon because of the difficulties arising from the specific properties of this element".

VI. RADON COMPOUNDS

As stated above, the most useful isotope for the preparation of radon compounds and the study of their properties is the longest-lived of the isotopes (^{222}Rn). For experiments under dynamic conditions we can also use the short-lived radon isotopes ^{219}Rn and ^{220}Rn, formed by the reactions

$$^{226}Th_{1,9\ yr} \xrightarrow{\alpha} {}^{224}Ra_{3,64\ day} \xrightarrow{\alpha} {}^{220}Rn_{54,55} \xrightarrow{\alpha} {}^{216}Po$$
$$^{227}Th_{18,17\ day} \xrightarrow{\alpha} {}^{223}Ra_{11,7\ day} \xrightarrow{\alpha} {}^{219}Rn_{3,92s} \xrightarrow{\alpha} {}^{215}Po$$

There are several methods of preparing inert gas fluorides, differing either in the choice of fluorinating agent or in the method of initiating the reaction.

Elementary fluorine, halogen fluorides, oxygen[63] or nitrogen[64] fluorides, fluorinated organic compounds[65], and some other substances have been used as fluorinating agents. The reaction can be initiated by heating the mixture, by u.v. irradiation[66,67], by an electric[68–71] or a microwave discharge[72], or by exposing the reaction mixture to γ-rays[73], to high-energy electrons[74,75], or to protons[76].

Radon fluoride can be prepared, apparently, by all these methods. It was first obtained by the most widely used of them (reaction between the inert gas and fluorine at high temperature). Heating a mixture of a small amount (1–100 μCi) of radon with fluorine at ~4×10^4 Pa pressure for 30 min at 673 K produced radon fluoride[77]. The evidence for the formation of a chemical compound was the large decrease in the volatility of radon (elemental radon sublimes easily at 195 K) produced by the admission of fluorine into the reaction vessel. The resulting fluoride had a very low volatility, and could not be sublimed in a vacuum of 10^{-4} Pa at temperatures below 500–520 K. Fluorination of a mixture of xenon and radon by a ten-fold excess of fluorine at 673 K and 3.4×10^5 Pa pressure produces di- and tetrafluorides of xenon, which readily sublime in a vacuum at 323 K, and radon fluoride which is fully retained in the reaction vessel[52]. However, when the experiment was repeated with a 100-fold excess of fluorine (573 K, 9.8×10^6 Pa) a significant amount

of radon was distilled at room temperature together with the xenon hexafluoride which was formed under these conditions[5]. The radon fluorides obtained in this work were not identified: the only comment[77] was that the product can be quantitatively reduced by hydrogen at 773 K. Micro-quantities of radon fluoride do not show any radiation-induced decomposition[77].

Chemical combination of radon by reacting with ClF_3, KrF_2, or xenon fluoride at room temperature has been reported[5], and also by reacting with oxygen monofluoride at 173 K.[78] These reactions can take place in metal, quartz, or Teflon vessels, but the reaction in a Teflon vessel yields a more volatile compound: the product of the reaction in a glass vessel is less volatile, and a still less volatile product is obtained in a metal vessel. These observations show that the surface of the reaction vessel plays an important role in the synthesis of radon fluoride.

Sufficiently large samples of radon react with liquid fluorine or solid ClF_3 (but not with chlorine) under the influence of the intrinsic α-radiation to give an involatile product. Unfortunately, a detailed description of these experiments[5] has not yet been published.

The oxidation of microgramme quantities of radon with liquid BrF_3, BrF_5, or ClF_3 also yields involatile forms of radon[61]. Between 0.12 and 0.32 Ci of radon was condensed at 73 K in a reaction vessel containing the freshly prepared fluorine halide, the mixture was thawed out, and it was allowed to warm up to room temperature over 1.5 h. Under these conditions the radon was concentrated in the lower part of the reaction vessel, and the fluorine halide which was pumped out was inactive. Dilution of an aliquot of the solution (containing 15 mCi of radon per mole of BrF_3) with fresh bromine trifluoride caused a change in specific activity strictly related to the dilution. The solutions could be centrifuged and decanted without loss of radon. The solid obtained by evaporating the solution to dryness was not identified, but the similarity between its hydrolysis and that of xenon difluoride was pointed out. It was suggested[61] that the radon is present as Rn^{2+} cations in these solutions.

