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Continuous Isotope Separation in Systems with Solid Phase. I. Gas-Phase Separation of Isotopes of the Light Elements

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ABSTRACT

A continuous countercurrent process for separation of isotopes of the light elements in gas–solid systems is reported. To separate hydrogen isotopes a hydride-forming palladium adsorbent and a zeolite 13A were used as a solid phase. The molecular sieve was also used to separate oxygen, nitrogen, and argon isotopes. The simulated moving-bed technique (SMB) was employed to organize a countercurrency between gas and solid phases without actual movement of the latter phase. Details of the experimental SMB unit operating in the gas phase are presented, and the results of the separation tests are discussed. A multichannel disk rotary valve was used to switch the flows. It is shown that the process takes advantage of a continuous operation and possesses very high separation efficiency with HETP values comparable to those for the equivalent conventional chromatographic process. It appears that the process proposed is ideally suited for processing tritium-containing streams to produce commodity-grade tritium and may be effectively used to separate isotopes of other light elements.

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INTRODUCTION

Production-scale separation of stable isotopes employs common countercurrent processes in vapor–liquid or gas–liquid systems. It is known, however, that high values of the equilibrium separation factor α, defined as

$$\alpha = \frac{y(1 - y)}{x(1 - x)}$$

(where y is the atom fraction of an isotope in the solid phase and x is the atom fraction of this isotope in the gas phase) are also observed in many gas–solid systems. Isotope fractionation in such systems can be due to physical adsorption of the gas, comprising isotopes to be separated, as for example in the case of adsorption of H_2, O_2, N_2, or Ar on zeolites (1). Separation effects may also be caused by the isotope chemical exchange between two different chemical species when gas and solids are in contact (2), as in the case of exchange between molecular hydrogen and a hydride of palladium (3).

A vast amount of data is available in the literature on both thermodynamics and kinetics of the isotope exchange in gas–solid systems (see, for example, citations in Ref. 4). That information clearly indicates that many of these systems may be employed to operate various isotope separations efficiently. Thus, the system hydrogen–solid adsorbent is of particular interest for application to recover tritium at nuclear power plants (5). Creating safe and robust hydrogen isotope separation apparatuses for tritium fuel cycles and for securing radiation safety in the ITER (international thermonuclear experimental reactor) is another important engineering problem where solid adsorbents of hydrogen isotopes are expected to be in growing demand (6). The substances to be used in these applications should not be subject to radiolysis by tritium radiation, which is why most of the vapor(gas)–liquid systems currently in use for industrial isotope separations cannot be applied here. At the present time the cryogenic distillation of hydrogen, developed by Sulzer Brothers Ltd., seems to be the only continuous process for tritium-containing streams processing which has been brought into production. It is designed to recover tritium from a heavy water moderator at the experimental nuclear reactor in Grenoble (7). It is not clear, however, whether this method can be used to process hydrogen isotope feeds in thermonuclear reactor programs because of the potential hazard associated with high tritium inventory needed in the process, very complicated schemes and equipment, and high energy consumption (8).

As a result, considerable effort has been expended in recent years to develop new techniques for separating hydrogen isotopes with particular emphasis on systems with high radiation stability. Thus, Suzuki and Kimura reported hydrogen isotope separation by a palladium alloy membrane (9). They developed a cascade design in which membrane columns were used as unit cells. This system, as shown, can separate and concentrate isotopes of hydrogen. Matsumoto et al. performed the separation of hydrogen isotopes in a "cryogenic-wall" thermal diffusion column (10). The separation were carried out with the H-D system under total reflex and in the continuous feed operation mode.

Most work, however, has been devoted to studying gas chromatographic systems utilizing hydride-forming metals, intermetallic compounds, or molecular sieves as the solid adsorbent of hydrogen (11–15). Despite high separation factors and fast isotope exchange kinetics observed, gas–solid systems have not been brought into industrial isotope enrichment and fractionation because of the periodical nature of chromatographic separation. Some research has attempted to optimize conventional chromatography process schemes. Embury et al. (12) investigated a rapid cycling, computer-operated displacement chromatograph for the separation of hydrogen isotopes. This device incorporated several features that optimized product throughput and purity. The operations were carried out with a protium–deuterium mixture with palladium as adsorbent. A semi continuous metal hydride-based hydrogen isotope separation process was studied by Horen and Lee from the Savannah River Laboratory (13). An experimental, large-scale unit was tested using protium and deuterium. A 55% protium and 45% deuterium mixture separation resulted in protium and deuterium products with purities better then 99.5%.

