

Overview on Recent Trends and Developments in Radioactive Liquid Waste Treatment Part 1: Sorption/Ion Exchange Technique

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ABSTRACT *Radioactive liquid waste treatment is a subject that have received considerable attention worldwide in recognition of its importance for the protection of human health and the environment from adverse effects of radiation associated with these wastes. In this work, the radioactive waste management policy principles will be presented briefly, and an overview of proven liquid radioactive waste treatment technological options and approaches and their alternative designs will be provided, as part of an integrated waste management system. The paper will focus on the development of the sorption technique, as one of the most widely used techniques for the treatment of liquid radioactive wastes, presenting some of the latest important results and giving a source of up-to date literature on it. The progress in utilizing different sorbents in radioactive liquid waste treatment will be highlighted. Moreover, the most important aspects referring to new trends and visions in the application of these sorbents in liquid radioactive wastes treatment are overviewed.*

Keywords *radioactive waste; regulatory aspects; treatment technologies.*

1. INTRODUCTION

The safe management of radioactive wastes starts with planning to produce or use radioactive material and/or to operate research or power reactors. Different technical and non technical factors are affecting the waste management decision makers; these include the existence of sufficient socio-economic benefits and adequate legal, institutional, financial, and technical resources to manage the full cycle impact of the proposed activity. International Atomic Energy Agency (IAEA 2000) has identified some major legislative and governmental mechanisms to ensure the achievement and maintenance of high safety level in the management practice, these mechanisms include:

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- (1) Legislative and statutory framework must be established to regulate the safe management practice,
- (2) Regulatory body must be established and maintained independent of organizations or bodies charged with the promotion of nuclear technologies or responsible for facilities or activities. This is to ensure that regulatory judgments are made without pressure from interests that may conflict with safety,
- (3) Responsibility must be assigned to the regulatory body for authorization, regulatory review and assessment, inspection and enforcement, and for establishing safety principles, criteria, regulations and guides, and
- (4) An effective system of governmental emergency response and intervention capabilities must be established and emergency preparedness must be ensured.

Radioactive waste regulations, as most forms of regulations, involve a number of identified elements arranged in a hierarchy structure ranging from board policy principles to objects and enforceable standard (IAEA 2002). The first level in that structure is the policy principles set that serve as national commitment to address the radioactive waste issues in any country in a coordinated and cooperative manner. The International Atomic Energy Agency (IAEA) has developed a comprehensive set of principles for the safe management of these wastes. These principles are applicable to all countries and can be applied to all types of radioactive wastes, regardless of its physical and chemical characteristics or origin. They include the protection of human health and the environment, now and in the future without imposing undue burden now or in the future (IAEA1995). In addition to the internationally accepted principles, each country has its own policy principles that defined the aims and objective for the regulatory framework; these might includes administrative and operational measure i.e. control of radioactive waste generation, safety of facilities, decision-making criteria (Abdel Rahman *et al.* 2005). The responsibility of developing the overall radioactive waste management policy lies with the central government. After the establishment of the policy principles set, primary and secondary legislations are created. Generally, the main national legislative body is responsible for the development of primary legislations that might be in the form of laws and/or acts, where the development of secondary legislations that includes regulation rules, ordinance, and decrees is the responsibility of ministries or governmental department (NEA 2005). There are two philosophies that could be adopted, at the first there is a need to develop specifications, standards and guides to direct the implementer on how to implement the primary and secondary legislations. At this philosophy, the regulator has some responsibilities and the operator elaborate the detailed specifications then the reviewer and decision is made by the regulator. In the second philosophy, the regulation system is based only on the primary and secondary legislations (NEA 2005, Norrby and Wingefors 1995).

Radioactive waste management schemes differ from country to country, but the philosophical approach adopted generally is to dispose these wastes in environmentally acceptable ways. During the planning for such scheme, the collection and segregation

of wastes, their volume reduction and appropriate conditioning into a form suitable for future handling, transportation, storage and disposal should be considered. Pertinent technological activities in managing radioactive waste are schematically given in Figure 1.

