Review

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Abstract: Although a slightly radioactive element, thorium is considered very toxic because its various species, which reach the environment, can constitute an important problem for the health of the population. The present paper aims to expand the possibilities of using membrane processes in the removal, recovery and recycling of thorium from industrial residues reaching the municipal waste processing platforms. The paper includes a short introduction on the interest shown for this element, a weak radioactive metal, followed by highlighting of some common (domestic) uses. In a distinct but concise part, the bio-medical impact of thorium is presented. The classic technologies for obtaining thorium are concentrated in a single schema, and then the speciation of thorium is presented with an emphasis on the formation of hydroxo-complexes and complexes with common organic reagents. The determination of thorium has been highlighted both on the basis of its radioactivity, but especially through methods that call for extraction followed by an established electrochemical, spectral or chromatographic method. Membrane processes are presented based on the electrochemical potential difference, rapidly presenting barro-membrane processes, electrodialysis, liquid membranes and hybrid processes. A separate sub-chapter is devoted to proposals and recommendations for the use of membranes in order to achieve some progress in urban mining for the valorization of thorium.

Keywords: thorium removing; thorium recovery; thorium recycling; thorium separation; thorium transport; thorium separation; thorium membrane separation; thorium membrane concentration; thorium determination

1. Introduction

Thorium is a relatively exotic element, although it is known to have a significant natural abundance, compared with that of lead [1]. Component of the actinide series at position #90 and a weight of the gram atom equals 232.03, it is unstable (radioactive) in all its isotopes except isotope ^{232}Th [2,3]. The half-time of ^{232}Th is so long, that it is considered stable joining uranium, which also occurs naturally [4]. The interest in thorium as a nuclear material should have justified an increased interest both on the part of researchers and on the part of energy or armament producers [5–7].

However, the number of existing publications highlighted in Google Scholar [8] on various key-terms of scientific interest is relatively moderate or even low (Table 1):

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Table 1. The number of publications highlighted in Google Scholar on various key-terms related to thorium.

Keywords *)	Publications number on periods		
	Any time	2014-23	2021-23
Thorium separation	162,000	82,000	12,900
Thorium concentration	199,000	12,200	6,200
Thorium recovery	79,000	17,900	9,200
Thorium removing	62,000	17,500	13,800
Membrane thorium separation	21,900	10,600	3,730
Membrane thorium concentration	27,600	19,000	4,610
Membrane thorium recovery	18,000	8,600	3,850
Membrane thorium removing	21,600	12,000	4,500
"Thorium separation"	883	244	79
"Thorium recovery"	611	204	87
"Thorium recycling"	50	18	4
"Thorium membrane"	7	2	-

*) accessed on June 24th, 2023.

Analyzing Table 1, researchers would be discouraged to start research on aspects of recovery, recycling or removal of thorium from various sources, but its common applications have determined its presence in urban waste, in surprisingly high concentrations.

This last observation led to the initiation of this paper, whose aim is to warn both the researchers and environmental officers on the danger of uncontrolled spread of thorium as well as the proposal of simpler solutions of removal, recovery and recycling, based on processes very close to the "urban mining".

2. Applications of thorium

Thorium, but especially thorium dioxide, have found relatively numerous applications for a radioactive element, even if this radioactivity is weak [9,10]. Various and unexpected, many abandoned, the applications of thorium are so common (Figure 1), that they have become dangerous [11], especially since after the use of various materials and in the conditions of inattention in recycling or selective collection, thorium ends up in the environment [12].

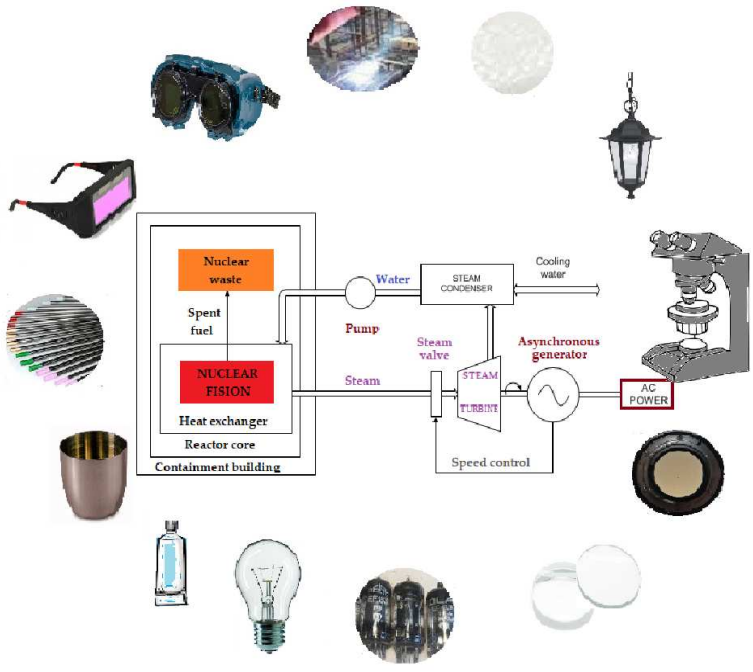

Figure 1. Domestic applications of thorium and thorium dioxide, along with the alleged use in generating energy in nuclear power plants.

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From its surprising use in toothpaste, the dating of hominids, contrast agent in certain radiological examinations or as a filament in the incandescent light bulbs, lamps, lanterns, thorium mantles [13] it is also used technically for quite solid applications and in which it is practically irreplaceable: crucibles for high temperatures, welding electrodes and alloys (aluminum, magnesium, steel), lamps for special electronic equipment, mantles in the metallurgical industry, industrial catalyst (ammonia, sulfuric acid, cracking hydrocarbons), the manufacture of thorium mixed oxide tablets and uranium, oxygen detector, lenses for various optical and opto-electronic devices (excellent wavelength dispersion and high refractive index) [14,15].

We can conclude that thorium, although radioactive, will be found in the aerospace industry, the automobile industry, the chemical and metallurgical industry, the electrotechnical industry, the electronic industry, dentistry (cements for dentistry, optical and surgical instruments, manufacturing), but also in the execution of art objects (alloys, jewelry, sculptures, statues) [16,17], which will certainly lead to finding thorium as an environmental pollutant [18–20].

3. Toxicity and bio-medical implications

Thorium is included on the list of carcinogenic substances [21], even though it was not always considered that it decomposes through alfa decay [22], and the emitted alfa radiation cannot penetrate human skin [23].

The dangers associated with its radioactivity, due to the use of thorium in various technologies that capitalizes on the high melting of thorium dioxide, lead to [24–28]:

- the amounts of thorium in the environment can be accidentally increased during processing;
- humans absorb thorium through food or drinking water (in areas adjacent to mining operations);
- the quantities in the air are very small (insignificant and generally neglected);
- near hazardous waste storage or processing sites;
- industrial laboratories or mining laboratories, milling minerals containing thorium.

The medical effects, observed over time, of those who acquire thorium at work [29–32]:

- the chances of developing lung diseases;
- occurrence of lung and pancreas cancer;
- changes in the genetic material;
- blood cancer;
- develop liver diseases (when injecting thorium for *x*-rays);
- stored in bones (long-term exposure) can lead to the generation of bone cancer.

Being a heavy metal, the medical effects of thorium as well as the precautions for working with it must be included in this field [33,34].

At the same time, natural thorium is in secular equilibrium with its descendants, which makes it necessary to consider their radiotoxicity, for this reason being classified among the most dangerous radionuclides [35].

4. Classical technology

Thorium is found in monazite (1 to 15%) in concentrations that allow it to be exploited on industrial scale, through classical technologies [36,37]. At the same time, thorium will appear in mining processes, especially those aimed at obtaining rare earths or uranium [38–40].

The combined scheme in Figure 2 shows the main operations that lead to obtaining thorium from monazite through the acid or basic attack of impurities, but in principle any mineral is considered as a source of thorium, the series of technological operations being the same [41–48].

