Production of stable isotopes by membrane method

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Separation of Water Isotopomers by Porous Hydrophobic Membrane

Water enriched with its natural isotopes plays an important role in research and technology. Heavy water (HDO, D_2O) is used in nuclear technology and research and the increasing market demand is expected in future if nuclear fusion will be used for energy production.

Water enriched in ^{18}O is used in research and medicine in trace experiments, as is water enriched in ^{17}O. Recently there appeas to be significant market demand for increased production of heavy oxygen (^{18}O) (*see next chapter*). Its role is becoming more important as large amounts of heavy oxygen is used for PET scanning.

The new method of separation of water isotopomers proposed in the project is thermal evaporation through a porous hydrophobic membrane (membrane distillation).

The unit separation factor for the process was determined in experiments carried out on a simple laboratory apparatus, equipped with PTFE flat sheet membrane. The experiments show the membrane process is characterized by higher separation factors than distillation of water. Since distillation is the only method for heavy oxygen production the proposed process has particular importance for this isotope production. In some cases it can be also applied for a production of heavy water. Preliminary engineering calculations based on the cascade theory showed many advantages of membrane permeation. Employing the system of two counter-current cascades combined in series results in savings in stage number, reflux ratio, and energy demand. The technical and economic evaluation of permeation as compared to other enrichment methods showed the competitiveness of the membrane process. The process is tested now on two multi-stage cascades operating under different conditions.

The method can be applied for a separation of isotopes of hydrogen and oxygen in natural water. It can be used separately or in combination with other separation processes.

Production of heavy oxygen (^{18}O)

^{18}O is widely used in research on the mechanisms of catalytic reactions. Double-labelled water with ^{18}O and D is employed in metabolism studies to measure energy expenditure and the total body water composition in human subjects especially when subjected to extreme conditions, e.g. during surgical operations, persons under treat, etc.. To increase precision of measurements triple-labelling is sometimes employed (^{18}O, ^{17}O and D). In contrast to other oxygen isotopes ^{17}O possesses a magnetic moment, which allows easy detection using NMR.

Over the past few years the world has witnessed a continuously increasing demand for enriched oxygen isotopes, especially ^{18}O, due to a large consumption of $H_2{}^{18}O$ by positron emission tomography (PET, a new medical diagnostic technique used principally for tumour detection). PET uses short-lived positron emitters, like carbon-11, nitrogen-13, oxygen-15 and fluorine-18 incorporated into bio-chemically active tracer molecules absorbed preferentially by the tumour. The subsequent radioactive decay monitored by sophisticated position sensitive detectors permits to tumour or target organ to be mapped at high resolution. Several different target materials are used for a production of these isotopes (Table 1), among them water enriched in ^{18}O.

Table 1. Target system.

Nuclide	*Half-period [min]*	*Reaction*	*Target material*
^{11}C	***20.4***	***^{14}N (p, α) ^{11}C***	***Nitrogen gas (natural)***
^{13}N	***9.97***	***^{16}O (p, α) ^{13}N***	***Water (natural)***
^{15}O	***2.07***	***^{15}N (p n) ^{15}O***	***Nitrogen gas (^{15}N - enriched)***
^{18}F	***109.8***	***^{18}O (p, n) ^{18}F***	***Water (^{18}O – enriched)***

Heavy oxygen water ($H_2{}^{18}O$) is used as a target material for production of the short-lived radioisotope ^{18}F used in PET scanning. Fluorine–18 is obtained efficiently using the nuclear reaction: ^{18}O (p, n) ^{18}F, induced in small PET cyclotrons (~11 MEV). As ^{18}F and the other product isotopes in Table 1 are short-lived, the cyclotrons are installed directly in hospitals or clinics. A typical tomography centre comprises specialized cyclotron for short-lived positron isotope production, a laboratory for the synthesis of labelled tumor-specific compounds, and a positron tomograph.

In the 1990's at Institute of Nuclear Chemistry and Technology the new method of heavy oxygen enrichment in natural water was elaborated. The method based on permeation through porous, hydrophobic membrane, called membrane distillation produces higher separation factors than distillation of water. Unit separation factors in membrane process were determined in the experiments carried out with a simple laboratory apparatus equipped with flat sheet PTFE membrane (Fig. 1).

The experiments showed the separation factors of membrane permeation process are markedly higher than those obtained for distillation of water. Since the distillation is the main process used for heavy oxygen enrichment the membrane process is of particular importance. Preliminary engineering calculations based on the separation cascade theory showed the advantages of membrane permeation. The application of the double system of counter-current cascades connected in series (Fig. 2) resulted in the reduction of number of stages, reflux ratio, energy consumption. Technological and economical evaluation of permeation in comparison with other methods used for oxygen isotope enrichment showed the competitiveness of membrane process. The method proposed in patents [1–5] can be used separately or in combination with other separation processes.

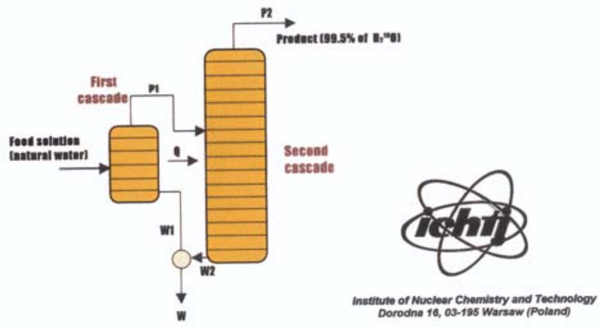

Fig. 2. Double system of separation cascades for isotopes enrichment.

The methods of heavy oxygen enrichment are very expensive and very often difficult in their technological accomplishment. Effective processes as thermodiffusion or chemical isotope exchange are characterised by low kinetics. NO distillation exhibits a large separation factor (Table 2), however it is disadvantageous because of high price of feed material, its toxicity, difficulty with handling and inconveniently low process temperatures.

Table 2. Comparison of heavy oxygen enrichment methods.

Process	Unit separation factor	Energy consumption per 1 kg $H_2{}^{18}O$ [GJ]	Apparatus	Industrial hazard
Water distillation	**1.0032**	**4–8**	**Simple, Normal carbon steel**	**Safe**
Water permeation	**1.005-1.04**	**1–12**	**Simple, Normal carbon steel, plastics**	**Safe**
NO distillation	**1.0406**	**–**	**Complicated, special materials, corrosion hazard**	**NO-toxic substance**

PATENTS

1. A. G. Chmielewski, G. Zakrzewska-Trznadel, N. Miljević, W.A. Van Hook, Sposób wzbogacania wody w składniki wody ciężkiej, PL161104 (31.05.1993).
2. A. G. Chmielewski, G. Zakrzewska-Trznadel, N. Miljević, W.A. Van Hook, Sposób wzbogacania wody naturalnej w tlen-18, PL 16 1105 (17.05.1994).
3. W. A. Van Hook, A. G. Chmielewski, G. Zakrzewska-Trznadel, N. Miljević, Enrichment of Water in Components of Heavy Water, U.S.Patent 5,084,181, (Jan. 28, 1992).
4. W. A. Van Hook, A. G. Chmielewski, G. Zakrzewska-Trznadel, N. Miljević, Method of Enrichment of Oxygen-18 in Natural Water, U.S. Patent No 5, 057, 225 (Oct. 15, 1991).
5. A. G. Chmielewski, G. Zakrzewska-Trznadel, N. Miljević, W.A. Van Hook, Sposób wzbogacania wody naturalnej w tlen-18, PL 168 152 (09.08.1995).