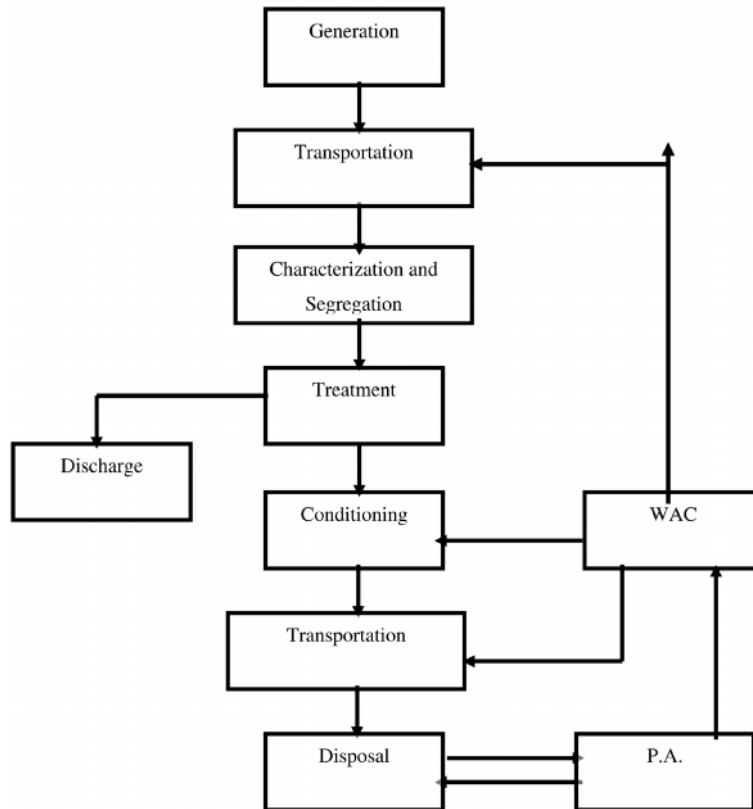


Figure 1: Schematic Diagram for Different Radioactive Waste Management Activities
Radioactive Waste Management Activities (Developed from Abdel Rahman 2005)

2. TREATMENT OF LIQUID RADIOACTIVE WASTES

Radioactive waste treatment involves operations intended to benefit safety and/or economy by changing the characteristics of the waste. The selection of a treatment technology should be based on the individual characteristics and sources of the waste, failure to account for the nature of waste may result in inadequate management practice. The selection of a liquid waste treatment system involves making decisions on a number of factors. That can be grouped into five main categories (IAEA 1984):

- Characterization and segregation of liquid waste,
- Discharge requirements for decontaminated liquors,
- Available technologies for treatment and their costs,

- Conditioning of waste concentrates resulting from the treatment,
- Storage and disposal of the conditioned concentrates.

The rest of this section will briefly present processes available for treating liquid radioactive wastes:

2.1. Pyro-metallurgical Processes

Pyro-metallurgical processes include liquid-liquid extraction using either immiscible molten metal phases or immiscible molten metal-molten salt phases, electrorefining in non-aqueous media, vacuum distillation, fractional crystallisation, melt refining, zone melting and gas-solid reactions. These processes have limited applications in fuel recycle to provide greater radiation resistance, be more compact, produce smaller waste volumes and be more proliferation resistant than PUREX process. These processes possess many disadvantages that limited its application in radioactive waste treatment i.e. they are more difficult to carry out than aqueous separations, small decontamination factors, and the produced waste forms is not suitable for direct immobilization (Ackerman, 1991).

2.2. Hydrometallurgy

Many of the treatment processes used in liquid radioactive waste management may be categorized as hydrometallurgical processes that use of aqueous reagents to soluble metals. Often the aqueous process streams are used in conjunction with liquid or solid process materials.

2.2.1. Liquid-liquid Extraction

Liquid-liquid extraction is most established separation technology for aqueous liquid waste recycling This process is principally used for large-scale operations where the concentrations of contaminants are high (see Figure 2). Solvents are becoming increasingly selective, allowing specific chemical species to be separated from the aqueous phase while others are retained. However, the capital outlay for such equipment can be expensive, large volumes of organic extractants are required and performance is often limited by hydrodynamic constraints such as flooding and entrainment, and the potential for cross-contamination of the aqueous stream with the organic solution.