A continuous countercurrent process for separation of hydrogen isotopes was studied by Singh (16) in a moving-bed column. A silica gel and an activated carbon were used as a solid phase. This process, commonly referred to as hypersorption, is a continuous operation. It encountered many problems when operating because of difficulties associated with actual solids movement (attrition, cumbersome solids handling system for recycling, nonuniformity of solid flow, etc.).

The solids problems, however, may be avoided by using a simulated countercurrent moving-bed process (SMB) instead of a moving-bed operation. This approach was originally developed for continuous chromatographic separation (17). One of the variants of its practical realization implies the use of a number of connected sections with a stationary fixed-bed solid phase (18). In SMB units the solid beds are fixed and their continuous movement is simulated by a discrete movement obtained by shifting the feed and withdrawal points at discrete times along the column axis in the same direction as the fluid flows. Very recently this principle has been applied for separation of hydrogen isotopes in the gas phase (19) and for nitrogen isotope separation in the liquid phase (20).

It is believed that gas–solid systems may also be efficiently utilized for separation of isotopes of light elements such as O, N, Ar, etc. In some cases the equilibrium separation factors in these systems noticeably exceed those for gas–liquid or vapor–liquid systems when separation is carried out by distillation or chemical exchange (4). These traditional industrial methods are characterized by high energy consumption and increased ecological hazard due to the high volumes of chemicals consumed and large amounts of wastes (4). When a zeolite is used for separation of isotopes by adsorption, the reflux of the streams in the separation column can be carried out in a thermal swing operation mode and, therefore, without spending chemicals. This may substantially reduce the operating costs and improve environmental aspects of the process.

In the present work we describe the principles and report the results of the separation of isotopes of light elements (H, O, N, and Ar) in a countercurrent SMB unit operating in the gas phase. Two modes of operation—under total reflux and with a continuous feed operation—will be considered. Details of the experiment will be given, and the results of separation tests will be discussed. To separate hydrogen isotopes, a hydride-forming palladium adsorbent and a zeolite 13A were used as a solid phase. A molecular sieve was also used to separate oxygen, nitrogen, and argon isotopes.

SCHEMATICS OF A SIMULATED MOVING BED PROCESS FOR CONTINUOUS GAS-PHASE SEPARATION OF ISOTOPES

A schematic representation of the SMB separation unit consisting of five fixed-bed sections is shown in Fig. 1. Sections in Positions 2, 3, and 4 form the separation zone with gas and solids in counter contact. Sections in Positions 1 and 5 serve as flow conversion units in which isotopes to be separated are transferred from one phase to the other. In the adsorber (Position 5) the gas is adsorbed or chemically bound by solids. In the desorber (Position 1) it is desorbed, forming a fluid flow along the column. It is noteworthy that in the countercurrent adsorptive separation unit the adsorber and the desorber play essentially the same roles as the reboiler and condenser in a distillation column.

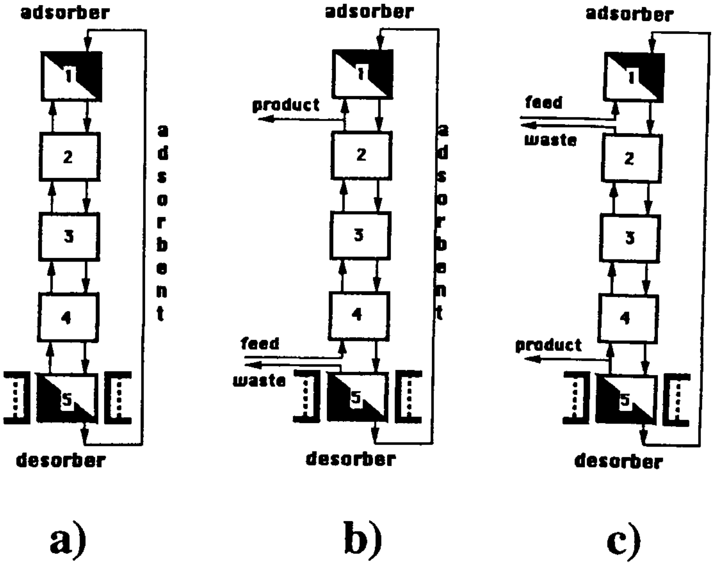

FIG. 1 Schematic representation of the simulated moving-bed process for continuous isotope separation in gas–solid systems. (a) column with total reflux, (b) and (c) continuous feed operation: (b) column concentrating less strongly adsorbed isotope, (c) column concentrating more strongly adsorbed isotope.

