Treatment of gaseous effluents issued from recycling: A review of the current practices and prospective improvements

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Abstract

The objectives of gaseous waste management for the recycling of nuclear used fuel is to reduce by best practical means (ALARA) and below regulatory limits, the quantity of activity discharged to the environment. The industrial Purex process recovers the fissile material U(VI) and Pu(IV) to re-use them for the fabrication of new fuel elements e.g. recycling plutonium as a mixed-oxide (MOX) fuel or recycling uranium for new enrichment for pressurised water reactor (PWR). Meanwhile the separation of the waste (activation and fission product) is performed as a function of their pollution in order to store and avoid any potential danger and release towards the biosphere. Raffinate, that remains after the extraction step and which contains mostly all fission products and minor actinides is vitrified, the glass package being stored temporarily at the recycling plant site. Hulls and end pieces coming from PWR recycled fuel are compacted by means of a press leading to a volume reduced to 20% of initial volume. An organic waste treatment step will recycle the solvent, mainly tri-butyl phosphate (TBP) and some of its hydrolysis and radiolytic degradation products such as dibutyl phosphate (HDPB) and monobutyl phosphate (H_2MBP). Although most scientific and technological development work focused on high-level waste streams, a considerable effort is still under way in the area of intermediate and low-level waste management. Current industrial practices for the treatment of gaseous effluents focusing essentially on iodine-129 and krypton-85 will be reviewed along with the development of novel technologies to extract, condition and store these fission products. As an example, the current industrial practice is to discharge ^{85}Kr, a radioactive gas, entirely to the atmosphere after dilution, but for the large recycling facilities envisioned in the near future, several techniques are potential technologies to retain the gas, i) cryogenic distillation and selective absorption in solvents; ii) adsorption on activated charcoal; iii) selective sorption on chemical modified zeolites; iv) diffusion through membranes with selective permeability.

Introduction

Over the last 50 years the principal reason for reprocessing used nuclear fuel (UNF) has been to recover unused uranium and plutonium in the used fuel elements in order to provide fresh fuel for existing and future nuclear power plants, and thereby avoids the wastage of a valuable resource. The PUREX process is the predominant method for reprocessing commercial reactor fuel throughout the world. In the original PUREX process [1,2] both the hexavalent uranium, and tetravalent plutonium are extracted from a nitric acid solution of dissolved irradiated fuel by an organic phase composed of 30% tri-butyl phosphate (TBP) mixed with an inert aliphatic diluents such as odourless kerosene, or dodecane. The main release of gaseous radionuclides occurs when UNF is breached and dissolved in boiling nitric acid. In this paper we will focus mainly on the radionuclides iodine-129 (^{129}I), and krypton-85 (^{85}Kr), because they are likely to appear in the off-gas of an air sparge of the dissolver in a conventional Purex fuel-reprocessing flow sheet. Tritium (^{3}H) and carbon-14 (^{14}C) are also volatile radionuclides but ^{3}H has a short half-life (12.3 yrs) and its low beta decay energy (18.5 keV) makes it a relatively mild radiological hazard. No measures are currently in place to immobilise the relatively small quantities of ^{3}H arising from fuel reprocessing plants, it is commonly assumed that discharge of ^{3}H to the ocean, with subsequent isotopic dilution, is an adequate management mode at coastal sites [3]. ^{14}C is also only weakly radioactive (half-life, 5 730 yrs, beta decay energy, 156 keV) and is not abundant in fuel reprocessing plants. Like ^{3}H, it is not currently subject to control measures at fuel reprocessing plants and the requirement for separation and immobilisation is marginal. The off-gases usually contain nitrogen (~80%), oxygen (0-20%), xenon (~500 ppm), krypton (~70 ppm) and various impurities such as NOx (NO, NO_2, N_2O) gas, steam and traces of hydrogen, carbon dioxide, nitric acid, iodine and hydrocarbons [4]. Gaseous radionuclides which may become a problem in the future are tritium, krypton-85 and possibly iodine-129. While the capacity of the environment with respect to tritium, krypton-85 and iodine-129 is not well established, it has been estimated that the global concentrations of tritium and krypton-85 may approach acceptable limits during the first half of this century [5].