2.2.2. Chemical Precipitation

The objective of this process is to remove radionuclides from liquid waste by the formation of an insoluble finely divided solid material. A typical chemical precipitation method involves the addition of reagents and/or adjustment of pH to form the precipitate, flocculation, sedimentation, and solid-liquid separation. The waste volume reduction and decontamination factors achieved with precipitation strongly depend on the method of solid-liquid separation used. Various possibilities include sedimentation and decantation, filtration or centrifugation. A wide range of different precipitants exists including metal hydroxides, oxalates, carbonates, peroxides and

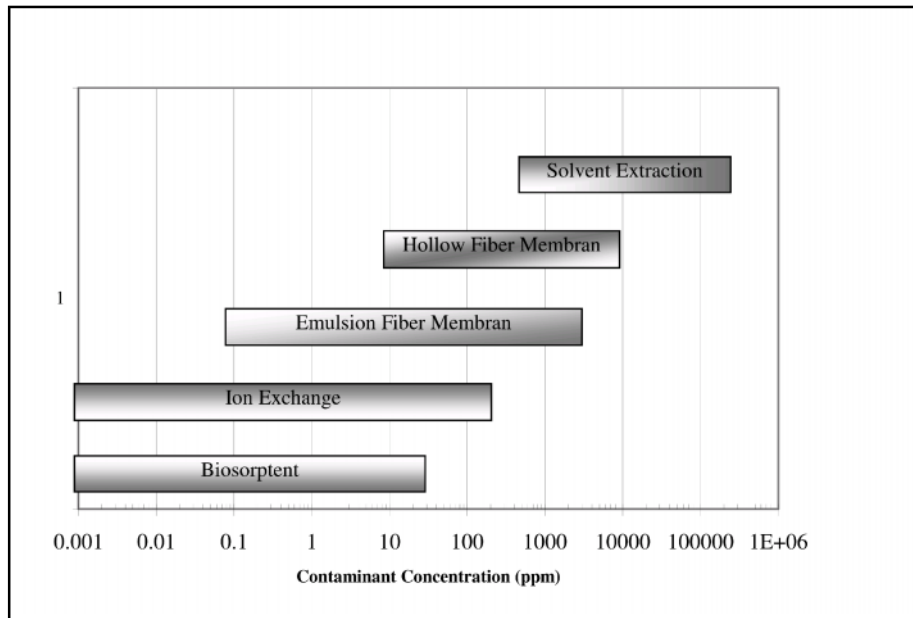


Figure 2: Decontamination Ranges for Different Treatment Technologies
(Developed from Kentish and Stevens 2001)

fluorides. Often, small quantities of selected adsorbents are added during a floc process to improve or provide decontamination for specific radionuclides (IAEA 1992).

2.2.3. Evaporation.

It is a proven method for the treatment of liquid radioactive waste providing both good volume reduction and decontamination factor. Water is removed in the vapor phase of the process leaving behind non-volatile components such as salts and most radionuclides. This process produces a clean condensate that can be discharged to the environment and a concentrate which may be encapsulated in cement or other media for long term storage or disposal. The main disadvantages of this process are high capital, energy and maintenance costs, corrosion, scaling or foaming. Evaporation is probably the best technique for waste having relatively high salt content and for nitric-acid-containing effluents, i.e. having a high electrical conductivity, a relatively low volume and needing high decontamination factors.

2.2.4 Extraction Chromatography

Chromatography is based on the principle of multiplication of single-stage separation factors by arranging the separation medium such that the products of one separation stage directly feed additional stages, thus significantly enhancing the degree of separation obtained (NEA 1997). Extraction chromatography combines the favorable characteristics of solvent extraction and ion exchange. It involves impregnating a porous granular material with a liquid that can selectively extract chemical species of interest

from solutions containing a mixture of chemical species. The granular material is placed in a column and separations are carried out in much the same way as in ion exchange, except that solvent extraction rather than ion exchange is the effective separation process. Despite the limited capacity of the reagent-loaded material, extraction chromatography is very promising, especially for the decontamination of fairly small quantities of actinide elements in liquid radioactive wastes (Eschrich, 1980). Extraction chromatographic methods with TBP and DHDECMP (Kimura 1990, Kimura and Akatsu, 1991, Mathur *et al* 1995, Abdel Rahman *et al* 2008) have been extensively applied to the separation of actinides in nitric acid media.