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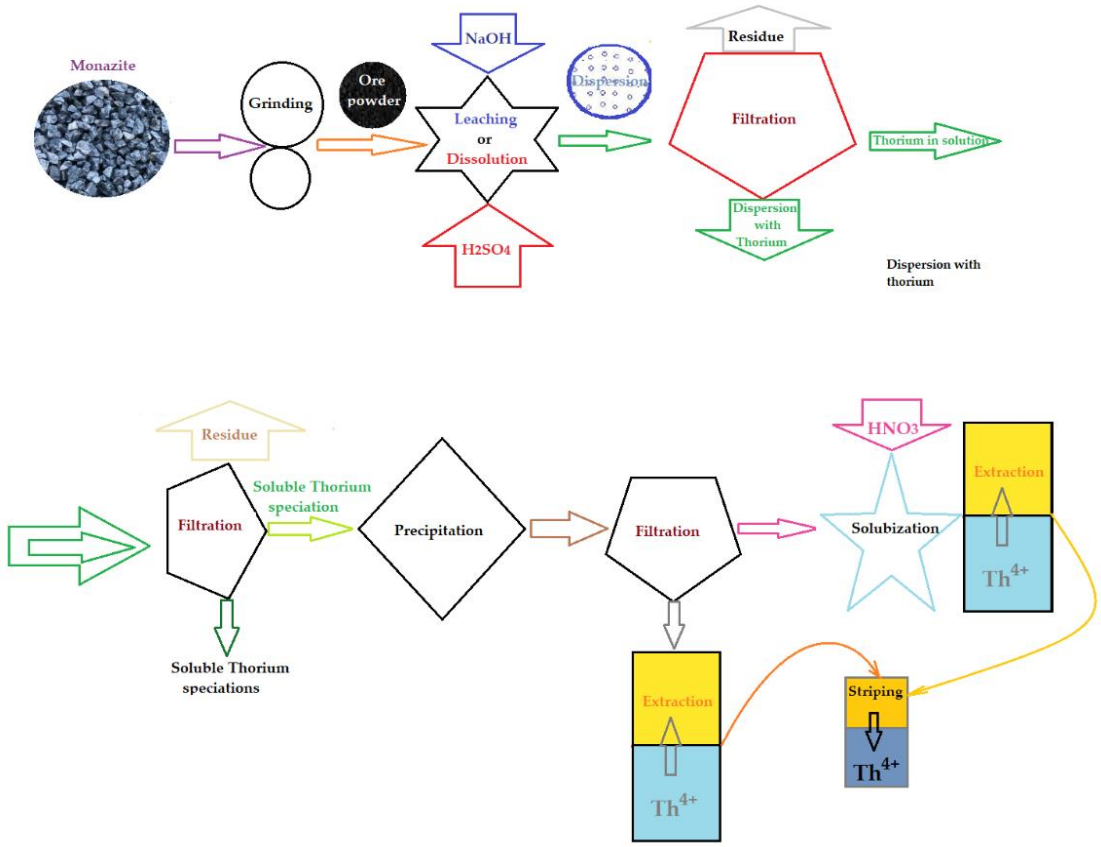

Figure 2. The combined scheme of the procedures for obtaining thorium by acid and/or basic attack of monazite.

Practically, the mineral (source of thorium) is brought to a state of fine grinding in order to be attacked by (sulfuric acid) or basic (sodium hydroxide), so that the parts of the mineral not containing thorium pass into the solution, while others are removed by filtration. The filtrate containing thorium (colloidal) can be directly processed (when purification is not done in this technology) or precipitated, filtered and finally subjected to purification by extraction in an organic solvent (TBP) [49], or an organic solvent and an amine or a selective complexant and re-extraction [50].

If the source of thorium is a mineral containing rare earths or the residue obtained during the processing of various minerals in order to obtain rare earths (REE), then the basic procedures used in the separation, concentration and purification of thorium are: leaching [51–53], precipitation [54–57], solvent extraction [58,59] and ion exchange [60].

Obtaining thorium from pure compounds (halogens, halides) or alloys can be done by physical (thermal) or chemical (reduction) processes [61,62].

For the current work that involves obtaining thorium from industrial residues or by-products (waste), the diagram in Figure 2 presents, as narrow technological points, the filtrations and extractions, operations likely to be replaced to avoid environmental pollution [63,64].

5. Thorium speciation

Thorium compounds are relatively few compared to other elements, even the less reactive ones [65,66]. Thus, thorium dioxide, halogens or a nitride are encountered, but the speciation of the thorium ion (Th^{4+}) in aqueous solutions is of practical importance, when countless hydroxylated chemical species can be generated: $[ThOH]^{3+}$, $[Th(OH)_2]^{2+}$, $[Th(OH)_3]^{+}$, $[Th(OH)_4]$, $[Th(OH)_2\ (CO_3)_2]^{2-}$, $[Th_2(OH)_2]^{6+}$,.$[Th(H_2O)_9]^{4+}$ [67–69]. Hence the importance of the operational parameters (pH, ionic strength, temperature, contact anion -F^-, NO_3^-, CH_3COO^-, Cl^-, CO_3^{2-}, SO_4^{2-}, HPO_4, CDTA, NTA DTPA, EDTA, EGTA, HEDTA, IDA, Humic acids, Citric acid, Succinic acid, Malonic acid, Oxalic acid) in the aqueous solution [70–73], which would be the object of the study in membrane processes.

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When dissolving thorium nitrate (for example) in water, the mentioned hydroxyl species are formed, but also combinations that may include carbon dioxide (present in the environment). Considering the formation of only thorium hydroxides in aqueous solution, a series of chemical species are formed as a result of some equilibria with proton exchange which is dependent on pH can be shown hypothetically by the diagram in Figure 3. The degree of formation in solution of various chemical species can be determined exactly if the acidity constants of the chemical species and/or stability constants of the hydroxyl complexes are known [57].

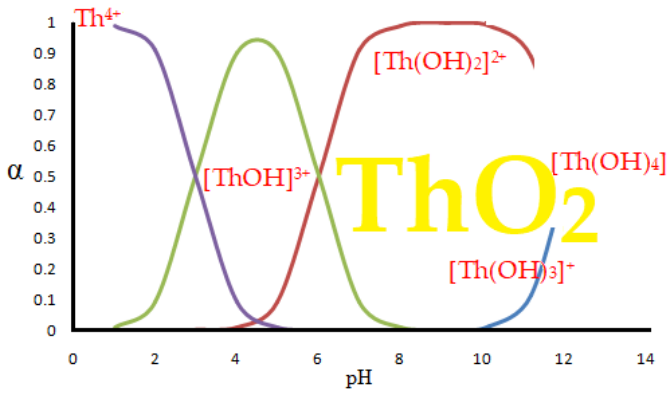

Figure 3. Hypothetical stability diagram of thorium hydroxo-complexes in aqueous medium.

The appearance of thorium dioxide is also related to the pH, ionic strength and temperature of the solution, but as a solid phase, it depends essentially on the concentration of thorium obtained at a given moment in the phases of a technology, but more importantly, on the distribution of thorium in the environment [70,73].

At the same time, if we consider that thorium is obtained in the source solution as Th^{4+} ion, then a wide series of organic complexants (Figure 4) can contribute to the formation of some speciations involved in the concentration and recovery, especially through extraction, of thorium [74–85].

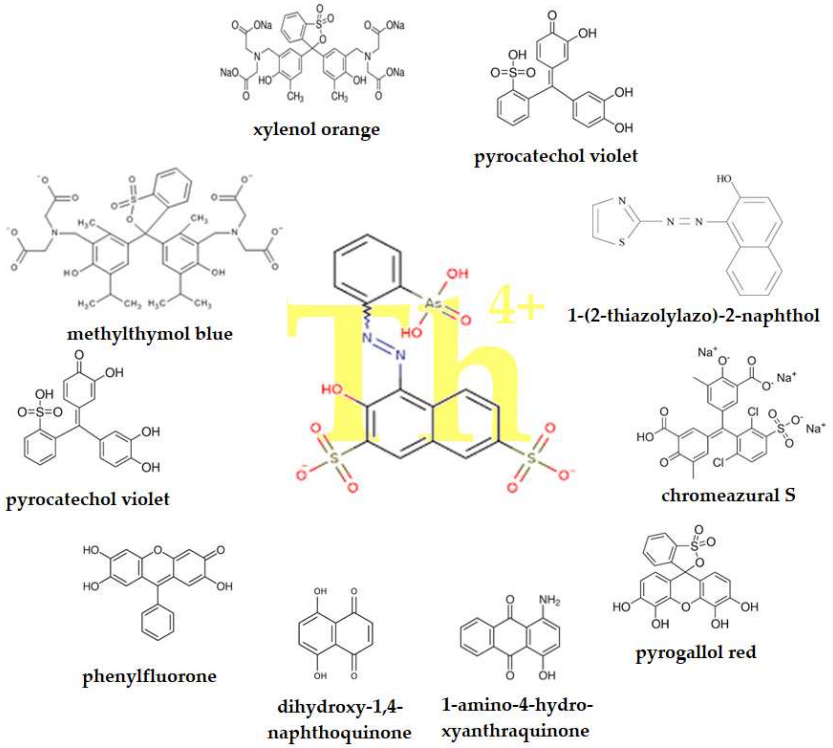

Figure 4. Common organic reagents involved in thorium ion complexation and/or extraction.

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6. Thorium determination

Although a radioactive element, physical-chemical analysis based on specific reactions finds permanent use in various applications [86–89]. The main reagent, studied exhaustively and used with excellent results in various working conditions is Thorin, 1-(2-Arsonophenylazo)-2-hydroxy-3,6-naphthalene-disulfonic acid sodium salt, 2-(2-Hydroxy-3,6-disulfo-1-naphthylazo)-benzene-arsenic acid sodium salt; Empirical Formula (Hill Notation): $C_{16}H_{11}AsN_2Na_2O_{10}S_2$ [76].

Otherwise, the radiation analyzes for thorium are few, although they refer to the entire radiation register (α, β or γ) [90–94] (Table 2).