Concentration and recovery of iodine

^{129}I is produced by both natural and man-made sources. The natural sources include the spontaneous fission of uranium and the interaction of cosmic rays with xenon in the atmosphere. The annual production through these natural paths is approximately 10 mg/year. Up to the year 2000, the European reprocessing plants emitted about 3 500 kg (23 TBq) ^{129}I, of this 70% had been released by La Hague reprocessing plant [6,7]. A single light water reactor (LWR) will produce 234 g of ^{129}I based on uranium burn-up of 30 000 M Wd/t [8]. In handling UNF, a single 5 t/d nuclear fuel recycling facility would have to process $3.2*10^5$ g of ^{129}I per year [8]. The main factors steering the development of processes for the collection and isolation of ^{129}I are its long half-life (15.5 Ma), its low concentration standard prescribed by 10 CFR 20, and its tendency to concentrate in the thyroid gland. An additional impetus for development of radioiodine capture is the release limit for ^{129}I, prescribed by EPA criteria in 40 CFR 190, which requires a high efficiency of collection and retention [9]. Extensive work in the area of iodine removal from reprocessing plant off-gas streams using various types of solid sorbent materials has been conducted world wide over the past three decades [10]. More recently, the use of silver-containing sorbents has been the subject of considerable research [11]. The most recent work in the United States has addressed the use of silver-exchanged mordenite-type-zeolites [12]. In the industrial PUREX process, a small fraction of the total amount of ^{129}I is released into the shear off-gas as elemental iodine (I_2); however, organic iodides are produced by reactions with organic contaminates in the dissolver off-gas (DOG) and vessel off-gas (VOG) systems [10,13]. The predominant compound formed by these restrictions is methyl iodide (CH_3I), which may amount to several per cent of the total iodine in the DOG and VOG systems. In the current PUREX process, the