MEMBRANE SEPARATION OF RADIOACTIVE SUBSTANCES

I.M. Buntseva, I.N. Beckman

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Summary

This paper describes current research which aims to develop the potential of membrane processes for a cleaner environment from radionuclides, radioactive waste management, and marine pollutants monitoring.

Abstract

Membrane separation of gases. The apparatus designated for the determination of selective transport parameters of radioactive gases in the polymer membranes with the use of the gas permeability method is described. Within the frame work of this study the gas permeability kinetics of radioactive gases, namely, tritium, radon, tritium water, ^{85}Kr, ^{133}Xe, $^{14}CH_4$, $^{14}C_6H_6$, etc., in polyethylene, polypropylene and polyviniltrimethylsilane (PVTMS) were measured. Diffusion coefficient, solubility constant and permeability coefficient of various gases in polymers as well as the productivity and permselectivity of membranes were calculated using the experimental results. The effect on these parameters of such factors as temperature, gas pressure, the gas mixture composition, membrane thickness, thermal, mechanical and radiation pre-history of the polymer has been studied. The prospects for the application of asymmetric membranes from PVTMS for the separation of heavy inert gases and for the removal of radioactive krypton, xenon and radon from their mixtures with air are demonstrated. The separation factor value for the gas pair Xe-Kr is 2.

Membrane gas absorption. Mobile liquid membranes can be used to separate gas mixtures which are difficult to separate by conventional methods even if the permeabilities of gases are practically the same.

Flat-sheet membrane permabsorber operating in the circulation mode has been tested by separation a two-component gas mixture Rn-Ar. Asymmetric non-porous membranes produced from PVTMS and aqueous glycerine solutions of different concentrations served as the carrier of radon. The separation factor value for the gas pair Rn-Ar is more than 10.

Permstraction. The recovery of heavy metals (^{232}Th, ^{238}U, etc) from aqueous solutions by means of membrane-based liquid-liquid extraction, using buthyl phosphate in benzol as extractant, has been studied. Microporous polypropylene films were used. Permstractor consists of two cells (extraction and re-extraction cells) and the liquid between them. The overall mass transfer coefficients were determined under different hydrodynamic conditions in both phases. It is possible to recuperate radioactive heavy metals from industrial effluents by means of membrane-based extraction using extractor-reextractor system operating in the circulation mode.

METHOD OF RADIONUCLIDES SORPTION DEFECTOSCOPIC IN DIAGNOSTIC OF THIN LAYER OF FLUORINATED SURFACE OF POLYETHYLENE

I.N.Beckman, I.M.Buntseva

(Chair Radiochemistry)

Resume

The method of radioactive gases sorption fault detection is used for the control of the barrier characteristics of a thin surface layer of polyethylene of low density modified by fluorination, sulfonation and oxidation. As radioactive gas probes applied vapours of bensol, cyclohexan or tetrachloro-methane marked by ^{14}C . The influence on the thickness of the modified layer, it defects and the diffusion in it vapours of organic substances of such factors, as a type of the modifying agent, mode of updating, cleanliness and degree of oxidation of a surface of a sample, irradiation of polymer by the large dozes of ionization radiation is investigated. Is shown, that from the point of blocking process of mass-transfer by most effective is a method fluorination in comparison with sulfidation. The homogeneous distribution of the ftuorinated layer on surface of polymer is achieved only after careful cleaning of a surface. The preliminary oxidation of a surface of polyethylene essentially worsens the barrier characteristics of layer near to a surface of a material. In process of growth of a doze of an irradiation the protective characteristics fluorinated of a layer fall as the loose layers practically which are not slowing down migration through them gases and vapours are formed.

MEMBRANE SEPARATION OF RADIOACTIVE SUBSTANCES

I.M.Buntseva and I.N.Beckman

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Effective methods for the separation and concentration of radioactive substances are required to provide the operation safety of nuclear and thermonuclear plants. This paper describes current research at the Moscow State University which aims to develop the potential of membrane processes for a cleaner environment from radionuclides, radioactive waste management, and marine pollutants monitoring.