That is why, alongside the highly developed spectrophotometric methods [95], the studied reagents are today widely used for preconcentration [96], with a view to the permanent development of new methods: electrochemical, optodes, electrochemical sensors, coupled spectral methods [97–121].

Table 2. Analysis methods for thorium and applications.

Analytical Methods	Samples and/or applications	Refs.
Inductively Coupled Plasma Mass Spectrometry	Microwave Digestion Technique for the Analysis of Rare Earth Elements, Thorium and Uranium in Geochemical Certified Reference Materials and Soils	[97]
Quadrupole-ICP-MS	Determination of trace element concentrations and stable lead, uranium and thorium isotope ratios by in NORM and NORM-polluted sample leachates	[98]
Inductively coupled plasma–atomic emission spectrometric	Chemical separation and determination of seventeen trace metals in thorium oxide matrix using a novel extractant – Cyanex-923	[99]
ICP-AES with MSF	Determination of Th and U	[100]
Inductively coupled plasma-atomic emission spectrometry	Determination of trace thorium in uranium dioxide	[101]
ICP-AES after matrix solvent extraction	Determination of REE, U, Th, Ba, and Zr in simulated hydrogeological leachates	[102]
Inductively coupled plasma mass spectrometry and on-line-coupled size-exclusion chromatography.	Determination of thorium and light rare-earth elements in soil water and its high molecular mass organic fractions	[103]
ICP-OES	Determination of Trace Thorium and Uranium Impurities in Scandium with High Matrix	[104]
Electrothermal atomization atomic absorption spectrometry	Thorium, zirconium, and vanadium as chemical modifiers in the determination of arsenic	[105]
Cyclic voltametric	Uranyl ion in sulfuric acid solutions. Application to some nuclear materials characterization.	[106]
Chemically modified electrode	Determination of thorium by adsorptive type with a poly-complex system	[107]
Fluorogenic thorium sensors	Based on 2,6-pyridinedicarboxylic acid-substituted tetraphenylethenes with aggregation-induced emission characteristics	[108]
Selective optode	Design and evaluation of a thorium (IV)	[109]
Electrothermal vaporization — inductively coupled plasma-atomic emission spectrometry	Trace metal determination in uranium and thorium compounds without prior matrix separation	[110]
Micellar electrokinetic chromatographic	Analysis of thorium, uranium, copper, nickel, cobalt and iron in ore and fish samples	[111]
laser induced breakdown spectrometry	Determination of trace constituents in thoria	[112]
preconcentration and inductively coupled plasma-mass spectrometric (ICP-MS)	Determination of thorium(IV), titanium(IV), iron(III), lead(II) and chromium(III) on 2-nitroso-1-naphthol impregnated MCI GEL CHP20P resin	[113]
laser induced breakdown spectrometry	Determination of trace constituents in thoria	[114]
Electrochemical and spectro-electro-chemical	Studies of bis(diketonate) thorium(IV) and uranium(IV) porphyrins	[115]
Electrochemical detector based on a modified graphite electrode	With phthalocyanine for the elemental analysis of actinides	[116]
selective extraction and trace determination of thorium	Synthesis of UiO-66-OH zirconium metal-organic framework and its application in water samples by spectrophotometry	[117]

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Anodic polarization of thorium and electrochemical impedance spectroscopy	Study at tungsten, cadmium and thorium electrodes	[118]
High performance liquid chromatographic	Studies on lanthanides, uranium and thorium on amide modified reversed phase supports	[119]
Extraction of thorium on resin	Using a commercially available extraction chromatographic resin	[120]
ion exchange and extraction chromatography	Separation of actinium from proton-irradiated thorium metal	[121]

7. Thorium separation and/or pre-concentration [122–140]

In analytical or technological interest, the concentration of thorium has permanently constituted a special problem especially because it accompanies the rare earths (REE), but especially uranium [122,123].

The concentration process that was often followed and then technologically capitalized was the pre-concentration of thorium [124,125].

Table 3 presents results that can form the basis of the development of urban thorium mining and which are focused on the concentration, pre-concentration, separation, extraction, ion exchange, the sorption or bio-sorption of thorium or thorium-uranium couple from various samples or aqueous solutions [126–149].

Table 3. Concentration and separation of thorium through various techniques.

Application	Materials or techniques	Refs.
Thorium Removal	Different Adsorbents	[126]
Removal of Thorium (IV) from Aqueous Solutions.	Modification of Clinoptilolite as a Robust Adsorbent for Highly-Efficient	[127]
Preconcentration of Uranium in Natural Water Samples	New Polymer with Imprinted Ions	[128]
Adsorption of Trace Thorium (IV) from Aqueous Solution	Mono-Modified β-Cyclodextrin Polyrotaxane Using Response Surface Methodology (RSM)	[129]
Novel Malonamide Grafted Polystyrene-Divinyl Benzene Resin for Extraction	Pre-Concentration and Separation of Actinides	[130]
Using Mesoporous	Selectivity of Th (IV) Adsorption as Compared to U (VI), La (III), Ce (III), Sm (III) and Gd (III)	[131]
α-Aminophosphonates, -Phosphinates, and -Phosphine Oxides	Extraction and Precipitation Agents for Rare Earth Metals, Thorium, and Uranium	[132]
Removal of Polyvalent Metal Ions (Eu(III) and Th(IV)) from Aqueous Solutions by.	Polyurea-Crosslinked Alginate Aerogels	[133]
Patented Chinese method	Method for Separating Cerium-Fluoride and Thorium	[134]
α-Aminophosphonate Extractant.	Extraction and Recovery of Cerium(IV) and Thorium(IV) from Sulphate Medium	[135]
Selective Extraction and Separation of Ce(IV) and Th(IV) from RE(III)	Sulfate Medium Using Di(2-Ethylhexyl)-N-Heptylaminomethylphosphonate	[136]
Selective Extraction and Separation of Ce(IV) from Thorium and Trivalent Rare Earths	Sulfate Medium by an α-Aminophosphonate Extractant	[137]
Extraction and Separation of Heavy Rare Earths from Chloride Medium	α-Aminophosphonic Acid HEHAPP.	[138]
Solvent Extraction and Separation of Rare Earths from Chloride Media Using	α-Aminophosphonic Acid Extractant HEHAMP.	[139]
on PAN/Zeolite Composite Adsorbent.	Study of the Behavior of Thorium Adsorption	[140]
Tulul Al-Shabba Zeolitic Tuff, Jordan	Adsorption of Thorium (IV) and Uranium (VI)	[141]
Sodium Clinoptilolite	Removal of Thorium from Aqueous Solutions	[142]
Adsorptions Performance towards Thorium.	Studies of Modification of Zeolite by Tandem Acid-Base Treatments	[143]
Tetraazonium based ionic liquid	Selective cloud point extraction of thorium (IV)	[144]
Deoiled karanja seed cake	Removal of thorium (IV) from aqueous solutions. Optimization using Taguchi method,	[145]

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	equilibrium, kinetic and thermodynamic studies	
Peat moss	Retention of uranyl and thorium ions from radioactive solution	[146]
To obtain photo-responsive metal-organic frameworks (MOFs)	Photocatalysis and adsorption	[147]
Th(IV) and Ce(III) in ThF_4-CeF_3-LiCl-KCl quaternary melt	Electrochemical behaviors and electrolytic separation	[148]
Hybrid mesoporous adsorbent as benzenesulfonamide-derivative@ZrO2	Selective removal of thorium ions from aqueous solutions	[149]
Extraction Using Sodium Diethyldithiocarbamate/Polyvinyl Chloride	Rare Earth Group Separation from Lamprophyre Dykes Leachate.	[150]
Metal-Organic Framework Based Fluorescent Sensors	for Hazardous Materials Detection.	[151]
Zeolite Adsorption.	Separation of Radionuclides from a Rare Earth-Containing Solution by	[152]
Acidic (Chelating) and Organophosphorus Ligands.	Equilibrium Constants of Mixed Complexes of Rare Earth Elements	[153]
Thenoyltrifluoroacetone:	Molecule for Solvent Extraction of Metals	[154]
8-Hydroxyquinoline Immobilized Bentonite.	Removal of Uranium and Thorium from Their Aqueous Solutions	[155]
New Polymer with Imprinted Ions Samples and Determination by Digital Imaging.	Preconcentration of Uranium in Natural Water	[156]

The remarkable results in development of organic ligands (Table 2), but especially of selective materials (Table 3) allow a confident approach to the recovery and recycling of thorium from electrical and electronic waste, but more generally (considering the slightly selective separation of waste) of residues that reach the integrated municipal storage and waste platforms (especially from construction).

8. Membrane and membrane processes

Membranes and processes have evolved from laboratory scale installations to industrial ones, having at the same time an increased economic and commercial importance [157]. Membrane processes have not only replaced some of the conventional separation processes, but also gave remarkable results in areas where conventional techniques were exhausted or very expensive [158,159]. Among the problems that have determined the exponential development of membrane processes are also those of environmental protection, since technologies based on membranes and membrane separation techniques are recognized as ecological technologies [160].