Schematic (a) in Fig. 1 represents the configuration of an SMB column under total reflux, i.e., when neither feed nor products are introduced or withdrawn. After gas is completely released from the solid phase in the desorber (Position 1), this section is moved up and becomes an adsorber (Position 5). Simultaneously, the section in Position 4 shifts down and becomes a desorber. The section which was in Position 5 moves to the separation zone, and so on. In other words, each section moves down one position countercurrently to the gas flowing up. As a result of this simulated countercurrent moving-bed operation, the less strongly adsorbed isotopic species is concentrated in the upper part of the column (in the adsorber) and the more strongly adsorbed isotopic species is concentrated in the bottom (in the desorber).

When the less strongly adsorbed isotope is subject to concentration (heavy hydrogen isotopes in the H_2–Pd system and light isotopes of H, O, N, or Ar in the system with zeolite), the feed has to be applied to the bottom of separation zone [section in Position 2, Fig. 1(b)]. After flowing sequentially through the three sections in the separation zone, the gas eventually enters the adsorber. Part of it, enriched with the less strongly adsorbed isotope, is withdrawn as a product. In turn, the solid phase, depleted of the less strongly adsorbed isotope (and enriched with the more strongly adsorbed component), moves down and enters the desorber operating at elevated temperature. The desorbed gas is taken out of the column as a waste. The solid phase, free from sorbate, is then recycled to the upper end of the column.

When a more strongly adsorbed isotope has to be concentrated, the feed to the column in composed of solids: the gaseous feed is applied to the adsorber so that the solid phase in it becomes saturated with gas of the initial isotope concentration. After the flows are switched, this section shifts to Position 4, Fig. 1(c). Passing sequentially from Positions 4 to 2 in the separation zone, the solids becomes enriched with the more adsorbable isotope (light hydrogen isotopes in the H_2–Pd system, and heavy isotopes of hydrogen, oxygen, etc. in the system with zeolite). Finally, this section becomes a desorber. The desorbed gas is turned back to the column, and part of it may be withdrawn as a product.

When both more and less adsorbable isotopes are subject to concentration, the feed has to be applied to the middle of the column.

EXPERIMENTAL SETUP

Based on the process principles outlined in the previous section, an experimental SMB unit operating in the gas phase has been designed and constructed. A schematic diagram of the experimental setup is presented in Fig. 2.

The simulated moving bed consisted of five uniform sections packed with granulated adsorbent. The bed was 11 mm in diameter for all experiments whereas the height of a bed in a single section varied from 10 to 56 mm. Two sections were always used as flow conversion units (adsorber and desorber), and the other three formed the separation zone. The desorber was maintained under the temperature at which a complete desorption of gas occurred (493 K for palladium-based adsorbent and 298 K for molecular sieve). The adsorber, along with the sections in the separation zone, were maintained at the operating temperature (273–353 K for palladium-based adsorbent and 77 K for zeolite).

To operate the flow switches in the simulated moving-bed system, a specially designed multichannal disk rotary valve was built. All fixed bed sections were connected to the turning part of the valve, whereas the communications between them along with the side streams were connected to the stationary part of the valve. Both parts of the valve were fabricated from stainless steel with a hard-alloy coating applied by welding