radioiodines ^{129}I and ^{131}I can be eliminated from gaseous waste streams by counter-current scrubbing of the gas with aqueous solutions of caustic, mercuric nitrate, nitric acid or by chemical adsorption on zeolites treated with silver or on other metals with an affinity for iodine [4]. If voloxidation or thermal out-gassing is

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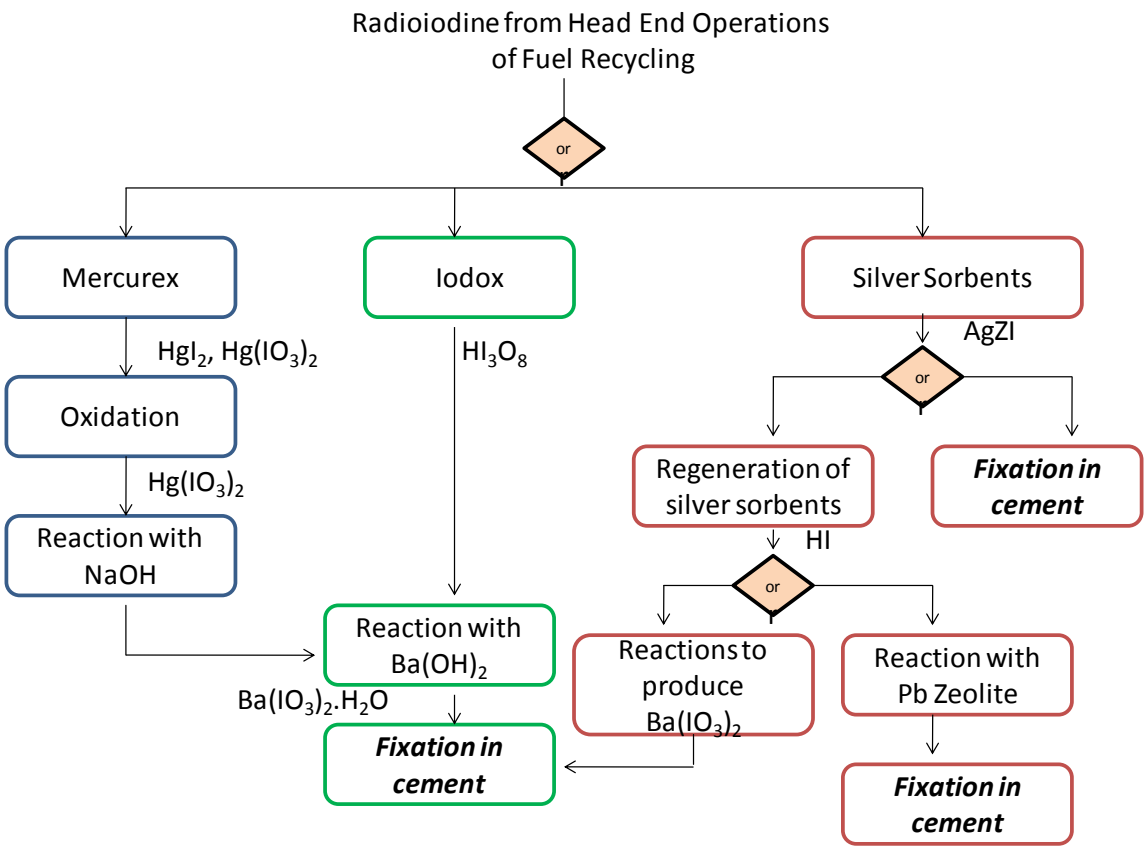
employed, a major fraction of the iodine might appear in the off-gas of these operations. If these operations are not used, most of the iodine present in UNF will proceed into the dissolver. The complex chemistry of iodine poses difficulties in its collection because iodine tends to distribute into several fuel-reprocessing streams. Without voloxidation or thermal out-gassing, most of the iodine is expected to appear in the DOG. During the past three decades, various systems have been studied to reduce the iodine release to the environment. Three liquid scrubber systems have been investigated: the Iodox system, the mercuric nitrate-nitric acid (Mercurex) system and caustic scrubbers. Solid sorbents have also been studied, either as secondary systems to provide final filtering following use of the liquid scrubbing techniques or as primary systems to replace the liquid methods. These solid sorbents include silver faujasite-type-zeolite (but it has been demonstrated that faujasite is adversely affected by water and nitric vapours), silver mordenite, alumina silicates (but there is a lack of acid resistance), and macroreticular resins. Figure 1 summarises the different separation technologies of ^{129}I from reprocessing/recycling plant. Activated carbons have also been examined as a reference material. However, activated carbon cannot be considered as a primary sorbent for treating reprocessing plant off-gas because of its low ignition temperature and its adverse reactions with nitrogen oxide which could lead to formation of explosive compounds and to poisoning by organic contaminants in the off-gas [8,10,13].