8.1. Introduction in membrane and membrane processes

If we focus on membrane processes, it can be stated that the membrane is a window of a multi-component system (Figure 5), with selective permeability for chemical species of the system [161]. This membrane allows the separation of the complex system formed by a solvent in which ionic chemical species, molecules and macromolecules, dispersed macromolecules and/or molecular aggregates and particles are dissolved, can be separated in components by classical or membrane processes [162]. In order for the separation process to occur, the system must be subjected to an electrochemical potential difference or driving force ($\Delta\mu$) [163].

Most important driving forces on membrane processes are [164–167]:

- P = transmembrane pressure difference;
- Δc = concentration difference between the two compartments separated by a membrane;
- ΔE = potential difference.

It should be emphasized that in the last decade membrane processes under potential gradient, thermal, magnetic, interfacial tension, volatility have also developed a lot [168–171].

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In this subchapter, we will briefly present the essential aspects regarding the processes with pressure or concentration gradient (liquid membranes).

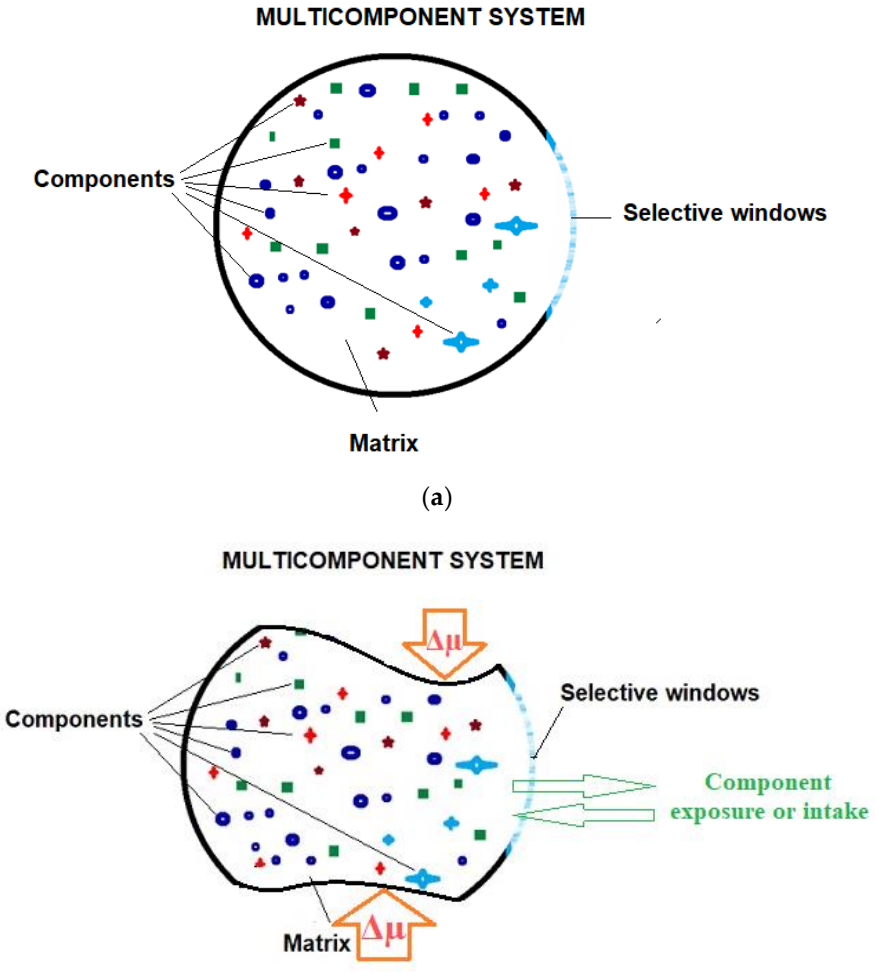

Figure 5. Multicomponent system bordered by a selective window: ions, small molecules, macromolecules, nanoparticles, micro-particles, microorganisms, and viruses, suspended particles: (**a**) system in equilibrium; (**b**) system subject to an electrochemical potential difference (Δμ).

8.2. Barro membrane processes

In the practice of membrane processes, the pressure difference (Δp) was constituted as a technically and economically accessible driving force, leading to many applications which are representative in a series of the processes such as: microfiltration, ultrafiltration, nano-filtration, reverse osmosis (hyperfiltration) [172–175]. The first and most developed application was the obtaining of drinking water from sea water (Figure 6a), when it was found that by applying a pressure higher than the sea water, mostly the solvent passes (96–99%) through a semi-permeable membrane [176,177]. But these processes have applications on industrial scale, and their introduction in a certain technology is a flow optimization problem (Figures 6b and 6c) [178–180], which depends on the load in chemical species to be removed from the solvent that constitutes the feed [181]. There is the option of operating in a dead-end filtration of cross-flow filtration system [182]. The design of filtration devices may differ, chemical equipment manufacturers competing to create prototypes with increasingly high performances by improving the flow on the membrane (Figures 6d and 6e) [183,184]. Because in filtration processes, no matter how hard you try, the membrane gets dirty, clogs or concentration polarization (solute accumulation) occurs on the layer adjacent to the membrane,

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process engineering is complemented by the introduction of ultrasonic cleaning devices into the technology, cavitation, magnetic stirring, or pulsatile flow vibrations [185].

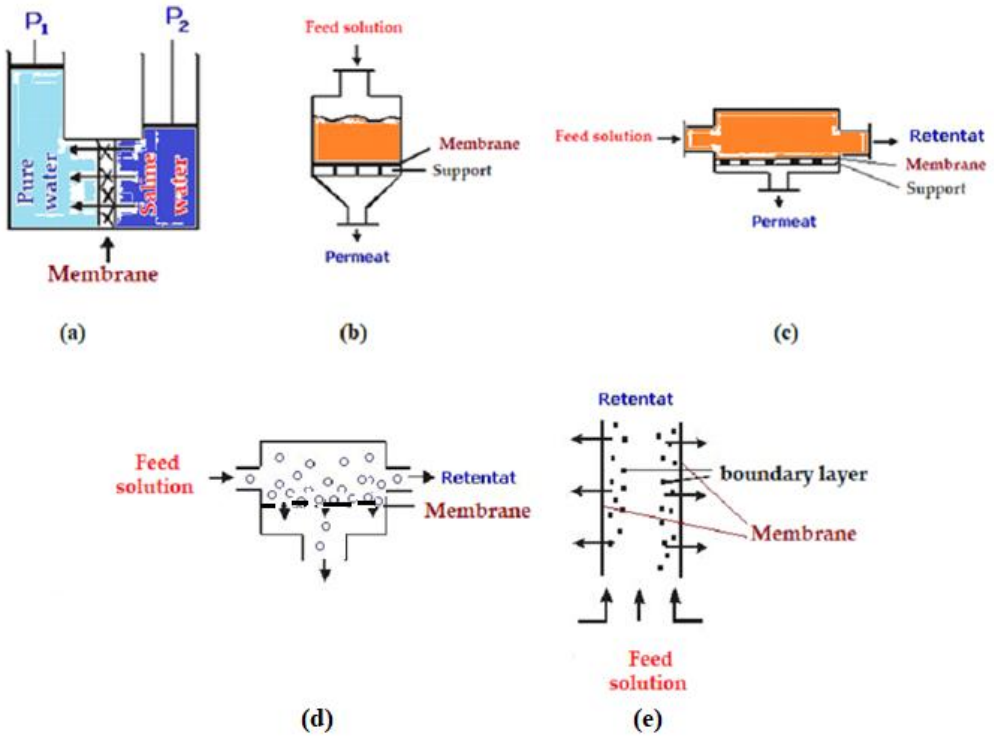

Figure 6. Aspects presented schematically on membrane separation processes under pressure difference: (**a**) obtaining drinking water through reverse osmosis; (**b**) piston type (dead end filtration); (**c**) tangential flow; (**d**) tangential flow through large sections; and (**e**) flow through tubes.

However, in essence, the feeding can be done through large cylindrical, tubular, spiraled or capillary (hollow fiber) spaces, in which, along with the flow through and/or on the membrane and avoiding fouling (contamination, soiling), the aim is to increase the area of the contact surface of the membrane with the dispersed system of feeding (Figure 7) [186]. Of course, the operation can be done by introducing in the feeding solution, as in Figure 7, but most often the feed solution being dirtier, it is inserted between tubes or fibers for a possible physical cleaning [187,188].