The same method was used to prepare stable solutions of oxidised radon in a large number of non-aqueous solvents[79]: ClF, ClF_3, ClF_5, BrF_3, BrF_5, IF_7, an equimolar mixture of bromine and BrF_3, solutions of BrF_3 in HF and in IF_5, of BrF_5, in HF, and of K_2NiF_6 in HF. The use of arsenic, antimony, or iodine pentoxides as fluorinating agents, like the use of hydrogen fluoride, did not yield radon compounds. These observations allowed an estimate to be made[79] of the free energy of formation of radon fluoride, which must lie between the highest value of the reduction energy in the series of fluorides which oxidise radon [$E(BrF_3)$ = 167 kJ mole^{-1}] and the lowest value in the series of fluorides which fail to oxidise it [$E(AsF_5)$ = 256 kJ mole^{-1}]. When recalculated for the standard state this estimate becomes 121–214 kJ mole^{-1}.

The electro-migration of the radon compound in various solutions was studied in order to determine the charge and the chemical properties of the oxidised forms of radon[79]. The results were surprising. In pure BrF_3 solutions the radon migrates only to the cathode, but it is not discharged and it accumulates in the cathode compartment. Evidently in this solution the radon fluoride is ionised as follows[80].

$$RnF_2 \rightleftarrows RnF^+ + F^-$$
$$RnF^+ \rightleftarrows Rn^{2+} + F^- .$$

In solutions of BrF_3 in HF small quantities of radon are found in the anode compartment, and in a solution of BrF_3 in IF_5 no migration is observed. However, by saturating this solution with a strong electrolyte (KF or CsF) both cationic and anionic forms of radon are produced. This behaviour can be explained by assuming that the radon fluoride (for example, RnF_2) behaves as a Lewis base towards BrF_3 (i.e. as a donor of fluoride ions), and as a Lewis acid (i.e. as an acceptor of fluoride ions) towards compounds with a strongly developed donor function such as KF:

$$RnF_2 + BrF_3 \rightleftarrows RnF^+ + BrF_4^-$$
$$RnF_2 + KF \rightleftarrows K^+ + RnF_3^- .$$

Processes of this type are known for the fluoride of many elements in solutions of BF_3, BrF_3, BrF_5, ClF_3, etc.[81]

These workers assumed that the most likely product of the interaction between radon and fluorine or other fluorinating agents is radon difluoride, though no strict proof was given. Attempts to analyse the radon fluoride mass-spectrometrically were unsuccessful: no peaks could be recorded other than those for the full decomposition products (radon and fluorine)[79].

As stated above, one way of determining the composition of the resulting radon compounds is to compare their properties with those of the known xenon compounds. This approach has been used[82,83] to identify the chemical forms of radon produced by oxidation with strong fluorinating agents. The co-precipitation of the resulting radon fluoride with the xenon fluorides and their derivatives was studied in a special series of experiments. The macro-components were molecular crystals of the xenon fluorides themselves (XeF_2 and XeF_4), adducts of xenon difluoride and iodine pentafluoride ($XeF_2 \cdot 2IF_5$), or ionic crystals of a fluorine–xenon salt ($XeF^+Sb_2F_{11}^-$), and the solvent was iodine pentafluoride or bromine pentafluoride. The radon fluoride was prepared by treating radon with elemental fluorine and bromine pentafluoride, or with xenon di- and tetrafluoride or their solutions in iodine pentafluoride[82] or in bromine pentafluoride[83]. The co-crystallisation experiments showed that whatever the choice of fluorinating agent the radon compound obtained can be incorporated in the lattice of both molecular (XeF_2, $XeF_2 \cdot 2IF_5$)[58] and ionic compounds of bivalent xenon ($XeF^+Sb_2F_{11}^-$),[59] but not in quadrivalent xenon compounds. This suggests a similarity, both chemical and structural, between the radon compound and xenon difluoride.