Figure 1: Radioiodine separation technologies from reprocessing/recycling plant [9]



Iodox process and products

The Iodox process [10] involves passing the DOG or VOG streams through 20 to 22 M HNO_3 in a bubble cap column. The iodine product is obtained by evaporating the nitric acid from the liquid effluent from the bottom of the bubble cap column. This results in a solid iodine product. This solid product is, however, water soluble and must undergo further treatment prior to final disposal. The recommended conversion step converts the soluble iodic acid into insoluble

$Ba(IO_3)_2$ by simple reaction with $Ba(OH)_2$. Excess $Ba(OH)_2$ should be used to ensure complete reaction and does not affect the cement stability. Up to 9 wt.% of iodine can be incorporated in a cement matrix as barium iodate. About 3 m^3 of cement would be required to immobilise the 340 kg of halogen fission products.

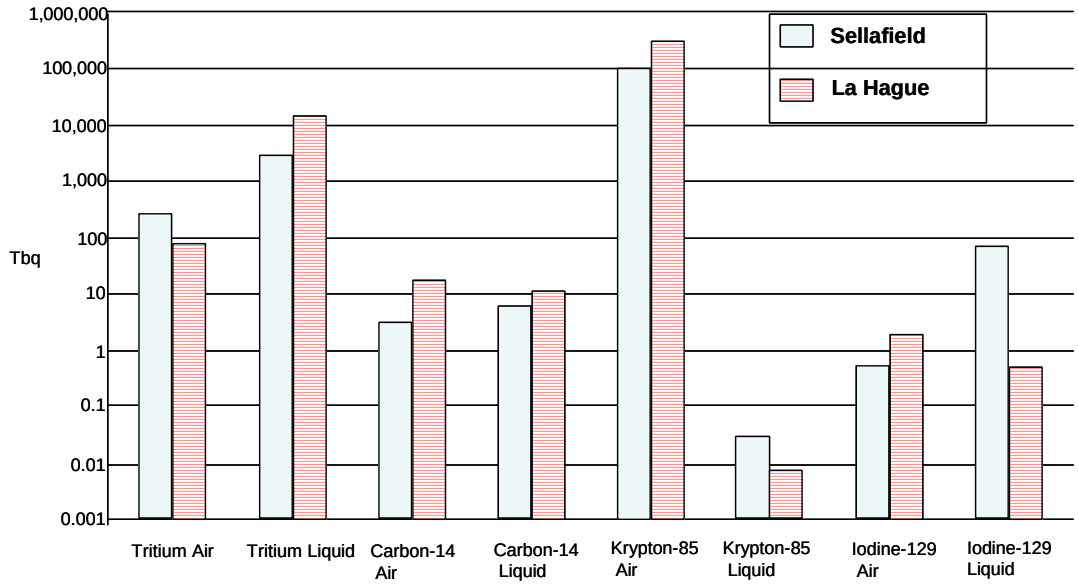
Mercurex process and products

In the Mercurex process, the iodine is absorbed in a $Hg(NO_3)_2$-HNO_3 solution and converted to iodates and mercury complexes. The scrub solution is 8-12 M HNO_3 and 0.2-0.4 M $Hg(NO_3)_2$. The used Mercurex solution loaded with iodine is treated in a subsequent process in which iodine and mercury are separated. After separation, mercury can be recycled to the Mercurex process while iodine is precipitated and conditioned for storage [14]. An alternate process involves contacting process solutions with caustic soda to produce $NaIO_3$ followed by conversion to $Ba(IO_3)_2$ as described for the Iodox process [10]. The Mercurex process followed by adsorption on silvered products was retained by SCK•CEN (Belgium) for investigation on both organic and inorganic retention of iodine compounds in recycling plant off-gases in the late 80 s.

Caustic scrubbing

Caustic scrubbing involves the conversion of elemental iodine to iodide and iodate. Organic iodine compounds pass through this type of scrubber virtually unreacted. CO_2 and NO, react in the scrubber to form carbonates and nitrates. The caustic scrubbing solutions are generally recirculated through the scrubbing tower with a small make-up stream of fresh caustic to maintain acceptable caustic and carbonate concentrations. A side stream is also generally withdrawn which contains the iodides and iodates. The iodine containing species are, for the most part, only minor constituents, and the process is generally limited by the solubility of Na_2CO_3 if NaOH is used. Variations to the process have been proposed to overcome the problems of sodium by replacement of potassium hydroxide because the carbonate form is more soluble [10]. The ^{129}I waste generated by caustic scrubbing in the United Kingdom Windscale-FRP, as well as in France La Hague (Figure 2) has been routinely discharged to the sea.

Figure 2: Discharges in 1999 from Sellafield reprocessing plant, United Kingdom and La Hague reprocessing plant, France (Tbq, logarithmic scale) [15]



Source: BNFL, 2000; Cogema, 2001

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Iodine capture on silver sorbents

The leading approach to capture radioactive gaseous iodine involves sorption onto zeolites containing silver. For several decades, the silver-containing zeolite mordenite (MOR) has been a benchmark for radiological iodine capture; however, the molecular basis for its performance remained largely unexplored. To rationally optimise the capture process, Nenoff's group [12] are presently examining the structure and distribution of the AgI when iodine is captured by silver containing MOR. Using a differential approach they have recently shown that the structure and distribution of the AgI when iodine is captured by silver containing MOR depends on whether the silver is reduced.

Iodine immobilisation

Three different waste forms were proposed for ^{129}I in the 80s [9]. Two of these utilised zeolites for capture. Two of the final waste forms incorporated cement into the system for immobilisation and the third consisted of pellets. As Trevorrow, *et al.* [9] stated in their report: "The very small release limit for ^{129}I specified by 40 CFR 190 (Table 1) makes extreme demands on technologies of collection and retention for this nuclide. Furthermore, the assessment of whether the technologies will comply with this limit is affected by uncertainties such as the distribution of iodine in the plant or the efficiency of the technology for collecting ^{129}I from gas streams" [16].

Table 1: Fraction of ^{129}I that must be isolated from the environment as indicated by 40 CFR 190

Release Limit 40 CFR 190, Ci/GWe-y	Quantity processed in UNF Ci/GWe-y[1]	Fraction[2] that must be isolated from environment [9]
$5*10^{-3}$	1.1	0.996

[1] Based on expected nuclide inventories in fuel from 2/1 ratio of PWR/BWR power plants {DOE/NE-0017} with a 1.5-y cooling period.