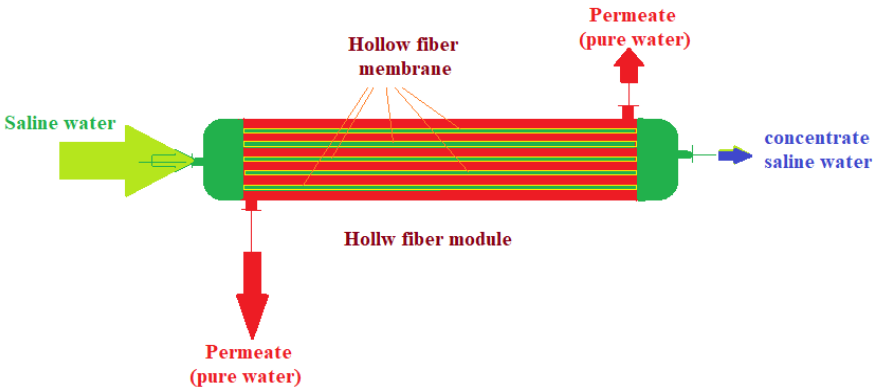

Figure 7. Advanced hollow fiber filtration modules.

A homogenous system can be separated by aggregation (segregation), so that instead of a high-pressure process (Table 4) a lower pressure one is used [189].

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Table 4. Characteristics of pressure gradient processes.

Type of membrane process	Pore diameter (nm)	Pressure (bar)	The obtained water content
Reverse osmosis	< 0.6	15–60	Pure water (poorly ionized)
Nanofiltration	0.6–10	6–20	Pure water (traces of molecular substances)
Ultrafiltration	7–70	4–15	Pure water, molecular substances and macromolecules
Microfiltration	50–800	0.5–2.5	Pure water, molecular substances and colloids

The first processes of this kind were promoted by Schamehorn, ultrafiltration of micellar systems (MUF) – which consists in transforming a solution into an ultra-micro-dispersed system by adding suitable surfactants, followed by ultrafiltration [190–192].

The condition for using micellar ultrafiltration is that the micelles contain the organic compound, which means an impurity of the concentrate [193].

The variants of ultrafiltration and nanofiltration have a huge development due to nano-species and nanomaterials (nanoparticles, nanotubes, nanofibers, proteins, soluble polymers, polyelectrolytes, micelles, vesicles) also used as carriers (Figure 8) in processes in liquid membranes [194].

The concentration polarization and the diffusion effects related to the sizes of solutes with low molecular masses, can influence the working conditions of nano- and ultra- filtration, the number of additives required being determined experimentally [195].

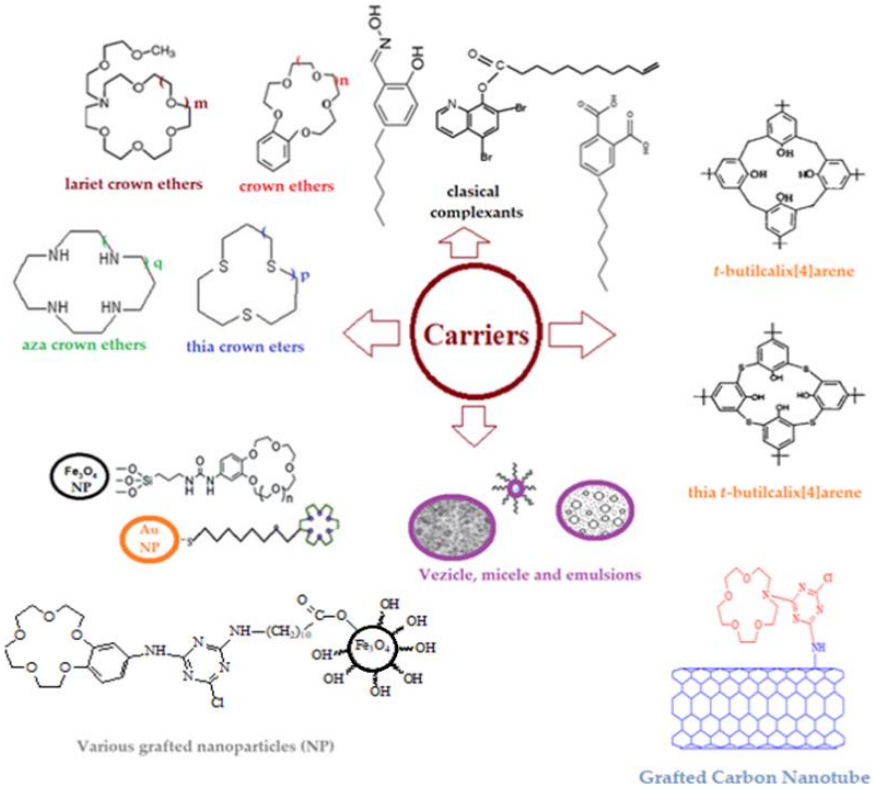

Figure 8. Common types of carriers: macrocyclic compounds, modified classical complexants agents, nano-species.

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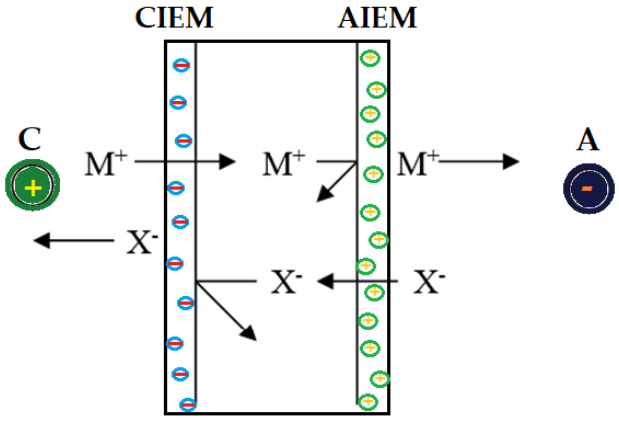
8.3. Electro-membrane processes

Electrodialysis is the most widespread separation process, carried out under an electric potential gradient, which involves ion exchange membranes [196]. In electrodialysis, the extraction, reconcentration and substitution operations are carried out without direct intervention of the electrodes [197]. They are placed at the end of the electrodialysis cells in order to maintain the electric potential difference between the compartments separated by membranes (Figure 9) [197–199].



M = cation; X = anion; C = Cathode; A = Anode
AIEM = Anion Ion-Exchange Membrane; CIEM = Cation Ion-Exchange Membrane

Figure 9. Scheme of an electrolysis cell for the concentration of a salt by electrodialysis with two ion exchange membranes.

If we associate an anion exchange membrane with a cathode, it is possible to eliminate an electrolyte whose cation can be deposited by electrochemical reaction on the cathode [200]. The electrolyte extracted from the diluted circuit by electrodialysis will be recovered in the concentrated circuit according to the principle in Figure 9. This electrolyte will not only be recovered, but can be reconcentrated. Recovery and reconcentration are possible because the ions cannot migrate over their compartment, the M cation being retained by the anion exchange membrane and X anion by the cation exchange membrane [201,202].

Conducting electrodialysis requires ways to interpose electrodes, aqueous phases to be processed and the membranes, so that the operation can lead at the same time to solute concentrations or to the recovery of deionized water [203–205].

8.4. Membrane processes carried out under a concentration gradient (liquid membrane)

Although the concentration gradient is also found in processes with solid membranes (osmosis, dialysis, forward osmosis), this paper deals with the processes with liquid membranes that have high chances of developing applications in the valorization of thorium [206].

Separation systems with liquid membrane (LM), or bulk liquid membrane (BLM) are formed by two homogenous liquid phases, immiscible with the membrane, called the source phase (SP) and the receiving phase (RP). The separation of the two liquid membranes is achieved with a third liquid, the membrane (M), that acts as a semi-permeable barrier between the two liquid phases [207].

An established graphic but also practical conception of liquid membranes (Figure 10), takes into account the density of the membrane, which is generally an organic solvent or a multicomponent system in which the continuous phase is the organic solvent [208].

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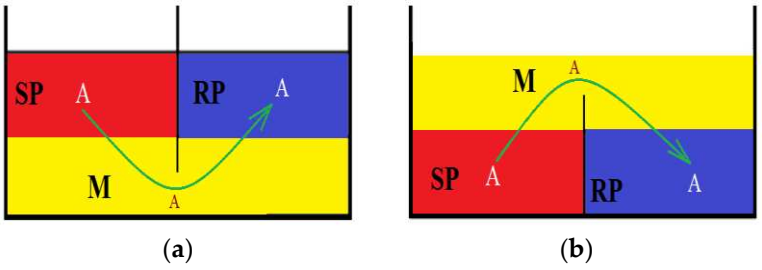

Figure 10. Schematic presentation of membrane systems with organic solvent: denser (**a**); or less dense than aqueous phases (**b**). Legend: M = membranes; SP = Source Phase; RP = Receiving Phase; A = chemical species of interest for separation.

The density of the membrane phase becomes unimportant if the membrane solvent is immobilized in or on a support [209], thus obtaining supported liquid membranes (SLMs). An interesting variant, but not yet sufficiently evaluated in separation processes, is the liquid membrane based on magnetic liquid (ferrofluid) [210], which also has no restrictions on the density of the organic solvent, but involves special aspects in stability and transfer of table [211].

If we focus on BLM, the technical problems to be solved are: the large volume of solvent used (V), the small mass transfer area (σ), the almost unit ratio between the volume of the source phase, the volume of the receiving phase (r) and the volume of the membrane organic solvent (OS or M) and therefore, implicitly, the long operating time (t) [212].