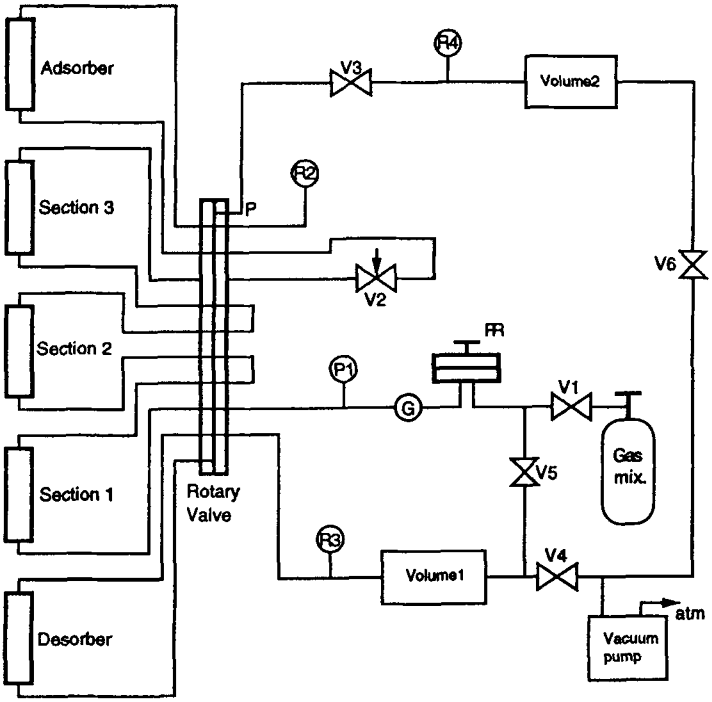

FIG. 2 Schematic diagram of the simulated moving-bed experimental setup. PR: Pressure regulator; P1–P4: pressure gauges; V1, V3–V6: on–off valves; V2: needle valve; P: port for analysis.

onto the disk surfaces. To avoid leakage, both disk surfaces were polished to a high degree of flatness and then lubricated.

The countercurrent flow between solid and gas phases was achieved by turning the moving part of the valve together with the sections in the direction opposite to that of the gas flow. The oven for the desorber and the thermostat for the other sections were arranged on a slab which was moved down and then lifted up every time the rotary valve was actuated and the sections were turned. The switching procedure was operated automatically. The gas flow rate in the column was adjusted by a needle valve (V2) at the adsorber inlet and was measured by a flowmeter (G). The system was maintained at constant pressure with a pressure regulator (PR).

Prior to the experiments under total reflux, the system was heated up under vacuum to desorb any gases from the adsorbent. All five sections were filled with the feed mixture by opening valves V1 and V5. Next, all the sections except for the desorber were cooled down to operating temperature so that the solid phase became saturated with sorbate. The temperature in the desorber was chosen so that no adsorption occurred under the operating pressure. When the first switch occurred, the desorber with solids, free of sorbate, was moved up and became an adsorber (see Fig. 1). Having been cooled in the thermostat, it immediately started to adsorb the gas. Simultaneously, the section which had been in Position 1 (Fig. 1) was shifted to the oven and became a desorber. The gas released from the solid phase flowed through Volume 1 (Fig. 2) and a pressure regulator (PR) toward the first section in the separation zone (Section 1 in Fig. 2). After the desorption was complete, [a condition recognized by no flux through the flowmeter (G)], a successive switch of the flows was executed.

The separation experiments were performed until a steady-state isotope concentration profile was formed. The concentration of the less strongly adsorbed isotope increased in the direction from the desorber toward the adsorber.

A drawback to the scheme under total reflux is that the overall separation factor attained in the column is reduced due to mixing of two streams with different isotope concentrations. Clearly, after desorption is complete in the desorber, the void space in this section still contains gas enriched with the more adsorbable isotope. Having then been transported to the top of the column, this section becomes an adsorber [Position 5 in Fig. 1(a)] and is fed with the gas enriched with the less adsorbable isotope.

The column startup in the continuous feed experiments [Schemes (b) and (c) in Fig. 1] was performed in a manner similar to that described above for the configuration under total reflux. The desorber was continuously pumped out by a vacuum pump through valve V4 whereas the valve V5 was off (see Fig. 2). When a section is moved out of the desorber oven, it contains no gas in the void space. Feed to the column was applied to Section 1 in the separation zone. When formed, the steady-state isotope concentration profile was characterized by the concentration of the more strongly adsorbed isotope increasing in the direction from the desorber toward the adsorber.

To determine the isotope concentration profiles in the column, the rotary valve was provided with a special port (P). Gas from each section could be desorbed to Volume 2 for analysis (see Fig. 2).

MATERIALS AND METHODS

For experiments on hydrogen isotope separation the feed was prepared by mixing natural abundance hydrogen and deuterium containing 99.95

atom% D, both of industrial grade purity. No further purification of the gases was attempted.

The hydride-forming palladium-based adsorbent, containing 75 wt% Pd and 25 wt% Al, was prepared. Palladium and aluminum powders (10–40 μm particle size) were mixed, pressed to pellets, and heated in a flow of inert gas. Then the resulting solid was crushed, sieved, and the 0.5–1.0 mm particle size fraction was packed into sections. Prior to the experiments the adsorbent was heated to 523 K under argon atmosphere and vacuumed to 1 Pa. It then underwent several hydrogen sorption–desorption cycles to obtain constant capacity.