2.2.5. Membrane Techniques

Membranes have found extensive use in separations of radioactive materials, both as sheets and in a variety of tubular or hollow fiber forms (Office of Science and Technology, 1997, and Kurath, *et al* 1995). They may function by ion exchange, extraction, or exclusion of ions or molecules. Supported liquid membrane and emulsion liquid membrane are widely used in the treatment of liquid radioactive waste (Porter, 1990, Gupta *et al* 1995, Bilewicz and Schenker 1992, E. E. Zaki *et al* 2000, M.S. Gasser *et al* 2001). In supported liquid membrane, the solvent extraction reagent is placed in the pores of a microporous membrane support material prior to service by a soaking procedure. The supports may be either organic or inorganic, and may assume a variety of geometries ranging from beads, to sheets or plates, and to tubes, and may be single or multilayer composites. The extraction and stripping operations then proceed simultaneously by passing the feed and the stripping solution across the side of this membrane (Figure 3a) (De Gyves and De San Miguel, 1999, Yi and Tavlarides, 1992 Nobel *et al* 1989). Where in emulsion liquid membrane, the aqueous phase is encapsulated (either receiving or strip phase) within a hydrophobic membrane liquid. This emulsion is then further dispersed within the continuous aqueous feed phase (Figure 3b) (Li 1968, Ho and Li 1996, Breembroek, *et al*, 2000, Sohair A. El-Reefy *et al* 2003).

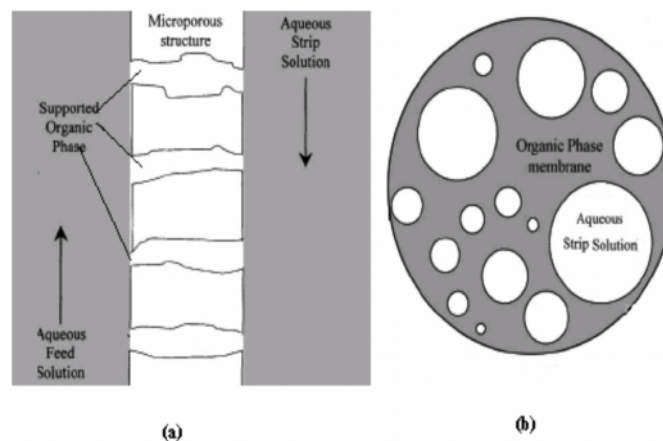


Figure 3: Schematic Diagram for (a) Supported Liquid Membrane and (b) Emulsion Liquid Membrane (Developed from Kentish and Stevens 2001)

2.2.6. Sorption/Ion Exchange

At low solute concentrations, the uses of solvent extraction or solvent-based membrane processes lose their advantage. Under this condition, sorption and ion-exchange of contaminants represent an effective alternative. The use of ion exchange procedures in water and wastewater treatments was well developed by the time this technique was first applied in the nuclear industry. Since then much progress has been made in improving the technology, and ion exchange methods have been widely used to remove soluble radionuclides from liquid waste (Huston, 1996, IAEA 1983, Navartile 1985, IAEA 1967). Most of ion exchange currently being used is commercially mass produced organic resins. Although these exchangers have a wide applicability, they have few limitations. One of their severest limitations is their poor thermal stability; i.e. the mechanical strength and removal capacity of ordinary organic ion exchange resins tends to decrease under high temperature conditions that are frequently encountered in processing liquid radioactive waste streams. In addition, degradation of ion exchange properties under high radiation field is also known to be another defect of organic resins. In order to overcome the above limitations, many investigators, have introduced composite resins consisting of inorganic sorbent and organic binding matrices.

3. NEW TRENDS AND DEVELOPMENT

To minimize the management costs, it is important to minimize the volume of the secondary waste to be conditioned and disposed and to change the characteristics of the main waste stream so it could be discharged or recycled in a technically less demanding and less costly manner. Environmental aspects are also very important, since the regulations on discharges are becoming increasingly more stringent. Research and development activities are developed worldwide to address these aspects this includes the preparation and evaluation of improved material and combined processes for the treatment of these wastes. In this section, recent development in the radioactive liquid waste treatment will be presented.