[2] Calculated by Trevorrow, *et al*. [9].

Concentration and recovery of krypton

^{85}Kr has a half-life of radioactive decay of 10.7 yrs and is produced by both natural and man-made sources. The natural sources include the interaction of cosmic rays with stable isotopes of Kr in the atmosphere. The man-made ^{85}Kr is principally produced during the fission reactions in LWR or during nuclear atmospheric tests; it is also released from nuclear-fuel reprocessing activities. Due to its atmophile nature, most Kr (>98%) resides in the atmosphere and becomes isotopically well-mixed within a few years [17]. There are no significant natural mechanisms (biological, physical or chemical) to concentrate krypton in the environment [18]. The annual production through these natural paths is approximately $4*10^{11}$ Bq [19]. Table 2 compares ^{85}Kr activities in the atmosphere for several accidents and annual release from reprocessing plants.

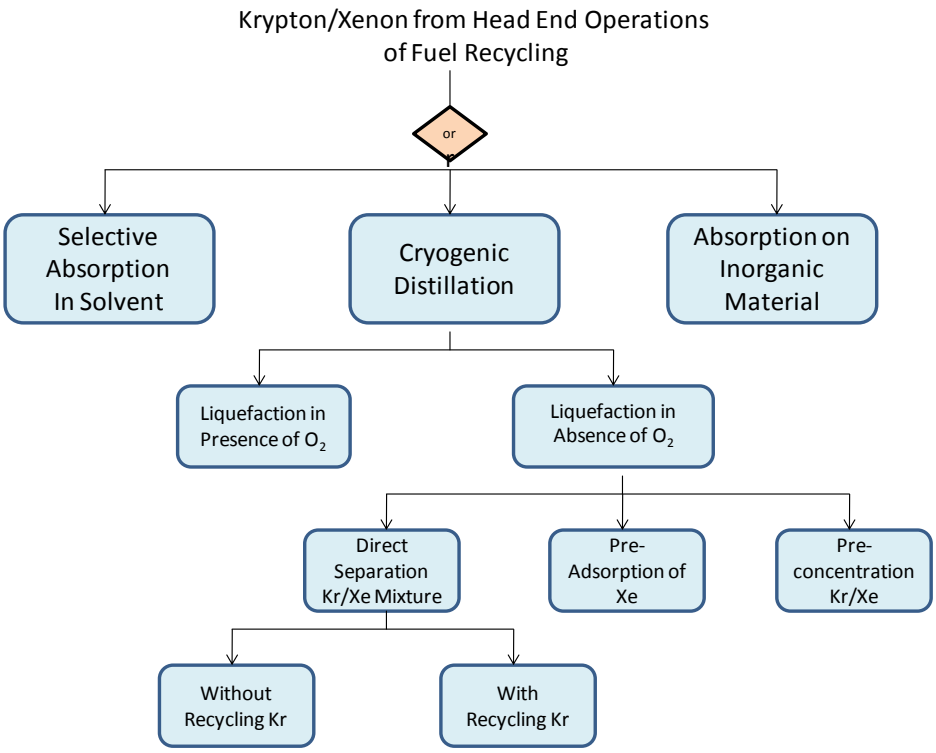
Table 2: Release of ^{85}Kr in the atmosphere [19]

Cumulated release	Activity in Bq (Ci)
Windscale accident (1957)	$5.9*10^{13}$ ($1.59*10^{3}$)
Chernobyl accident (1986)	$3.3*10^{18}$ ($8.92*10^{7}$)
Nuclear tests	$2.0*10^{17}$ ($5.41*10^{6}$)
Annual release	**Activity in Bq (Ci)**
Release from La Hague reprocessing plant, France (1999)	$2.9*10^{17}$ ($7.84*10^{6}$)
Release from Sellafield reprocessing plant, UK (1997)	$9.5*10^{18}$ ($2.57*10^{8}$)
Release from Tokai Mura reprocessing plant, Japan (1994)	$1.8*10^{18}$ ($4.86*10^{7}$)