Only the lower fluoride of radon is formed, apparently, because of the appearance of stable complexes of the $RnF^+MF_6^-$ type on the (fluoridised) metal surface of the reaction vessel, which prevents further fluoridation. The formation of complex fluorides is known to cause a substantial increase in the stability of inert gas fluorides. Thus, in contrast to krypton difluoride (which is unstable at room temperature), its adducts with antimony pentafluoride $KrF^+SbF_6^-$ are stable up to 353 K, and the $KrF^+Sb_2F_{11}^-$ adducts are stable up to 323 K.[84] The energy of the Kr–F bond is increased from 48 kJ mole^{-1} in KrF_2 to 135 kJ mole^{-1} in $KrF^+SbF_6^-$.[85]

Among the xenon compounds the tendency to form these adducts is more marked in the difluoride than in the tetrafluoride. The same is true, apparently, in the case of radon: furthermore, the stability of the radon double fluorides can be expected to be very much greater than that of the corresponding xenon compounds.

The radon double fluorides $RnF^+MF_6^-$ and $RnF^+M_2F_{11}^-$ can also be formed by treating radon with oxidising agents such as ClF_2SbF_6, BrF_2SbF_6, $BrF_4Sb_2F_{11}$, $IF_4(SbF_6)_3$, BrF_2BrF_6,[86] and O_2SbF_6.[87] We shall discuss these reactions and their potential practical applications in the next Section.

All attempts to prepare radon compounds other than fluorides have been unsuccessful. A study of the interaction (both thermal and photochemical) of radon with chlorine gave

inconclusive results[88]. No definite proof of compound formation was obtained in attempted reactions between radon and oxygen, ozone, or iodine monochloride[88], even when u.v. radiation and electrical or microwave discharges were used as reaction initiators[88,89]. The application of electrical or microwave discharges caused fixation of the radon on the walls of the quartz reaction vessel, possibly through the formation of very stable radonates. Thus, the radon daughter products ^{214}Bi, ^{218}Po, and ^{214}Pb are evolved when the vessel is heated to 873 K, but radon itself remains bound to the surface. However, the possibility of chemical stabilisation of the radon through the formation of oxides or silicates is not excluded[88].

VII. RADIOCHEMICAL SYNTHESIS OF RADON COMPOUNDS

In addition to the classical methods of synthesis, radiation-chemical methods and methods based on the chemical after-effects of nuclear reactions have been used in the preparation of compounds of the inert gases (β^-, β^+, and α disintegration, electron capture, isomeric transition, fission reactions, etc.). By using some of these reactions we can not only prepare known compounds but also (in some cases) predict the synthesis of new compounds.

The processes which have been most widely used in the synthesis of inert gas compounds involve the β^- disintegration of halogens incorporated in various compounds. The chemical reactions occurring during the β disintegration of atoms in molecular systems are the after-effects of nuclear, atomic, and molecular processes. The composition and charge of the nucleus (and, therefore, its nature) are altered by nucleonic processes such as the conversion of a neutron into a proton or a proton into a neutron. At the atomic level these processes can be described as

$$[^AM_z]^q \xrightarrow{\beta^-} [^AN_{z+1}]^{q+1};$$
$$[^AM_z]^q \xrightarrow{\beta^+} [^AN'_{z-1}]^{q-1},$$

where z is the charge on the nucleus, A is the mass number, M is the nucleus of the parent atom, N and N′ are nuclei of the daughter atoms, and q is the charge on the parent atom.

Table 9. Main physical characteristics* of nuclear reactions leading to the formation of radon isotopes[107].