3.1. Development of Material with Sorption-combined Properties

3.1.1. Composite Materials

Composite material consists of one or more sorbents combined with another material, which can be inorganic or organic and may be a sorbent. The produced material is granular with sufficient strength for column use from sorbents that do not form or only form weak, granules. Inorganic-organic sorbent consisting of an inorganic ion exchanger-modified polyacrylonitrile (PAN) binding matrix was proposed as composite material for the treatment of ILLW (Sebesta 1993, 1994, 1997, 2003). PAN beads are highly porous and can accommodate very high loadings of ion exchange material (5-95%). Fisera and sebesta (2006) reported that highly porous PAN beads improve the sorption kinetics and sorbent capacity owing to the increased availability of the sorbent material, easy modification of physico-chemical properties (hydrophilicity, porosity, mechanical strength) and simplified production.

3.1.2. Sorption-reagent Material

Materials having sorption/reagent properties have the capability to interact with the waste solution components and employ dual features of co-precipitation and sorption (IAEA 2003). Avramenko (1999) indicates that the sorption selectivity for different radionuclides such as strontium, cobalt, mercury, iron, or manganese from liquid waste containing highly complexing agents on Barium amorphous silicate increase by tens times when compared to standard ion exchanger resin due to the radionuclide co-precipitation on micro-particles formed inside the pores of the inorganic matrices in addition to acting as a sorbent.

3.1.3. Bio-sorbents and their Modifications

In bio-sorption process, biological materials, such as chitosan (Bassi *et al.*, 2000, Kurita *et al.*, 1979, Volesky and Holan 1995), marine algae or alginates (Chu *et al.*, 1997, Matheical and Yu 1999, Hashim *et al.*, 2000, sag *et al.*, 1995, yang and Volesky 1999), fungi or bacterial biomass (Hermann 1999, Kogetv 1998) are used as a chelating ion-exchange medium. Various naturally occurring biosorbents are widely applied specifically for the sorption of long-lived radionuclides. Among them, chitin-containing sorbents have been found to be very efficient on account of their abundance and the textural characteristics. Kosyakov *et al.* (1997) indicates that these chitin containing sorbents have fiber structure consisting of about 70% of chitin, 25% of R-glucans and 5% of melanin located in the cell walls could be attached to selective inorganic ion exchangers, e.g. ferrocyanides for cesium and impregnated KMnO_4 for strontium.

3.1.4. Inorganic Exchangers-Titanium Oxide

Inorganic exchangers have been investigated extensively for the treatment of radioactive waste. Recent studies reported the ion exchange kinetics on these materials, in this concern, the isotopic exchange of common metal ions on hydrous oxide has been studied extensively (H. F. Aly *et al.* 1998 and 1999, El-Naggar I. M. *et al.* 1996 and 2002, E.S. Zakaria 2004). Titanium oxide based materials will be presented in this section as representative inorganic exchangers.

Hydrous Titanium oxides are inorganic exchanger that has been investigated extensively for the treatment of radioactive waste, the crystalline structure of this material has a strong effect in the ion exchange properties (E. Metwally *et al.*, 2007, Ayoub 2005, Shabana, and El Dessouky , 2002). In alkaline solutions, it acts as a cation exchanger, while in acidic solutions, it behaves as an anion exchanger. Therefore, the isoelectric point (or point of zero charge) of the TiO_2 sorbents is one of their important characteristics. Samanta (1996), and Lehto (1996) indicate that for various materials the isoelectirc point, it may range from $\text{pH} \sim 1$ to $\text{pH} \sim 6$.

Photochemical mineralization of organic materials by ultraviolet radiation has been known as waste water treatment method (Schiavello, 1997). The photo-degradation efficiency can be enhanced by introduction of a heterogeneously dispersed semiconductor photo-catalyst in the system. Titanium dioxide has band gap energies sufficient for catalyzing a wide range of chemical reactions. Photo-catalytic degradation

of decontamination solutions including EDTA, oxalic and citric acids complexes using titanium dioxide based sorbents was quite efficient. Nearly all the organic complexants such as oxalic and citric acids and EDTA could be effectively degraded. The degradation was more efficient in acidic solutions, and was very slow in neutral and alkaline pH range (Danacikova *et al* 1999, Sebesta *et al* 2003).