Membrane separation of gases. The membrane gas-separation technique became important with the development of synthetic polymeric nonporous membranes, and for many years investigations have been made to develop convenient and reliable methods for separating radioactive gas mixtures by means of membranes. The apparatus designated for the determination of selective transport parameters of radioactive gases in the polymer membranes with the use of the gas permeability method consists of a diffusion cell and a number of systems for preparation and overlapping of gas mixtures of a given composition, radiometric recording, measuring and regulating the sample temperature, and radioactive gas absorption after the completion of the experiment. Spectrometry-type detectors are installed to detect various types of radiation and separate components of the gas mixture. In the process of the experiment, the apparatus also provides the automatic and continuous recording of the diffusion fluxes of gases inserted

at the input and output of the membrane. The installation is used to study the kinetics of the radioactive and stable gas permeability by employing various methods, such as, integral, differential, pulse and concentration waves methods. With the use of the gas permeability method with autoradiographic detection it is possible directly control the evolution of the diffusion process in the solid phase.

Within the framework of this study the gas permeability kinetics of radioactive gases, namely, tritium ($^{3}H_2$), radon (^{222}Rn), tritium water (HTO), 85Kr, ^{133}Xe, $^{14}CH_4$, $^{14}C_6H_6$, etc., in polyethylene, polypropylene and polyviniltrimethylsilane (PVTMS) were measured. Diffusion coefficient, solubility constant and permeability coefficient of various gases in polymers as well as the productivity and permselectivity of membranes were calculated using the experimental results. The effect on these parameters of such factors as temperature, gas pressure, the gas mixture composition, membrane thickness, thermal, mechanical and radiation pre-history of the polymer has been studied. The prospects for the application of asymmetric membranes from polyvinyltrimethylsilane for the separation of heavy inert gases and for the removed of radioactive krypton, xenon and radon from their mixtures with air are demonstrated.

Membrane gas absorption. Mobile liquid membranes can be used to separate gas mixtures which are difficalt to separate by conventional methods even if the permeabilities of gases are practically the same. The membrane permabsorber consist of two cells (absorption cell and desorption cell) and the liquid specifically selected as an agent for extraction that circulated between them. The gas mixture passes over the polymeric membrane in the absorption cell. The most permeable component of the gas mixture diffuses selectively through the nonporous polymeric membrane into the flowing liquid under it, is absorbed by this liquid, and is transferred to the desorption cell. Degassing of the liquid occurs in the desorption membrane cell through the nonporous polymeric membrane, leading to highly concentrated gaseous products.

The flat sheet membrane permabsorber operating in the circulation mode has been tested by separation a two-component gas mixture Rn-Ar. Asymmetric non-porous membranes produced from PVTMS and aqueous solutions glycerin of different concentrations served as the carrier of radon. The separator factor value for the gas pair Rn-Ar is more than 10.

Pertraction. The recovery of heavy metals (^{232}Th, ^{238}U, etc.) from aqueous solutions by means of membrane-based liquid-liquid extraction, using butyl phosphate in benzol as extractant, has been studied. Microporous polypropylene films were used. Permstractor consist of two cells (extraction cell and reextraction cell) and the liquid between them. The overall mass transfer coefficients were determined under different hydrodynamic conditions no both phases. It is possible to recuperation radioactive heavy metals from industrial effluents by means of membrane-based extraction using extractor-reextractor system operating in the circulation mode.

New construction of **combine integrated membrane systems (**membrane reactor + integrated system of gas separation) was proposed. Equipment integrates in one module different methods of processing and separation of gas mixtures: absorption, adsorption, extraction and liquid catalyse. For regeneration of liquid carrier can be put to use desorption and reexstraction processes. Equipment is dedicated to the cleaning of the liquid nuclear plant wastes from radionuclides. Its make possible remove from waste the organic volatile matter, two radioactive metals and two gasses.