Parent isotope	$T_{1/2}$	Type of reaction	$10^{13}E$, J	Daughter isotope	$T_{1/2}$
^{213}Ra	2.7 min	α	11.0	^{209}Rn	30 min
^{212}Fr	19.3 min	EC (56%)	—	^{212}Rn	25 min
^{219}Ra	10^{-3} s	α	12.8	^{215}Rn	10^{-6} s
^{220}Ra	10^{-2} s	α	11.9	^{216}Rn	10^{-4} s
^{221}Ra	30 s	α	10.8	^{217}Rn	10^{-3} s
^{222}Ra	38 s	α	10.4	^{218}Rn	$2\cdot10^{-3}$ s
^{223}Ra	11.2 day	α	9.2	^{219}Rn	3.92 s
^{219}At	0.9 min	β (3%)	—	^{219}Rn	3.92 s
^{224}Ra	3.64 day	α	9.1	^{220}Rn	54.5 s
^{226}Ra	1620 year	α	7.6	^{222}Rn	3.82 day

*$T_{1/2}$ is the half-life, E is the energy of the particles.

If an atom undergoing β disintegration is part of a stable molecular system, the resulting molecular system should also be stable[90] (by the iso-electronic triad rule).

Methods based on β disintegration processes have yielded a large number of inorganic (or organic) neutral (or charged) molecular forms of xenon[91–105], such as

$$*IO_3^- \xrightarrow{\beta^-} *XeO_3$$
$$*IO_4^- \xrightarrow{\beta^-} *XeO_4$$
$$*IF_5 \xrightarrow{\beta^-} *XeF_5^+$$
$$C_6H_5{}^*I \xrightarrow{\beta^-} [C_6H_5{}^*Xe]^+.$$

We should stress that organic compounds of the inert gases have not so far been prepared by classical chemical methods.

Unfortunately, in the case of radon the use of these methods is limited by the small choice of radioactive reaction chains suitable for the synthesis and identification of radon compounds. Radon isotopes are formed by the β^- disintegration of astatine, K-capture of francium isotopes, and α disintegration of radium isotopes. The main characteristics of these nuclear reactions are listed in Table 9.

We note from these results that only one case of β disintegration leading to the formation of a radon isotope is known:

$$^{219}At \xrightarrow{\beta^-} {}^{219}Rn .$$

However, the practical application of this transformation to the synthesis and study of radon compounds is complicated by the very small number of transitions leading to the formation of ^{219}At (6×10^{-5}%), the small probability of disintegration by the emission of β^- particles (3%), the very short half-lives of the parent astatine isotopes (54 s) and daughter radon isotopes (3.92 s), and our inadequate knowledge of the chemistry of astatine.

Table 10. Retention of ^{212}Rn in a chemically bound state in the disintegration of ^{212}Fr in a caesium salt[107].

Salt	Retention, %	
	in solutions	in crystals
$CsClO_4$	13±5	29±5
$CsIO_4$	11±2	25±5
$CsXeO_3F$	12±4	32±12

A more promising method of radiochemical synthesis of radon compounds is electron capture (EC) in ^{212}Fr nuclei. Following the disintegration of the nucleus by electron capture from one of the inner shells, the atoms of the daughter element acquire a large positive charge by a sequence of Auger transitions. The redistribution of this charge between the structural units of a complex molecule, and the subsequent Coulombic interaction between charges of the same sign, cause the molecule to dissociate into fragments. The neutralisation of these fragments by electrons can bring about a situation conducive to the formation of new chemical bonds. Thus, the Mössbauer effect has been used[106] to detect chemically bound forms of krypton produced by electron capture in the ^{83}Rb isotope present in isotopically enriched rubidium halides.