3.1.5. Nano-sized Magnetic Materials

Nano-sized magnetic chemisorbents can be promising candidate materials for treatment of low-level effluents resulting in effective decontamination and very high volume reduction of radioactive wastes. These sorbents also enhance the kinetics of radionuclide separation from liquid radioactive wastes. Hexacyanoferrates (II) are well known for their sorption properties (Ismail 1998, 1999, Ishfaq 1997). Potassium nickel (II) hexacyanoferrate (II) is a weak magnetic complex, the magnetic property being mainly due to paramagnetic nature of nickel ions. To impart superior magnetic property, the complex must be coated to a base magnetic material to economize on the magnetic separator cost requirements. If the base material is super paramagnetic, it can be flushed off once the field is switched off with no remnant magnetization thereby economizing on the filter regeneration requirements. However, the super paramagnetic particles are very fine sized with maxima around about 30 nm. These particles are highly susceptible to fluid drag forces and therefore very high magnetic fields would be required to capture these particles. A compromise between the magnetic force and Brownian force is to be considered in designing material for magnetic separation.

Potassium nickel (II) hexacyanoferrate (II) -loaded magnetite was synthesized and characterize for application in the removal of cesium from aqueous waste by magnetic means. Ambashta (2003) reported that loading low concentration of hexacyanoferrate (II) on magnetite ensures 100% loading of magnetic adsorbent on filtration matrix. This study indicates the possibility of the presence of hexacyanoferrate (II) as a coating on magnetite rather than admixture composite. In high alkaline range of pH 12, the coating prevents magnetite from becoming colloidal since it does not directly come in contact with solution. The prevention of formation of colloidal magnetite helps in improving efficiency of column uptake as the influence of Brownian forces is reduced.

New composite sorbents containing magnetite particles and ion exchange resin was prepared and its utilization for the separation of Cr (VI) from wastewater was studied (Sheha 2008). The presence of the magnetic particles in these sorbents increase their granular strength, and thermal stability. The kinetics and thermodynamic of the reaction was studied in batch mode, it was found that the sorption process is exothermic. The negative Gibbs free energy values confirm the feasibility and spontaneous nature of the process.

3.2. Development of Combined Processes for the Treatment of Liquid Waste

Recent developments have greatly extended membrane types and applications to include inorganic membranes, supported liquid membranes and polymeric membranes. Zakrzewska-Trznadel *et al* (2001, 2002, 2004) prepared ceramic membranes

made of titania and Zirconia, these membrane were tested successfully in laboratory and pilot plant scale. The laboratory experiments showed that for removal of radioisotopes that are present in radioactive wastes as small ions, a membrane process has to be combined with chemical complexation, chelation or sorption on seeds. The hybrid process is known in the literature as "seeded ultrafiltration", which is able to remove metals and other impurities from the wastes as efficiently as reverse osmosis. The pilot plant experiments include a ceramic 23-channel module. The results showed that the feasibility of ultrafiltration/complexation hybrid method for reduction of long-lived radioisotope concentration in the effluent.

Xiguang Su *et al* (2003) proposed the application of coagulation/precipitation, evaporation and ion exchange processes for the treatment of radioactive waste. The schematic diagram of this process is shown in Figure 4. The results demonstrate that the sorption precipitation process is suitable for the treatment of low level radioactive liquid waste containing high concentrations of ^{137}Cs and ^{134}Cs . In this concern, the separation of the solids and solution can be effectively reached by ultrafiltration. In the arrangement, where the ultrafiltration module inlet and outlet are interchanged during the operation, and therefore higher average flux can be achieved.

Single stage processes combining two or more treatment techniques that were developed for the treatment of radioactive waste included electro-sorption (Sarfazai and Ghoroghchian 1994, Trainham and Newman 1997, Karlin 1998, 1999). This method combines in a single stage process migration of ions under an electrical field with simultaneous sorption of radionuclides on a suitable sorbent. Conventional sorbents to be used effectively for treatment of liquid radioactive waste, must be sufficiently strong, have low hydraulic resistance, low adhesion to suspended and colloidal mixtures, and a well developed surface for fast internal diffusion of ions. In this method, the sorbent is placed between two porous diaphragms, which could be microfiltration or ultrafiltration membranes. The waste solutions are made to contact their outer surfaces and under the influence of the electrical field, the ions migrate and are picked up on the sorbent without the liquid passing through the sorbent. Electrosorption takes advantage of a combination of the high surface area and the electrical conductivity of porous carbons. It has been suggested as a minimally polluting, energy-efficient, and potentially cost-effective method for ion exchange, reverse osmosis, electrodialysis, and evaporation.