The hydrogen isotope separation experiments were also attained with the use of zeolite 13A particles ranging in size from 0.5 to 1.0 mm. Before the experiments the adsorbent was heated to 623 K while an inert gas was blown through the bed; the bed was then pumped out to 1 Pa.

Some characteristics of the adsorbents used are listed in Table 1. Analysis of hydrogen isotopes was by atomic spectroscopy.

For the experiments on separating the isotopes of oxygen, nitrogen, and argon, corresponding gases of industrial grade purity were used without any further purification. Zeolite 13A with particle sizes in the 0.5 to 1.0 mm range was used as a solid phase. Some adsorbent characteristics are given in Table 2. For analysis of isotopes of O, N, and Ar, isotope mass-spectrometer MI-1101 was used.

TABLE 1
Some Characteristics of the Adsorbents Used for Hydrogen Isotope Separation

Parameter	Pd adsorbent (T = 296 K)	Zeolite 13A (T = 78 K)
Apparent density, kg/m^3	1500	710
Capacity, mL H_2 (STP)/g sorbent (P_{H_2} = 1.0 bar)	45	120
Capacity of a fixed bed of 10 mm height, mL H_2 (STP)	64	80
Equilibrium separation factor (5, 21):		
Mixture	α	
H_2–D_2		2.07
H_2–HD	2.02	
HD–D_2	2.49	
H_2–T_2		2.57
H_2–HT	2.68	
HT–T_2	4.16	
D_2–T_2		1.24
D_2–DT	1.47	
DT–T_2	1.54	

TABLE 2
Some Characteristics of the Zeolite 13A Used for Separation of O, N, and Ar Isotopes

Parameter	Gas phase O_2	N_2	Ar
Capacity, mL (STP)/g sorbent	175	130	140
Capacity of a fixed bed of 10 mm height, mL (STP)	116	86	93
Equilibrium separation factor at T = 78 K (22):			
Isotopic mixture	$^{16}O_2$–$^{16}O^{18}O$	$^{14}N_2$–$^{14}N^{15}N$	^{40}Ar–^{36}Ar
α	1.020	1.016	1.016

RESULTS AND DISCUSSION

Hydrogen Isotope Separation

On the basis of the steady-state isotope concentration profiles obtained along the column, the overall separation factor K was calculated:

$$K = \frac{x_1/(1 - x_1)}{x_2/(1 - x_2)}$$

where x_1 and x_2 are the atomic fractions of the isotope to be concentrated and measured at the two ends of the column; subscripts "1" and "2" indicate the ends where this isotope is enriched and depleted, respectively. The height equivalent to a theoretical plate (HETP) was computed by assuming that isotope distribution along the column may be described by the Fencske equation (2):

$$K = \alpha^N$$

The experiments on hydrogen isotope separation by the palladium-based adsorbent were carried out with use of protium–deuterium and protium–tritium mixtures with various concentrations of heavy isotope in the feed. For the deuterium–tritium experiments the deuterium containing trace concentrations of tritium was used. The separation tests were performed by using various temperatures in the desorber and separation zone, various operating pressures, various hydrogen flow rates, and by varying the height of the bed in a single section, h_{bed}. The operating conditions as well the results of the runs carried out are presented in Table 3. As an example, Fig. 3 shows the steady-state deuterium and tritium concentration profiles along the SMB column for some of the runs listed in Table