In order to improve the performances, hollow-fiber supported membranes (HFLM) and emulsion membranes (ELM) have been greatly developed (Figure 11) [213].

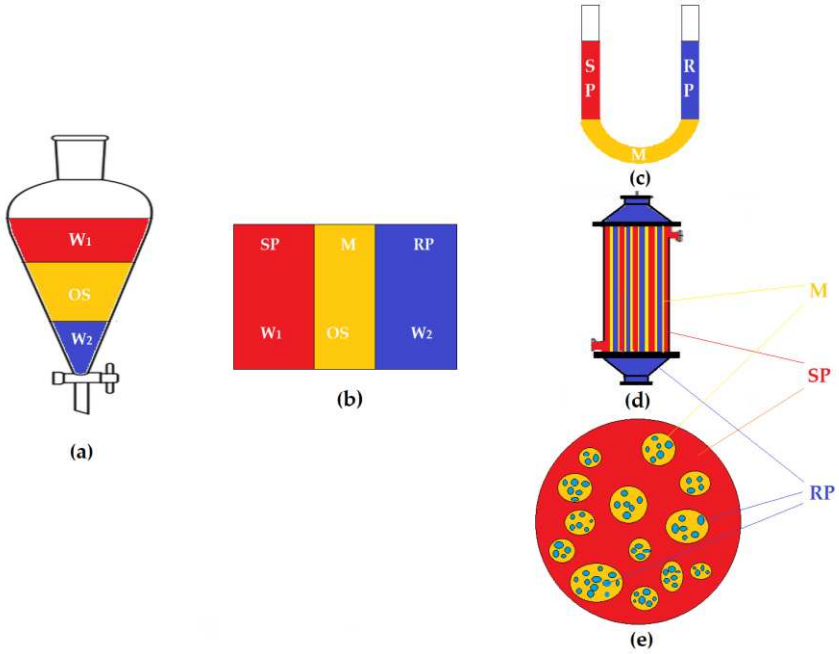

Figure 11. Schematic presentation of extraction and membrane systems with organic solvent: **(a)** water 1 (W1)–organic solvent (OS)–water extraction (W2); or: **(b)** liquid membranes (LM); **(c)** bulk liquid membranes (BLM); **(d)** supported liquid membranes (SLM); **(e)** emulsion liquid membranes (ELM). Legend: M = Membrane; SP = Source Phase; RP = Receiving Phase.

Recently, a BLM system with dispersed phases has been studied, in which the aqueous phases of the separation system disperse in / through the membranes. The membrane is a nanodispersed system of magnetic nanoparticles that have the role of ensuring both convection and transport for ionic chemical species in membranes based on saturated alcohols C_6–C_{12} [214,215]. The most recent design is shown in Figure 12, but other variants using chemical nanospecies are also known [216].

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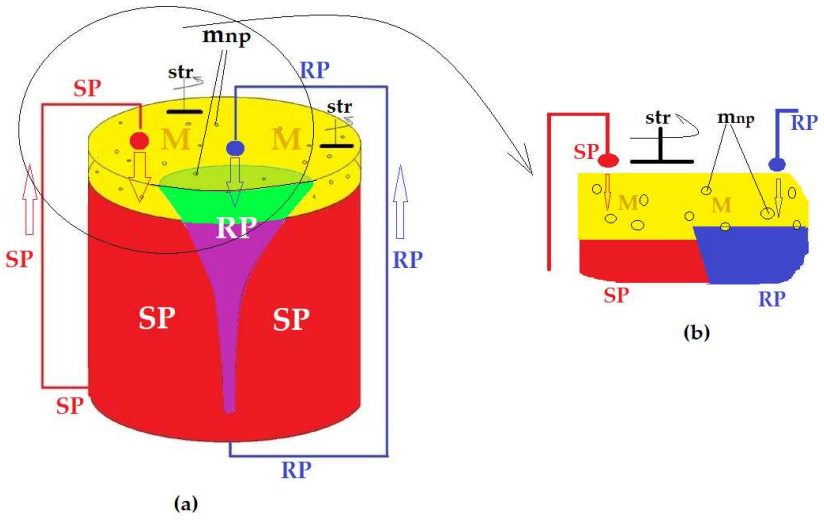

Figure 12. Schematic presentation of the permeation module with dispersed phases: (**a**) view; (**b**) cross-section detail;
Legend: SP—source phase; RP—receiving phase; M—organic solvent membrane; m_{np}—magnetic nanoparticles; str—stirrer with magnetic rods.

The BLM system with dispersed phases (based on Figure 8 carriers, for example) is close to the performances of liquid membranes on hollow fiber support or emulsion-type liquid membranes but has several limitations that restrict its applicability: stability of the membrane nanodispersion, control of the size of droplets in recirculating aqueous phases, losses of membrane material (solvent or nanoparticles) [217].

8.5. Transport in liquid membranes

The way of achieving the concentration gradient and the nature of the species dissolved in the phases of the membrane system led to various types of transport through liquid membranes [218–220]. Mainly, however, they can be narrowed down to those in Figure 13.

8.5.1. Physical "simply" shipping

The simple diffusion type of transport through the solution is usually followed by the permeation of the solute through the liquid membrane due to the concentration gradient (Figure 13a). In this case, the transport of the component from the source phase through the membrane phase occurs with a higher solubility or diffusivity of the solute in the membrane phase. In this type of transport, the mass transfer rate is low and depends on the solubility of the solute in the organic phase, as well as the solubility of the solute in the source and receiver phases [221,222].

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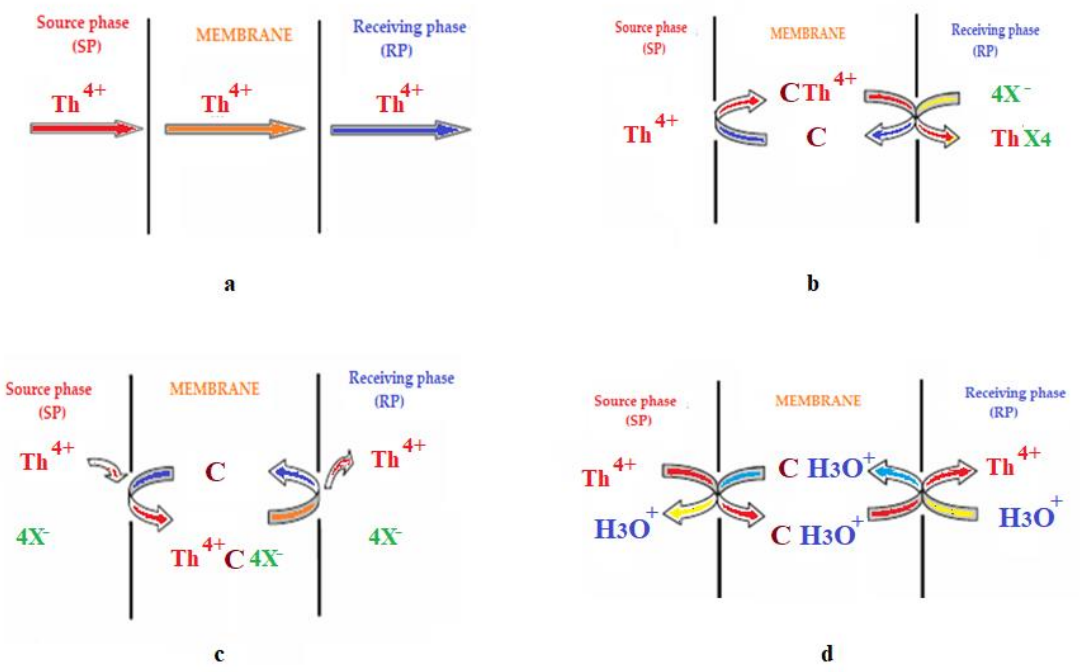

Figure 13. Schematic presentation of the transport mechanism by liquid membranes (C–carrier, X–anion complexant).

8.5.2. Facilitated transport or carrier-mediated transport

In carrier-mediated transport, a carrier is added to the membrane phase in order to increase the mass transfer rate or separation efficiency of the liquid membrane. Therefore, it is called facilitated transport or transport mediated by a transporter [223]. In this case, the solute dissolved in the source phase, at the source phase–LM interface, reacts chemically with a transporter dissolved in the liquid membrane to form a complex. This complex reacts inversely at the LM–receiving phase interface, releasing the partitioned solute in the receiving phase (Figure 13b). In recent years, this type of transport mediated by a transporter has been intensively developed for the selective transport of cations, anions as well as neutral species through liquid membranes [224].

8.5.3. Coupled co- or counter-transport

In this type of transport, the transport speed of a certain ion is dependent on the concentration of another ion. In the case of coupled co-transport, the metal ion is transferred together with a counter-anion, the two species transport taking place in the same direction. In the coupled counter-transport type, the simultaneous transport of another ion from the receptor phase to the source phase takes place, thus the transport of the two species takes place in opposite directions [225–227]. Figures 13c and 13d show the types of co- and counter-coupled transport of a metal ion.