Kim *et al* 2002 studied the electrosorption of cobalt and strontium onto a porous activated carbon fiber (ACF) to treat radioactive liquid wastes resulting from chemical or electrochemical decontamination and to regenerate the spent carbon electrode. The schematic diagram of the electrochemical cell is illustrated in Figure 4. Result of batch electrosorption experiments showed that applied negative potential increased adsorption kinetics and capacity in comparison with open-circuit potential (OCP) adsorption for the studied radionuclides. The adsorbed radionuclides released from the carbon fiber by applying a positive potential on the electrode, showing the reversibility of the sorption process. The possibility of application of the electrosorption technique to the separation of radionuclides was examined. The result of selective

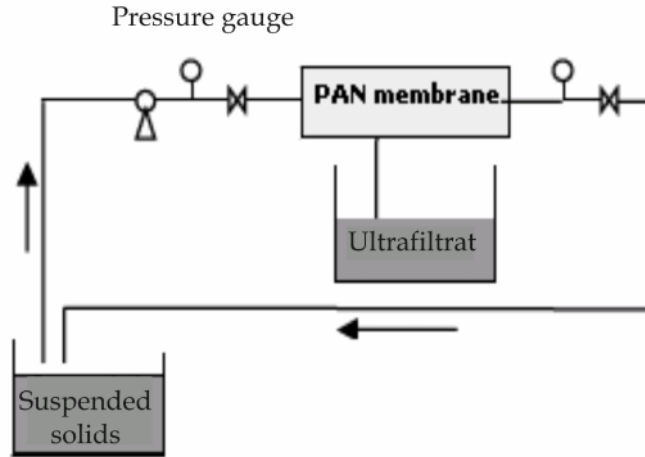


Figure 4: PAN Membrane

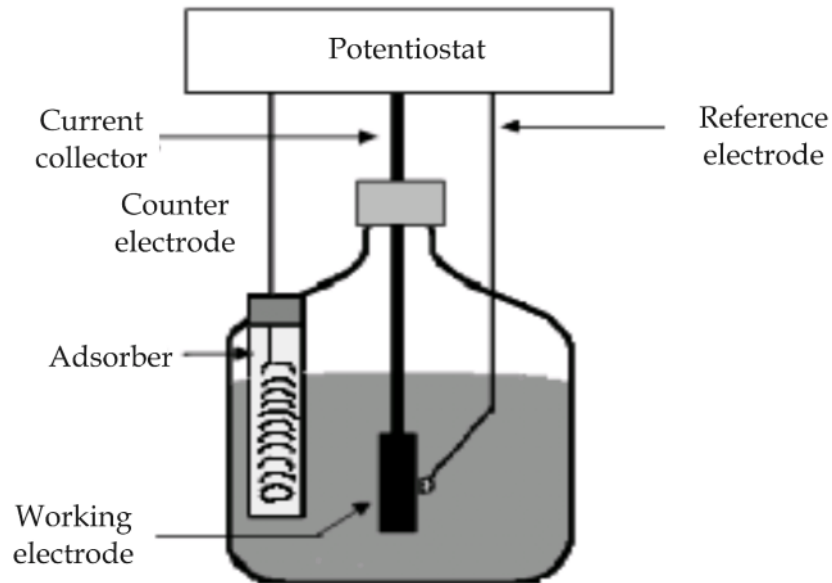


Figure 5: Schematic Diagram of Batch Adsorber (Developed form Kim *et al*, 2002)

removal experiments of a single component from a mixed solution showed that perfect separation of cobalt and strontium was achieved.

Jung *et al* (2005) confirmed that the electrosorption process using an activated carbon fiber felt electrode was effective in the removal of uranium to treat uranium-containing sludge. The selective adsorption of uranium in the Na^+ , Ca^{2+} , and NH_4^+

cations was obtained at -0.9V. they attribute the electrosorption behaviors for U(VI) to the double layer charging, and an electrochemical precipitation of the uranyl ions.

Tran *et al* (1997) studied the electrosorption capability of the carbon aerogels is the selectivity for Cr (VI) over naturally-occurring alkali metal cations, halide anions, and oxyanions in 530 ppm TDS ground water. The selective removal of uranium, cadmium, lead, nickel, and cobalt, has also been demonstrated.

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