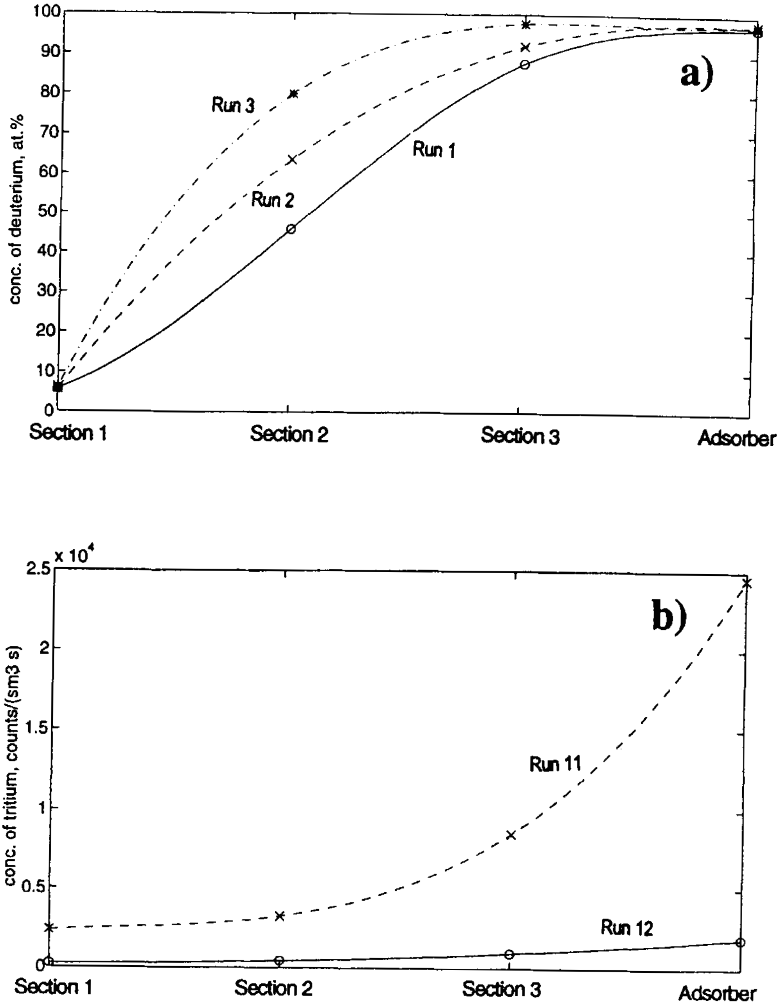

FIG. 3 Steady-state isotope concentration profiles along the SMB column. (a) protium–deuterium separation (Runs 1–3 in Table 3); (b) deuterium–tritium separation (Runs 11 and 12 in Table 3).

3. From the results given in Table 3, one can see that the data obtained confirm the very high separation efficiency of the process. Thus, at a total bed height in the separation zone of 30 mm, the deuterium was concentrated from 5.7 to 96.9 atom% (Run 1) with HETP = 2.0 mm and the overall separation factor $K = 528$.

TABLE 3
The Results of the Hydrogen Isotope Separation Experiments with Use of Palladium-Based Adsorbent

						Average concentration of heavy isotope (atom%)				
Run	Isotopes separated	h_{bed} (mm)	P (bar)	T (K)	Flow rate of H_2 (L/hr)	Section 1	Section 2	Section 3	Adsorber	HEPT (mm)
1	H–D	10	1.0	296	6.8	5.7	45.9	94.8	96.9	2.8
2	H–D	20	1.0	296	7.6	5.7	63.6	97.1	97.2	5.0
3	H–D	56	1.0	296	10	6.1	80.0	97.7	97.8	12
4	H–D	10	1.0	276	3.0	5.7	36.8	91.0	96.7	3.7
5	H–D	10	1.0	296	3.0	5.7	32.3	87.1	96.5	3.7
6	H–D	10	1.0	336	3.0	5.7	18.5	88.8	94.0	3.7
7	H–D	10	0.5	296	3.0	5.7	30.5	85.9	96.4	3.7
8	H–D	10	1.0	296	5.1	5.7	39.5	93.5	96.7	3.1
9	H–T	30	0.8	293	4.0	4.6	18.8	87.3	98.2	14
10	H–T	30	0.8	293	4.0	2.5	8.8	85.6	97.7	14
						Activity of tritium (counts/cm^3·s)				
11	D–T	10	1.0	296	5.9	2,370	3,250	8,500	24,500	3.5
12	D–T	10	1.0	296	5.1	240	440	950	1,900	3.5

Figure 4 shows the effect of the height of the bed in a single section on HETP for protium–deuterium experiments at 296 K and 1.0 bar. The separation efficiency of the column is decreases (HEPT increases) as h_{bed} increases. The reason for this effect may be due to the low number of the fixed beds in the separation zone (three), and as a result there is a quite poor approximation to the equivalent true countercurrent process. The absence of HETP dependence on the hydrogen flow rate (Fig. 5) may possibly be explained by the dominant contribution of the axial dispersion effects in the apparatus under the conditions tested. High variations in the hydrogen flow observed within the time interval between two successive flow switches is the likely reason of such behavior. Just after the flows were switched, a sharp increase in the hydrogen flow rate was always registered. The high adsorption rate in the adsorber immediately after switching flows resulted to a noticeable pressure drop in the system. This temporarily caused a high hydrogen flow rate through the column, so that equilibrium between the gas and solid phases in the sections was not quite attained. If diffusion inside the porous adsorbent is a slow step, HETP would be dependent on the hydrogen flow rate (23).