8.6. Hybrid membrane processes

The most common form of treatment of effluents containing heavy metal ions involves the precipitation of metals as hydroxide, base salt or as a sulphur. Precipitation is often followed by an additional treatment such as sedimentation or filtration processes [228–230].

The technique of liquid membranes also presents a huge potential for the application of the removal and valorization of heavy metals, especially for the purpose of environmental protection [231–233].

Currently, the most important commercial application of liquid membrane technologies is the treatment of wastewater and waste [232,234–236].

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However, the use of liquid membranes has encountered many obstacles, mainly related to the use of solvents with high toxicity [237,238], so that both the reduction of the amount of the membrane solvent required and their replacement with green solvents or nanodispersions have created better opportunities for this process [239,240].

The idea of using nanosystems has led to the development of hybrid processes, which basically follow the mechanism of liquid membranes, but the process design is more advanced [241,242].

9. Problems in application, achievements and development perspectives of an urban thorium mining

The analysis of the processes in which the minerals or waste containing thorium are processed shows that the classic technologies have material losses in the environment, which could be reduced with or through membrane techniques. Thus, in the classical thorium recovery technologies, some disadvantages of the operations were highlighted (Table 5), which require improvements especially from the perspective of the loss of thorium in the environment:

Table 5. Possible losses of thorium in the environment and remedial possibilities.

Technological operation	Losses of thorium or of thorium-contaminated materials	Means of remedial or reduction of losses
Crushing, grinding	Dust removal Mill shutdown losses Losses when cleaning the machine	Microfilter installation Micro and ultrafiltration of colloidal washing solutions
Solubilization or leaching	Incomplete solubilization with the chosen reagent Complete solubilization Too little concentration of thorium	Solubilization with a complementary reagent Selective reprecipitation and solubilization Concentration by precipitation and microfiltration
Filtration	Thorium retention in the precipitate Reduced concentration of thorium in the filtrate	Washing with solubilizing reagents Reprecipitation and micro or ultra filtration
Precipitation	Incomplete precipitation Precipitation of nanometric particles	Nanofiltration or reverse osmosis of the filtrate Colloidal ultrafiltration or nanofiltration
Extraction	Solvent losses Incomplete extraction	Solvent recovery Use of selective extractants
Ion exchange	Blockage of thorium in the ion exchanger (elution inefficiency) Incomplete retention	Change eluent Recovery of ion exchangers for destruction (burning)

Solving the problem of thorium separation, concentration and recycling can be approached by analyzing some of the contributions that offer both priority research directions and some viable technical solutions (Table 6) [243–266].

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Table 6. Aspects regarding the use of membranes and membrane materials with possible implications regarding thorium separation.

Topics	Application	Refs.
Ionic Liquid	Gas Separation Membranes	[243]
Waste treatment	Liquid Radioactive Wastes Treatment	[244]
Ionic Liquids	Proton Exchange Membrane in Fuel Cells	[245]
Roles of Chitosan-Supported Ionic Liquids	Chitosan-Based Polymers as Proton Exchange	[246]
Strategy in Liquid Filtration	Membrane Surface Patterning as a Fouling Mitigation	[247]
Polymer Inclusion Membrane and a Chelating Resin	Sequential Determination of Copper(II) and Zinc(II) in Natural Waters and Soil Leachates	[248]
Polymers and Solvents Used in Membrane Fabrication	Sustainable Membrane Development.	[249]
Light Responsive Polymer Membranes	Miscellaneous application	[250]
Poly(vinylidene-fluoride-co-hexafluoropropylene) Polymer Inclusion Membrane Containing Aliquat® 336 and Dibutyl Phthalate	Extraction from Sulfate Solutions	[251]
Ionic Liquid	Based Electrolytes for Energy Storage Devices	[252]
Ionic Liquids	Their Toxicity to Living Organisms.	[253]
Modern Computer Application	Model Rare Earth Element Ion Behavior in Adsorptive Membranes and Materials	[254]
Bulk Hybrid Liquid Membranes Based on Dispersion Systems	Operational Limits	[255]
nanofiltration membrane	effect of the adsorption of multicharge cations on the selectivity of a	[256]
Nanofiltration	Extraction of Uranium and Thorium from Aqueous Solutions	[257]
Polymer Inclusion Membranes (PIMs) Doped with Alkylimidazole	Application in the Separation of Non-Ferrous Metal Ions.	[258]
nanofiltration process	Removal of fluoride by nature diatomite from high-fluorine water	[259]
reverse osmosis and nanofiltration	removal radioactive contamination of groundwater, special aspects and advantage	[260]
Selective concentration	uranium from seawater by nanofiltration	[261]
glutathione-based magnetic nanocomposite	Sequestration and recovery of thorium ions using a recyclable, low-cost	[262]
evaluation of sodium alginate/polyvinyl alcohol/polyethylene oxide/ZSM5 zeolite hybrid adsorbent	a case study of thorium (IV).	[263]
Use of response surface methodology for optimization of thorium (IV)	removal from aqueous solutions by electro-deionization (EDI)	[264]
Continuous bulk liquid membrane technique	thorium transport: modeling and experimental validation	[265]
Synthesis, characterization, and evaluation of thiocarbazide-functionalized maleic-based polymer	thorium (IV) removal from aqueous solutions	[266]

Recent studies on the separation, concentration, removal or recovery of thorium from aqueous solutions, including by membrane techniques [267–289] (Table 7), have led to promising results, reinforcing the idea that membrane or hybrid processes can contribute to the imaging of flows

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technological recycling of thorium from various residues, especially industrial, on municipal waste processing platforms.

The various compounds proposed recently [290–300], but also some previously used [301–305] can contribute to the construction of a scheme for recuperative separation of thorium on an integrated municipal platform for processing, mainly, the waste of electrical devices (lamps, tubes, mantles) and electronics, but also those from the construction industry (welding electrodes, metallic materials and alloys).

Table 7. Recent materials and processes for thorium recovery.

Materials or processes	Application	Refs.
Solvent extraction and separation of thorium (IV)	from chloride media by a Schiff base.	[267]
Leaching and precipitation of thorium ions	from Cataclastic rocks (Abu Rusheid Area, South Eastern Desert, Egypt)	[268]
Equilibrium ultrafiltration of hydrolyzed thorium (IV) solutions.	Solubility of thorium salts	[269]
Evaluation of inorganic ion exchange materials.	for purification of 225Ac from thorium and radium radioisotopes	[270]
graphene oxide nanoribbons/manganese dioxide composite material.	Thorium adsorption on	[271]
oxidized biochar fibers derived from Luffa cylindrica sponges	Thorium adsorption	[272]
onto activated bentonite.	Sorption behavior of thorium(IV)	[273]
amorphous silica	Adsorption of thorium(IV) response surface modelling and optimization.	[274]
titanium tetrachloride modified sodium bentonite.	Th(IV) adsorption	[275]
electrospun PVA/SA/PEO/HZSM5 nanofiber.	Evaluation of single and simultaneous thorium and uranium sorption from water systems	[276]
crystalline tin oxide nanoparticles.	Kinetics, isotherm and thermodynamics for uranium and thorium ions adsorption from aqueous solutions by	[277]
novel electrospun polyvinyl alcohol/titanium oxide nanofiber adsorbent modified with mercapto groups.	for uranium(VI) and thorium(IV) removal from aqueous solution	[278]
Biosorption	uranium and thorium	[279]
Synthesis and characterization of poly(TRIM/VPA) functionalized graphene oxide nanoribbons aerogel,	for highly efficient capture of thorium(IV) from aqueous solutions	[280]
vinyl-functionalized silica aerogel-like monoliths,	for selective separation of radioactive thorium from monazite	[281]
recyclable GO@chitosan based magnetic nanocomposite	Selective removal of uranium from an aqueous solution of mixed radionuclides of uranium, cesium, and strontium via a viable	[282]
graphene oxide (GO) and (aminomethyl) phosphonic acid–graphene oxide (AMPA–GO)	Study of kinetic, thermodynamic, and isotherm of Sr adsorption	[283]
bulk liquid membrane containing Alamine 336 as a carrier	Kinetic study of uranium transport	[284]