The possible use of electron capture processes in the synthesis of radon compounds is discussed in a study[107] of the chemical after-effects of the reaction $^{212}Fr \xrightarrow{E.C.} {}^{212}Rn$ in various caesium salts. Francium-212 ($T_{1/2}$ = 19.3 min) was obtained by irradiating a thorium target with protons of energy $E = 1.1 \times 10^{-10}$ J. The francium was removed from the target by a rapid method[108] involving co-precipitation of the francium with silicotungstic acid and separation of the francium by partition chromatography. Accumulation of

radon took place in the crystals of caesium salts containing the francium, or in solutions of these salts in 0.05 M xenon trioxide. The retention of radon in a chemically bound state is summarised in Table 10. It was suggested[107], by analogy with the xenon compounds, that the stabilisation form of the radon is the trioxide, but attempts at isolating the trioxide by coprecipitation with barium xenate or by chromatographic methods with XeO_3 as the carrier were unsuccessful.

Thus the chemical form of the trapped radon has not yet been established. However, the extent to which radon is retained in a chemically bound state strongly suggests the formation of oxygen-containing radon compounds.

The α disintegration of radium in various salts could also be a convenient radiochemical method of synthesis of radon compounds. Though α disintegration processes are far less commonly used for synthetic work than β disintegration, they have often been applied with success. For example, the formation of volatile lead compounds by the reaction $^{216}Po \xrightarrow{\alpha} {}^{212}Pb$ in the presence of gaseous hydrocarbons has been demonstrated[109]. A study of the after-effects of the α disintegration of ^{238}U in the uranylbenzoylacetonate complex has revealed a retention of $^{234}Th(UX_1)$ as high as 20–50% in acetone solution and 80–90% in the solid phase[110].

The recoil energy of the radon atoms formed by the α disintegration of radium-226 is ~1.4×10^{-14} J, and the mean free path of the radon recoil nuclei in the radium fluoride lattice is 200–300 Å.[62] Local temperature rise may favour the recombination of radon and fluorine atoms to give radon difluoride or even higher fluorides. Stabilisation of the radon formed by the α disintegration of radium in $RaCl_2$ as $RnCl_2$ cannot be excluded[62].

The first practical application of α disintegration processes to the synthesis of radon compounds was a study[111] of the stabilisation form of the radon produced by the α disintegration of ^{226}Ra in strongly oxidising solutions. The authors[111] noted differences in the amount of elemental radon extracted by hexane from a 10^{-7} M aqueous solution of $RaBr_2$ and from solutions containing HNO_3, $K_2S_2O_8$, $KMnO_4$, and other oxidising agents. A chromatographic study of the chemical state of the resulting radon indicated the formation of radon trioxide in acid media and (by analogy with the xenon compounds) $HRnO_4^-$ in alkaline solutions. However, these results were obtained only when the radon was in equilibrium with its parent ^{226}Ra: all attempts to oxidise elemental radon separated from the radium were unsuccessful[111].

On repeating these experiments it was found[112] that the whole of the "oxidised" radon can be removed from the solution by centrifuging: in other words, the decreased extractability of the radon arises not from the formation of oxygen-containing radon compounds but from the formation of ^{226}Ra colloids.

Further studies of the possible application of chemical changes accompanying the α disintegration of ^{226}Ra to the synthesis of oxygen-containing radon compounds involved the accumulation of radon in solutions of barium (radium) salts in HCl, HNO_3, HNO_3 containing XeO_3 (as a carrier for RnO_3), $HClO_4$ containing XeO_3, and also in crystals of barium (radium) perxenate[113]. However, the whole of the radon formed by α disintegration was found to be in the elemental state. The absence of oxygen-containing radon compounds was explained[113] by postulating a zero mean charge on the radon recoil nucleus.

Other types of nuclear reaction can also be used to study the possible formation of chemical compounds by radon: in particular, nuclear reactions induced by accelerated elementary particles. These methods have been used successfully in the radiochemical synthesis of compounds of the lighter inert gases. Thus, irradiation of sodium perbromate crystals with protons of energy $E = 1.6 \times 10^{-10}$ J gave high retentions of krypton in a chemically bound state, possibly through the formation of oxygen-containing compounds[114].