For a few decades, ^{85}Kr artificial production shows a continuous increase of its average volumic activity into the atmosphere. In the North hemisphere the atmospheric concentration was of 0.1 Bq.m^{-3} in 1959, 0.8 Bq.m^{-3} in 1980 and in 2001, it was 1.2 Bq.m^{-3} [19]. One cannot say

that removal of tritium and krypton from gaseous waste is an immediate waste management requirement but it may become one in the future [5]. Opinions differ as to the need to isolate ^{85}Kr. Thus Geary [20] states that dispersal is almost certainly preferable to disposal, based on the relatively low inventories and hazards involved. Mellinger [21] suggested that the population risks arising from routine release of ^{85}Kr would not exceed the occupational risk associated with recovery, immobilisation and storage, and that both risks are very low. After 1 January 1983, the US EPA regulations (40 CFR 190) has limited the release of krypton-85 to the environment from commercial facilities, but criteria for final emplacement of nuclear wastes do not give specific guidance on the disposal of this nuclide. In order to comply with these regulations, krypton-85, which would be released during reprocessing of commercial nuclear fuel, would have to be collected and stored if a processing/recycling facility would be build in the US. Technology currently exists for separation of krypton from other inert gases, and for its storage as a compressed gas in steel cylinders. The requirements, which would be imposed for 100-year storage of krypton-85, have led to the development of processes for encapsulation of krypton within a stable solid matrix [9]. Satisfaction of not only final emplacement criteria but also transportation regulations will be difficult due to the large amounts of ^{85}Kr liberated in fuel reprocessing and the incomplete development of the packaging technology. A large fraction of the ^{85}Kr in UNF may be released into the shear off-gas, which will be combined with the off-gas from subsequent operations. Any ^{85}Kr remaining in sheared fuel will be released during dissolution to the DOG stream. Various methods of trapping the krypton contained in off-gases have already been considered (Figure 4).

Figure 4: Krypton/xenon separation technologies from reprocessing/recycling plant



Cryogenic distillation and selective absorption in solvents have been the subject of numerous studies [22]. Adsorption on activated charcoal has the advantage of simplicity and is widely used in reactors in the delay line for the decay of short-lived isotopes, while for its part ^{85}Kr is discharged in the atmosphere. By using two parallel beds operating cyclically in the adsorption/desorption mode it is, however, possible to trap Kr and Xe. At the same time, this method requires large volumes and fire hazards exist owing to the presence of oxidising agents.

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Cryogenic methods

In the air-products industry, cryogenic distillation is a well known technology for the commercial extraction of krypton, xenon, and other gases form the atmosphere. Cryogenic methods have therefore received much attention for the recovery of ^{85}Kr from reprocessing plants. A variety of conceptual plant designs are described in [22-23].

The cryogenic distillation process has been summarised by Bohnenstingl [24]:

- removal of ^{129}I and ^{131}I on silver-coated silica gel;
- deposition of particulate materials by HEPA-filters;
- elimination of O_2 and NOx by catalytic conversion with H_2 to N_2 and H_2O;
- drying of the gas stream with molecular sieve;
- deposition of xenon in solid form at about 80 K, while the remaining gas components are liquefied;
- enrichment of Kr by low-temperature distillation of liquid-gas mixture;
- withdrawal of the highly enriched Kr-fraction in pressurised steel cylinders for final disposal;
- purification of ^{85}Kr contaminated Xe for further industrial reuse by batch distillation.

Difficulties in this operation have been outlined by several authors [18,22,23,25]. The other volatile radionuclides should be removed from the off-gas stream before the krypton recovery stage. Thus, processes for recovery of ^{3}H, ^{14}C, ^{129}I should precede the krypton isolation process, which would be the final stage of off-gas treatment. Another issue observed is the formation of ozone due to the radiolytic decomposition of the liquid oxygen by the beta radiation emitted by ^{85}Kr. The accumulation of ozone, either alone or in combination with traces of hydrocarbons represents an explosion hazard, so most of ^{85}Kr recovery plants include oxygen removal in the pre-treatment stage. The presence of nitrogen oxides as radiolysis by-products or as components of DOG, may also react violently. Processes to remove these by-products include an alkaline wash (for NO_2 only) and catalytic decomposition or reduction. Finally the crystallisation of xenon at cryogenic temperatures appears surmountable by appropriate plant design.