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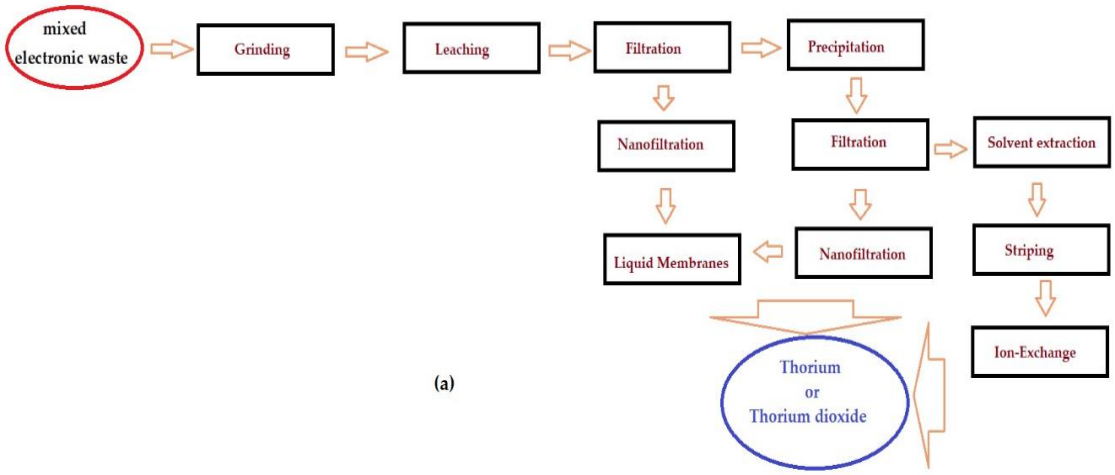
Continuous bulk liquid membrane technique	thorium transport: modeling and experimental validation	[285]
electrodeionization (EDI)	Use of response surface methodology for optimization of thorium (IV) removal from aqueous solutions	[286]
Magnetic chitosan composite particles	evaluation of thorium and uranyl ion adsorption from aqueous solutions	[287]
multi-walled carbon nanotubes decorated with magnetic nanoparticles	Sorption and preconcentration of uranium and thorium from aqueous solutions	[288]
Kinetic and isotherm analyses using response surface methodology (RSM)	for thorium (IV) adsorptive removal from aqueous solutions by modified magnetite nanoparticle	[289]

In the diagrams in Figure 14, several proposals for technical solutions for urban thorium mining are presented, starting from the raw material: waste that ends on integrated municipal waste management platforms.

Thus, in a first case (Figure 14a) it is assumed that the waste (it is assumed to be electrical and electronic waste or metal waste from metal construction materials) contains thorium and is totally unselected. This option would require the use of the classic scheme for the separation of thorium from poor sources, including the following operations: leaching, filtration (sedimentation), precipitation, filtration, solubilization at Th^{4+}, extraction, re-extraction, ion exchange. In this operating scheme, membrane processes that can be integrated to increase the performance of the process can be nanofiltration and/or liquid membranes.

A second case may be a raw material containing thorium alloyed with tungsten (filaments of incandescent lamps or other lighting fixtures, welding electrodes or building material alloys). Figure 14b shows the main operations that would consist of leaching, filtration, precipitation, filtration, nanofiltration, solubilization at Th^{4+}, extraction and stripping or membrane electrolysis and nanofiltration.

The third possible case would be a raw material consisting of various wastes from aluminum and magnesium alloys (Figure 14c). Such a waste content can be processed for thorium recovery by membrane electrolysis or acid attack, followed by filtration and nanofiltration.



(a)

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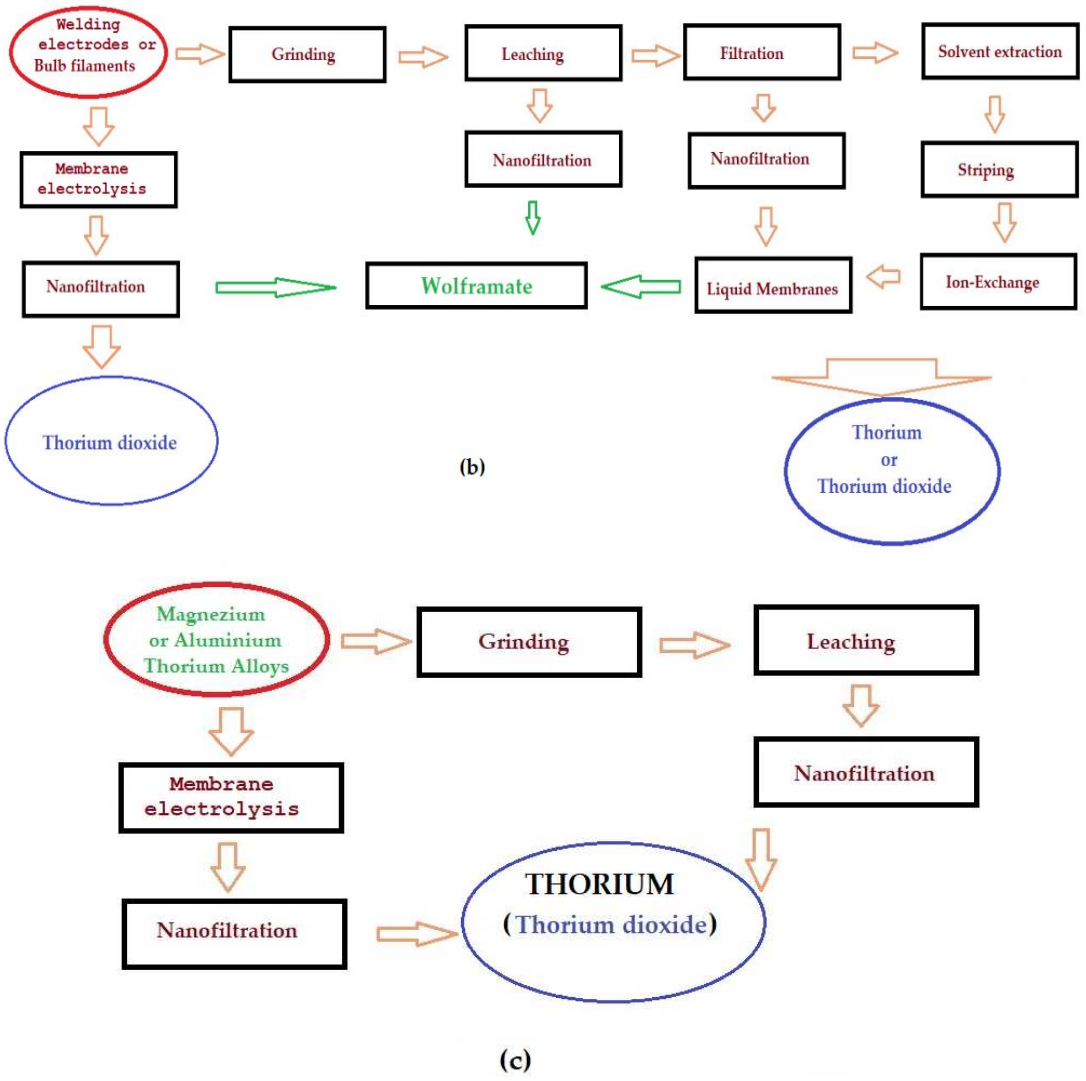

Figure 14. Scheme of proposals for the separation, recovery and recycling of thorium from waste of municipal waste management platforms.

The proposed operation schemes are highly dependent on the quality of waste selection that reaches the integrated municipal waste for management and processing platforms.

10. Conclusions

Although a radioactive element and a promising raw material for nuclear power generation, thorium, being a fairly abundant metal (similar to lead) has surprising domestic uses: toothpaste, dental cement, crucibles for high-temperature work, filaments for incandescent bulbs, welding electrodes, aluminum or magnesium alloys, jewelry, sculptures, coats and goggles, devices working at high temperatures, lamps for electronic devices.

Current regulations consider thorium to be a carcinogenic element, and its bio-toxicity and impact on human health (affects internal organs and blood), require the recovery and recycling of thorium, especially in the case of waste from municipal management platforms.

Classical thorium recovery processes require acid or base attack on the thorium-containing feedstock, filtration, re-solubilization, extraction, and ion exchange.

A variety of complexants and transporters have been used for the separation and preconcentration of thorium (especially for its analysis), which opens the perspectives of membrane applications (nanofiltration, colloidal ultrafiltration, liquid membranes, emulsion membranes) for thorium utilization.

Membrane processes can intervene throughout the thorium recovery and recycling stream, increasing the efficiency of the process and avoiding losses to the environment.

The proposed processing schemes for various wastes containing thorium are only a topic of reflection on the possibility of removal, recovery and valorization of thorium, practically opening an urban mining of this element.

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Abbreviation list

D	*dialysis.*
ED	*electrodialysis.*
EDI	*reverse electrodialysis.*
MF	*microfiltration.*
UF	*ultrafiltration.*
NF	*nanofiltration*
RO	*reverse osmosis.*
DM	*membrane distillation.*
SG	*gas separation.*
PV	*pervaporation.*
ELM	*liquid membrane extraction.*
TC	*facilitated transport.*
BLM (MLV)	*Bulk liquid membranes*
HLM	*hybrid liquid membranes*
HFCLM	*liquid membranes thin capillary or tubular fibers*
HFLM	*hollow fiber liquid membranes*
HMS	*hybrid multi-membrane systems*
P	*precipitation*
F	*filtration*
M	*milling*
E	*extraction*
S	*striping*
RE	*re-extraction*
N	*neutralization*
TBP	*tri-butyl phosphate*
REE	*rare earth elements*

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