The experiments on separating the protium–deuterium mixture on zeolite 13A were carried out at atmospheric pressure and a temperature of

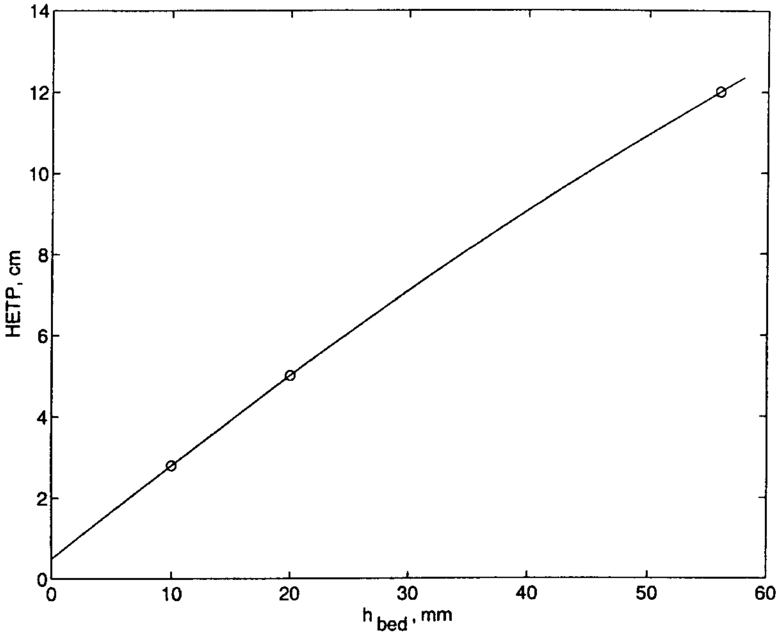

FIG. 4 Effect of the height of a bed in a single section on HETP. T = 296 K, P = 1.0 bar, H_2–D_2 mixture, solid phase: Pd adsorbent.

77 K. To accomplish this, the sections were placed in a tub containing liquid nitrogen. The temperature in the desorber oven was maintained at 296 K. The process was used to concentrate protium in accordance with Scheme (b) in Fig. 1. The results of the separation experiments are presented in Table 4. They show that hydrogen isotopes separation by the molecular sieve is also very efficient. Thus, an overall separation factor of 990 (Run 2 in Table 4) was attained at a total bed height in the separation zone of 30 mm. By comparison with the data listed in Tables 3 and 4, one may see that the HETPs obtained for the palladium-based adsorbent and for zeolite 13A are close, which means that the chemical reaction constituent of HETP is insignificant in both cases. This also agrees with the fact that there was no effect of temperature on HETP in the range of the operating parameters studied (see Runs 4–6 in Table 3).

On the basis of the results obtained in this study, a pilot-scale unit for the continuous processing of tritium-containing streams to produce commodity-grade tritium has been designed and constructed. It is currently being tested using real protium–tritium systems at one of the plants in the Russian Federation.