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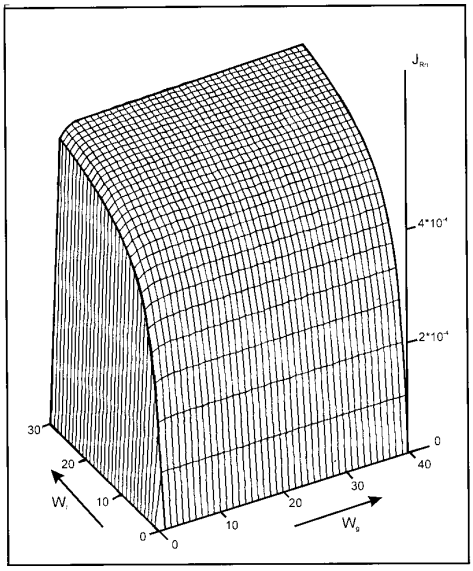
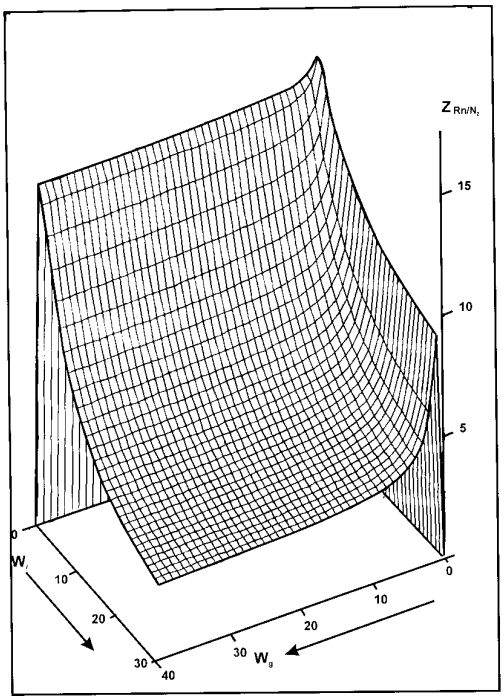


CALCULATED PRODUCTIVITY OF THE MODULE FOR SEPARETION OF ^{222}Rn FROM N_2.
W_g - gas flow rate, cm^3/s;
W_l - liquid flow rate, cm^3/s.
Retentate clearing, membrane absorption module of membrane contactor with flowing liquid carrier,non-porous membrane, polymer -PVTMS, liquid absorbent -water.



DEPENDENCIES OF CALCULATED SEPARATION FACTORS ($^{222}Rn/N_2$) UPON THE LIQUID AND GAS FLOW RATE.
W_g - gas flow rate, cm^3/s,
W_l - liquid flow rate, cm^3/s.
Retentate clearing, membrane absorption module of membrane contactor with flowinq liquid carrier, non-porous membrane, polymer - PVTMS, liquid absorbent - water.