Cryogenic distillation was used at the Idaho Chemical Processing Plant (ICCP) in the mid-1960s. This unit was the first large-scale plant based on the cryogenic distillation process; it was designed to recover and purify several thousand curies of ^{85}Kr daily. The process has seen extensive development work in Europe too, especially in Germany and Belgium [9]. Hutter, *et al.* [25] summarised in a 10 year development programme their cryogenic distillation investigation at the Forschungszentrum Karlsruhe, Germany. This three-stage process REDUKTION/ADAMO/KRETA culminated in a successful three-month continuous campaign and the krypton produced was stored in steel cylinders.

Other methods

Table 3 groups together various separation techniques and shows their advantages and disadvantages. Fluorocarbon absorption is a potential alternative to cryogenic distillation. It is based on the relatively high solubility of krypton and xenon, compared with oxygen, nitrogen and argon, in liquid Freon 12 (difluorodichloromethane, CF_2Cl_2). Krypton recovery efficiencies in excess of 99.9% using this method with a product purity of 97% were reached at the Oak Ridge Gaseous Diffusion Plant [18]. Following absorption, co-absorbed carrier gas is removed in a fractionator after which a rare-gas concentrate is recovered in a stripper column and the Freon is then reprocessed. One disadvantage is the CF_2Cl_2 release and its effect on the ozone layer which may constrain its use in a large-scale extraction process.

Table 3: Techniques for the separation of krypton-85 (adapted from [22])

Process	Advantages	Disadvantages
Cryogenic distillation	Reasonable cost Separation Kr/Xe Basic technology known and tested Potentially very reliable	Formation of ozone with risk of explosion Requires pre-treatment of gases
Absorption by solvents	Low cooling and solvents costs Little risk of explosion No major gas pre-treatment	Difficulties in Kr/Xe separation Corrosion risks Fluorocarbons as secondary wastes
Adsorption on activated charcoal	Simple operation Good reliability Low bed volume in the case of low-temperature adsorption	Fire and explosion risk No Kr/Xe separation High cooling cost in the case of low-temperature adsorption
Clathrates, kryptonates, chemical compounds of fluorine	^{85}Kr recovered as a solid or in a chemical compound	Poor stability

Over a dozen prospective adsorbents for krypton were studied and evaluated with respect to adsorption capacity and cost for DOG streams from nuclear reprocessing plants [26]. Results showed that, at sub-ambient temperature (-40°C to -80°C), the commercially available hydrogen mordenite had sufficient adsorptive capacity to be the most cost-effective material studied. Silver mordenite has a higher capacity for krypton retention, but is 50 times more expensive than hydrogen mordenite. The results indicated that a solid adsorbent system was feasible and competitive with other developing systems which utilise fluorocarbon absorption and cryogenic distillation. Further work by Ruthven, *et al.* [27] confirmed that a de-aluminated (i.e. aluminium depleted) hydrogen mordenite had the best selectivity for krypton of several molecular sieve materials they investigated. Excellent separation of krypton, initially present at 0.2% in N_2, was reported in a pilot plant operating at an average flow rate of 2.5 L/min.

Krypton immobilisation

Depending on regulatory criteria, ^{85}Kr, once separated should be immobilised for at least 100 years. The solid state chemistry is limited to a few highly reactive unstable compounds; therefore the immobilisation of ^{85}Kr will rely on physical methods. A variety of methods for ^{85}Kr encapsulation have been investigated such as: *i)* ion implantation/sputtering by which gases can be implanted into solids by bombarding the surface of the solid with energetic ions; *ii)* zeolite encapsulation, in which gases can be entrapped by sintering in solids such as zeolite or porous VYCOR®-type glass which have molecular defects in their structure; gas can be diffused into the solid, and encapsulated in a structural vacancy when the molecular lattice of the solid substrate is sintered at high temperatures [28].

Conclusion

This review on iodine and krypton gaseous waste management has shown that fundamental information was developed in the 1970s and 1980s. Currently, there are no plans to include krypton or iodine retention by the nuclear fuel reprocessing plants. It appears that there has been an apparent discontinuation of gaseous waste recovery process development implying that no new technologies have been developed recently. With the new fuel cycle R&D, research has restarted on this subject. It may become necessary in the future to contain ^{129}I or ^{85}Kr released from nuclear fuel reprocessing activities in order to reduce the exposure to the local population and the radioactive background throughout the world. Of course, whatever the process will be, either silver-containing zeolite mordenite for ^{129}I for example or cryogenic distillation for ^{85}Kr, the choice of the process will always depend on the plant operator and the regulatory requirements.

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