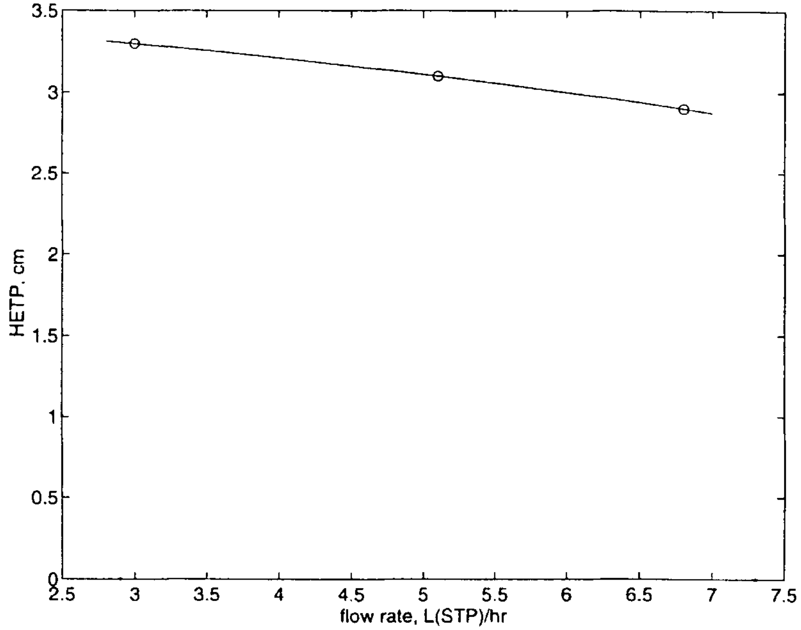

FIG. 5 Effect of the hydrogen flow rate on HETP. $T = 296$ K, $P = 1.0$ bar, H_2–D_2 mixture, $h_{bed} = 10$ mm, solid phase: Pd adsorbent.

TABLE 4
The Results of the Experiments on Hydrogen Isotope Separation with Use of Zeolite 13A

Run	h_{bed} (mm)	Hydrogen flow rate (L/h)	Concentration of protium in the feed (atom%)	Average concentration of protium in adsorber (atom%)	HETP (mm)
1	10	3.4	3.1	84.2	3.8
2	10	8.8	10.0	99.1	2.6

Separation of Oxygen, Nitrogen, and Argon Isotopes

The experiments on oxygen, nitrogen, and argon isotope separation were performed on the scheme under total reflux [see Fig. 1(a)]. The temperature in the separation zone and the adsorber was kept at 77 K with liquid nitrogen; in the desorber the temperature was 296 K. The

TABLE 5
The Results of the Experiments on Separation of O, N, and Ar Isotopes with Use of Zeolite 13A

			Average concentration of ^{18}O, ^{15}N, or ^{36}Ar (atom%)					
Run	Isotopes separated	*P* (bar)	Section 1	Section 2	Section 3	Adsorber	Overall separation factor	HEPT (mm)
1	^{16}O–^{18}O	0.197	0.460	0.433	0.409	0.388	1.186	3.5
2	^{14}N–^{15}N	0.790	0.00778	0.0743	0.00710	0.00681	1.142	3.5
3	^{40}Ar–^{36}Ar	0.263	0.342	0.356	0.377	0.390	1.140	3.5

height of the bed in each section was 10 mm, and the gas flow rate was set at 4.0 L/h (STP). For each system the operating pressure was adjusted to avoid condensation in the zeolite pores at 77 K. The results of the experiments are presented in Table 5. One can see that the overall separation factor obtained is close to unity for all systems. This is because the equilibrium separation factors for the isotopic mixtures considered lie in the 1.016 to 1.020 range, which is due to the much smaller difference in the physicochemical properties of distinct isotopic species with respect to hydrogen isotopes. The values of HETP in these experiments is as low as for the separation of hydrogen isotopes, which again indicates the high separation efficiency of the process.

It is interesting to note that equilibrium separation factors in systems with zeolite for the elements under consideration markedly exceed the values observed in corresponding vapor–liquid systems. Thus, for example, for the vacuum distillation of oxygen the value of α is 1.006, whereas it is 1.020 in the system used in our work. Similar behaviors are observed for the isotopes of nitrogen and argon.

CONCLUSIONS

A countercurrent simulated moving-bed process for the separation of isotopes of light elements in gas–solid systems was reported. The scheme under total reflux and continuous feed operation has been considered. The results of the experiments carried out in this study confirm the very high separation efficiency of the process and show that the SMB principle may be effectively employed to perform such "difficult" separations as the fractionation of isotopes.

Most of the work in this research is related to hydrogen isotope separation because of its particular interest for hydrogen energy and nuclear industries. The results on separating isotopes of other light element (oxygen, nitrogen, and argon) appear to be more illustrative and exploratory, and appropriate practical applications of the approach for these systems has yet to be found.

ACKNOWLEDGMENT

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SYMBOLS

K	$= [x_1/(1 - x_1)]/[x_2/(1 - x_2)]$, overall separation factor
N	number of theoretical plates in the column
P	pressure (bar)
T	temperature (K)
h_{bed}	fixed bed height (mm)
x	atom fraction of an isotope in the gas phase
y	atom fraction of an isotope in the solid phase

Greek Letter

α	$= [x/(1 - x)]/[y/(1 - y)]$, equilibrium separation factor

Subscripts

1, 2	ends of the countercurrent separation column

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