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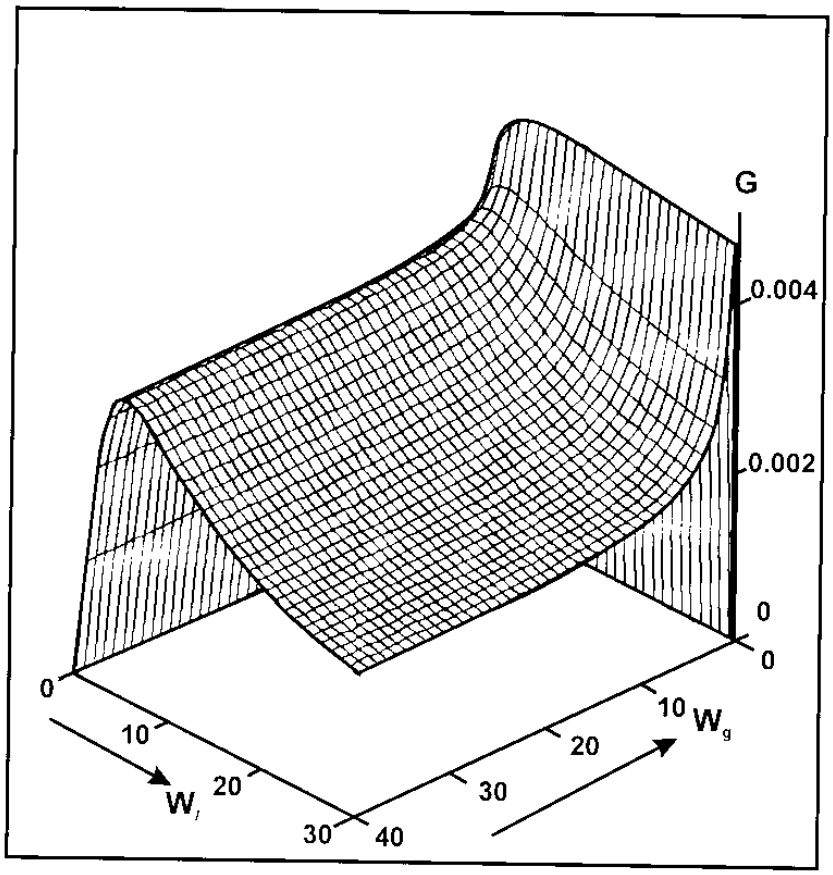
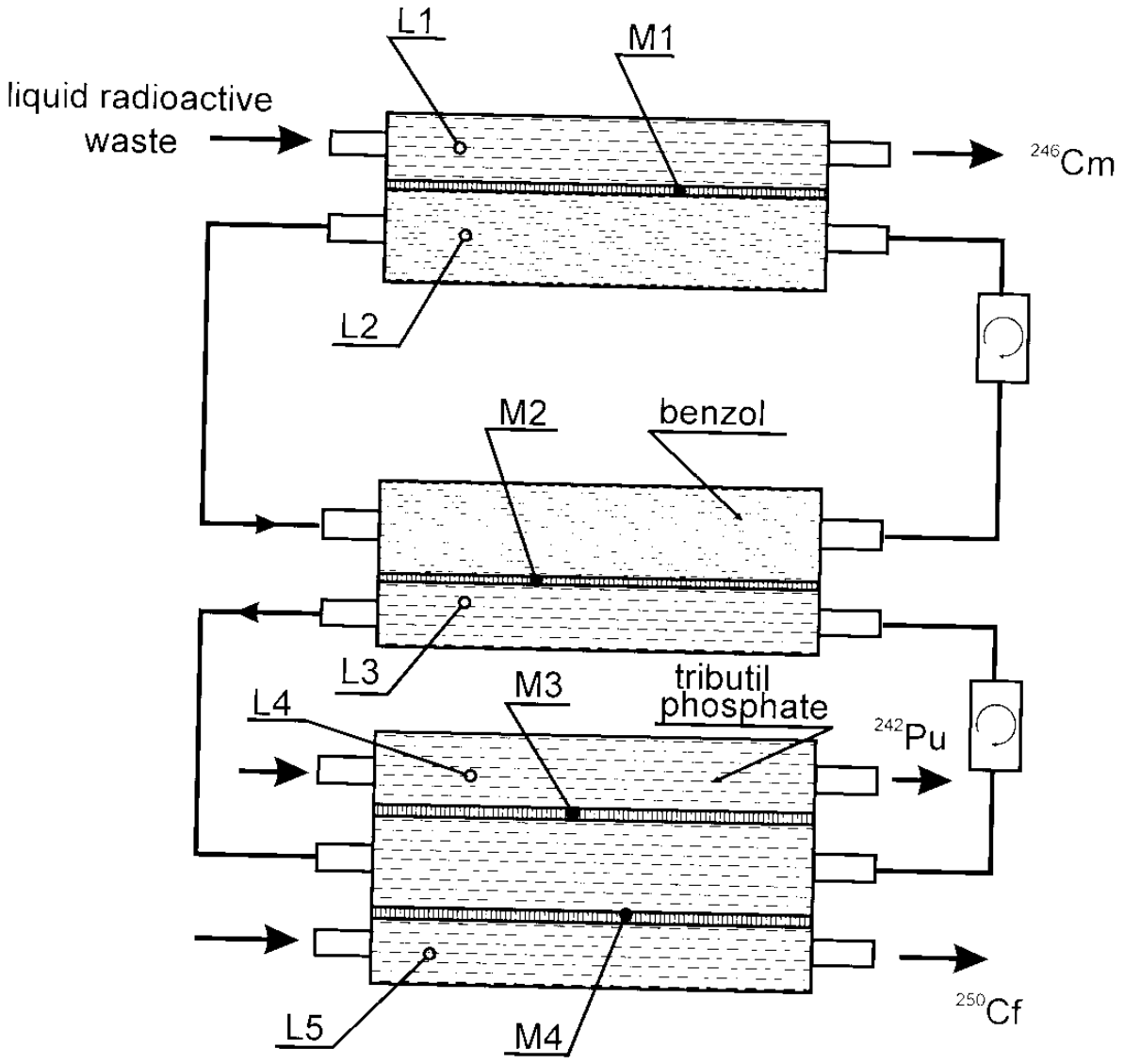
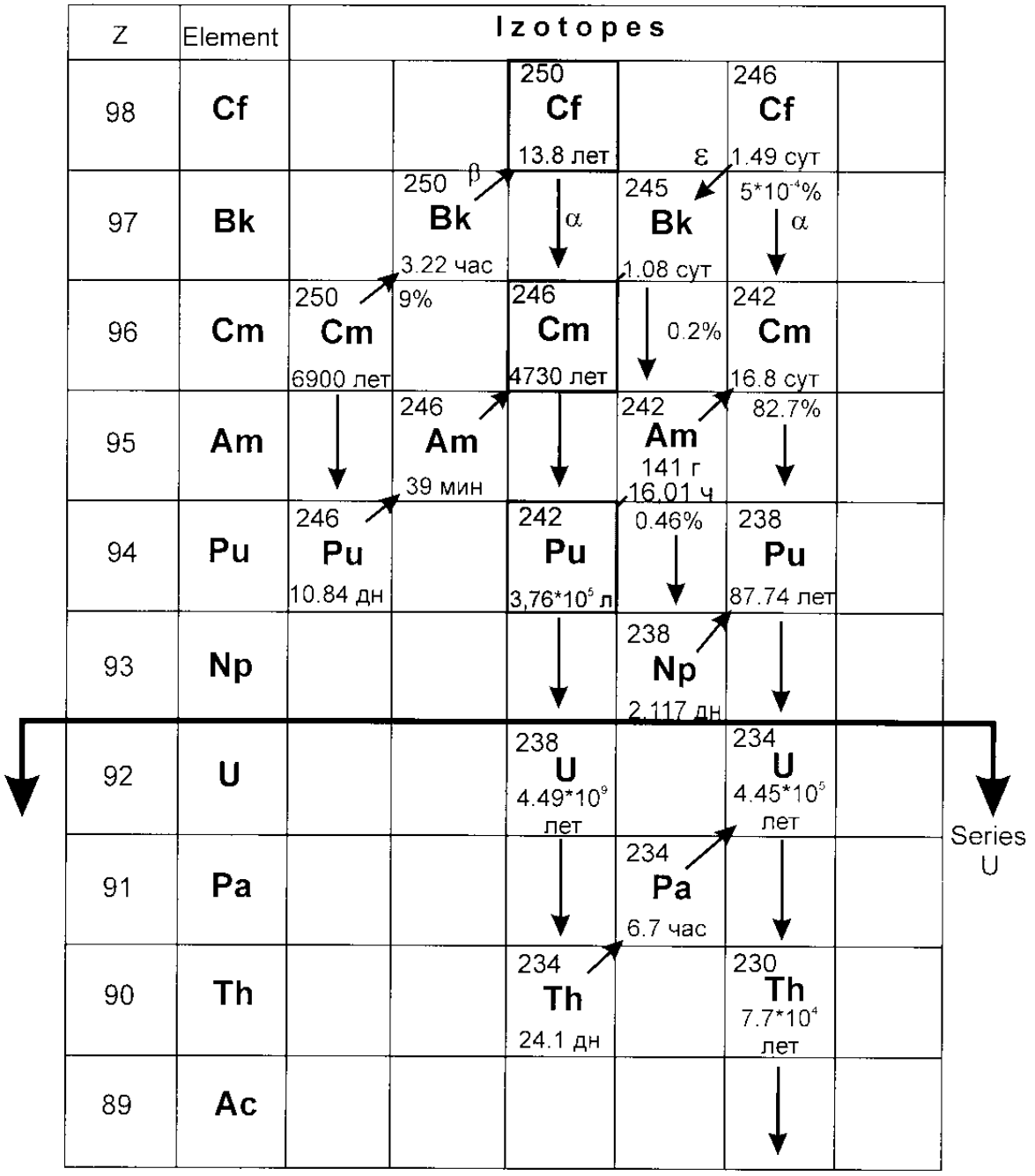


DEPENDENCIES OF OPTIMIZIATION FACTOR (G=JxZ) OF SEPARATOIN OF $^{222}Rn/N_2$ MIXTURE UPON THE GAS AND LIQUID FLOW RATES.
W_g - gas flow rate, cm^3/s
W_l - liquid flow rate, cm^3/s
Retentate clearing, membrane absorption module of membrane contactor with flowing liquid carrier, non-porous membrane, polymer - PVTMS, liquid absorbent - water.



PERMSTRACTOR WITH FLOWING LIQUID CARRIER, THE EQUIPMENT FOR PROCESSING OF LIQUID RADIOACTIVE WASTE OF NUCLEAR POWER PLANT



PROCESSORS OF URANIUM FAMILY - ONE OF RADIOACTIVE FAMILIESFORMED AT WORK FAST REACTOR.
EXTRACTOR OF THE SEPARATE MEMBERS OF SERIES OF RADIONUCLIDES ^{250}Cf, ^{246}Cm, ^{242}Pu ON THE PERMSTRACTOR WITH FLOWING LIQUID CARRIER.