A very interesting study of the chemical behaviour of radon formed by irradiating uranyl sulphate crystals with high-energy protons (1.6×10^{-10} J) has been reported[107]. The retention of radon in a chemically bound state was measured by blowing off the radon after adding a reducing agent (KI solution) to the solution from which the free radon had already been removed by a stream of carrier gas[113]. The various fractions of entrained radon were collected in traps containing activated charcoal at 195 K, and the contents of all the traps were analysed in a γ spectrometer. The retention values were determined by comparing the intensities of the spectral lines given by the trap contents before and after the addition of the reducing agent (with appropriate time corrections). For the radon isotopes the retention values were less than 5%. Because of these small retentions, and of the complexity of the emission spectra, the authors[107] were unable to identify the radon compounds chemically.

VIII. POSSIBLE APPLICATIONS OF RADON COMPOUNDS

The chemistry of radon is of practical as well as theoretical interest, since it underlies the solution of one of the most fundamental problems in the protection of the environment: the purification of air from radioactive isotopes of the inert gases. The need to solve this problem has been brought into sharp focus by the growing number of nuclear power stations, and also by the serious contamination by radon of the air of uranium mines.

New developments in the chemistry of the inert gases and of inorganic fluorine-containing oxidising agents have suggested new methods of removing radioactive isotopes of the inert gases from the atmosphere. All these methods rely on chemical interaction between the inert gases and strong fluorinating oxidisers. The possibility of using these processes was first suggested[115] in 1963, and the first practical application (reported[86] in 1972) used complex salts of composition ClF_2SbF_6, BrF_2SbF_6, and $IF_4(SbF_6)_3$ as solid oxidising agents able to remove radon quantitatively from the air. In later work the choice of oxidising agents was widened very substantially. Radon can be removed from the atmosphere also by using as oxidising agents BrF_4SbF_6, O_2SbF_6, N_2FSb_6, $N_2F_3Sb_2F_{11}$,[116] $XeFSbF_6$, and some fluorine halides[117].

Among the most convenient reagents for removing radon from the atmosphere are the dioxygenyl salts (in particular, O_2SbF_6). This salt interacts with radon with evolution of oxygen according to the equation

$$Rn(g) + O_2SbF_6(s) \rightarrow RnF^+Sb_2F_{11}^-(s) + O_2(g).$$

Dioxygenyl hexafluoro-antimonate is a fairly strong oxidising agent, and it can also oxidise xenon. Using air samples artificially contaminated with ^{223}Rn and ^{133}Xe, Stein[87] showed that the radon is quantitatively adsorbed in a column (5 cm long, 6.3 mm in diameter) packed with O_2SbF_6 powder, using a gas flow rate of 45 ml min^{-1} cm^{-2}. Dilution of the dioxygenyl salt with antimony pentafluoride lowers its oxidising power below the oxidation threshold of xenon, but not of radon. It is suggested[87] that this method of chemically binding radon could find practical application for the purification of radon-contaminated air in uranium mines and for the analysis of traces of radon in the air. However, the use of this type of oxidising agent in practice is complicated by the very high reactivity of dioxygenyl salts towards common atmospheric contaminants such as water and nitrogen and carbon oxides, as well as organic compounds[118,119,120].

The interaction of radon with the salts ClF_2SbF_6 and BrF_2SbF_6 has also been studied in detail[121]. Both these oxidising agents when supported on nickel powder can effectively absorb radon from a gas stream at 213–383 K and gas flow rates up to 50 litre h^{-1}. The efficiency of radon removal is strongly dependent (as in the case of O_2SbF_6) on the presence of water vapour in the gas. Thus, the volume of air which can be freed from radon by 1 mg of oxidiser is 50–60 litre if the air is dried with P_2O_5, approximately 15 litre after drying with anhydrone, and less than 0.5 litre after drying with calcium chloride.

This very effective reaction could be used as a method of concentrating radon in the determination of ultra-micro amounts of it, and also to purify air to a residual concentration of radon isotopes not greater than 10^{-15} Ci $litre^{-1}$, i.e. well below any likely statutory threshold.

The number of radon compounds likely to find practical application is probably not limited to those listed above. Further studies of the potential uses of the compounds of the inert gases are likely to prove rewarding.

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