ISOSILICON AS

New separation methods for production of light stable isotopes for use in nuclear technology

By

Dag Øistein Eriksen[1], Bruno Ceccaroli[2], Pierre Hilaireau[3], and Wieslaw Majewski[4]

[1] Primus.inter.pares AS, Norway; [2] Isosilicon AS, Norway; [3] Stonedge SA, Luxembourg, [4] Novasep SA, France

3rd – INCC, Sicily - Italy

18 - 23 September 2011

Outline

1. Use of isotopically pure elements
    1. Nuclear technology
    2. Medicine and science
    3. Electronics
2. Isotope separation technology
3. Isosilicon’s separation technology
4. Future isotopes of interest

Stable isotopes – Medicine and science

Medicine:

^{18}O is used as target (H_2O) for production of PET nuclide ^{18}F:
$^{18}O(p,n)^{18}F$

NMR/MRI:

Deuterated compounds and $D_2O = {}^{2}H_2O$
^{13}C-labelled compounds

Tracer studies:

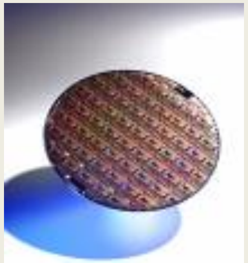
D_2O used as water tracer
^{15}N used in fertilizer studies as NH_4-salts

Stable isotopes – Electronics

NTD Neutron Transmutation Doped
(for high power devices)

$^{30}Si(n,\gamma)^{31}Si \rightarrow {}^{31}P$

homogeniously doped throughout the crystal

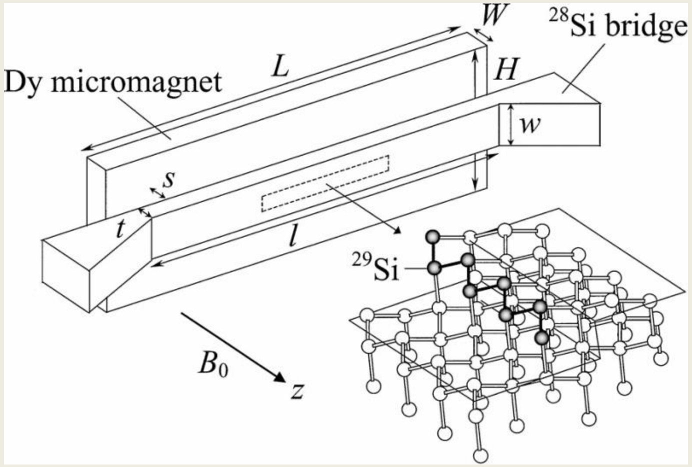


Spiking silicon with ^{30}Si will increase capacity of irradiation facilities

Use of ^{29}Si

Journal of Superconductivity: Incorporating Novel Magnetism, Vol. 16, No. 1, February 2003 (C° 2003)

E. Abe, K. M. Itoh, T. D. Ladd, J. R. Goldman, F. Yamaguchi, and Y. Yamamoto: **Solid-State Silicon NMR Quantum Computer**



Stable isotopes – nuclear technology

- Primarily ^{235}U to be enriched from natural abundance of 0.72%
- Thermal neutron absorbers:
  - ^{6}Li 7.59% $\sigma(n,\alpha)$ 940b
  - ^{9}B 19.9% $\sigma(n,\alpha)$ 3840b
  - ^{91}Zr 11.22% $\sigma(n,\gamma)$ 1.2b, but rep. 77% abs. in Zr
  - $^{155,157}Gd$: 14.8, 15.65% $\sigma(n,\gamma)$ $6.1 \cdot 10^{4}$, $2.54 \cdot 10^{5}$b
- Neutron reflectors:
  - ^{7}Li 92.41% $\sigma(n,\gamma)$ 0.045b
  - ^{10}B 80.1% $\sigma(n,\gamma)$ 0.005b

Gen IV - Very high temperature gas reactors

- HTTR, Japan
- GTMHR, Russia

} Prismatic blocks of fuel

- HTR-10, China
- PBMR, RSA

} Pebble beds of fuel

Common features:

- Graphite moderated
- He cooled
- Operated at approx. 1000°C

Pebble Bed Modular Reactor Pty., RSA

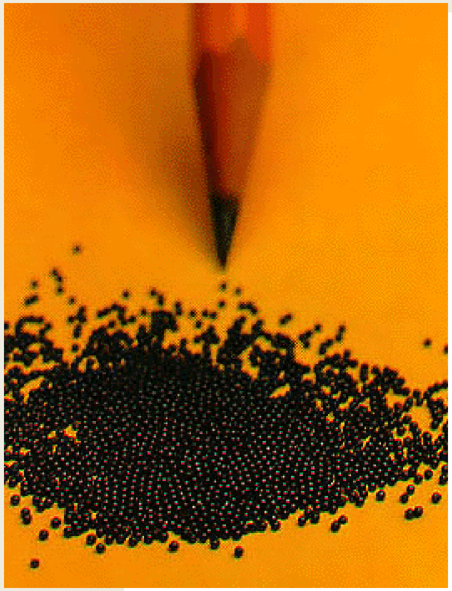
Time frame:

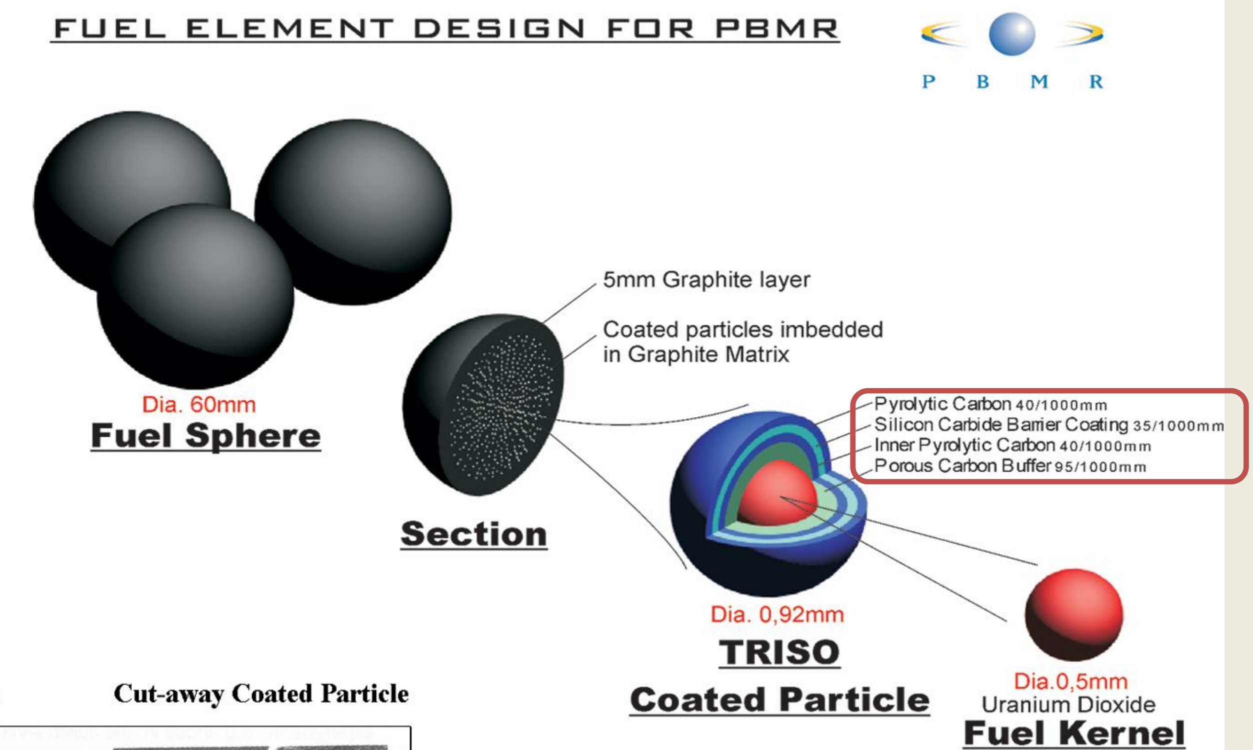
- 1993: Start of development
- 2009: Start construction of pilot reactor
- 2013: Fuel loading
- 2016: Start construction of first commercial PBMR, 165MW

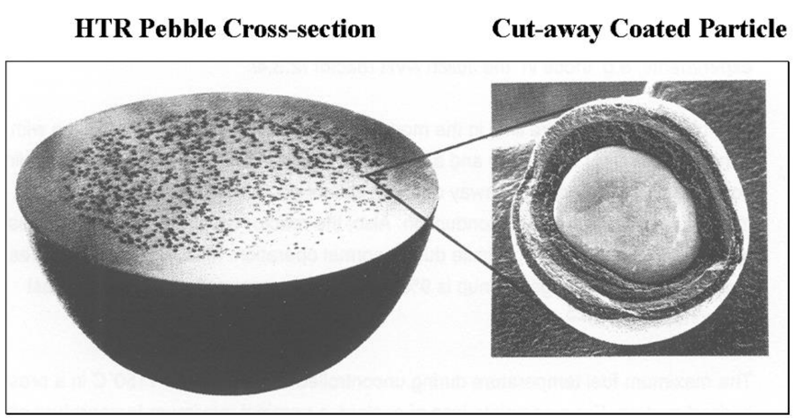
Technical key elements:

- He cooled
- Pressure of 9 bars
- Temperature 500°C in and 900°C out
- Moderator C (graphite) covered with SiC

Pebble Bed Modular Reactor







SiC: ^{28,29}SiC to avoid
$^{30}Si(n,\gamma)\ ^{31}Si \rightarrow\ ^{31}P$C

TRISO = triply coated ceramic particle fuel

Isotope separation technology

- Electrolysis
- Diffusion based
- Membrane based
- Distillation
- Electromagnetic
- Centrifugation
- Gas-jet centrifugation
- Separation nozzle
- Selective exitation by laser
- Ion-mobility
- Isotopic exchange
- Chromatography

Norsk Hydro's heavy water process

$^{1}H_2O \leftrightarrows {}^{1}H^{+} + OH^{-}$

$^{1}H^{2}HO \leftrightarrows {}^{1}H^{+} + O^{2}H^{-}$ or

$HDO \leftrightarrows H^{+} + OD^{-}$

$2\,^{1}H^{+} + 2\,e^{-} \rightarrow {}^{1}H_2$ $\quad E^0 = 0.000V$

$2\,^{2}D^{+} + 2\,e^{-} \rightarrow {}^{2}D_2$ $\quad E^0 = -0.044V$

Handbook of Chemistry and Physics, 64th Edition, CRC Press, Boca Raton, Fl 1983

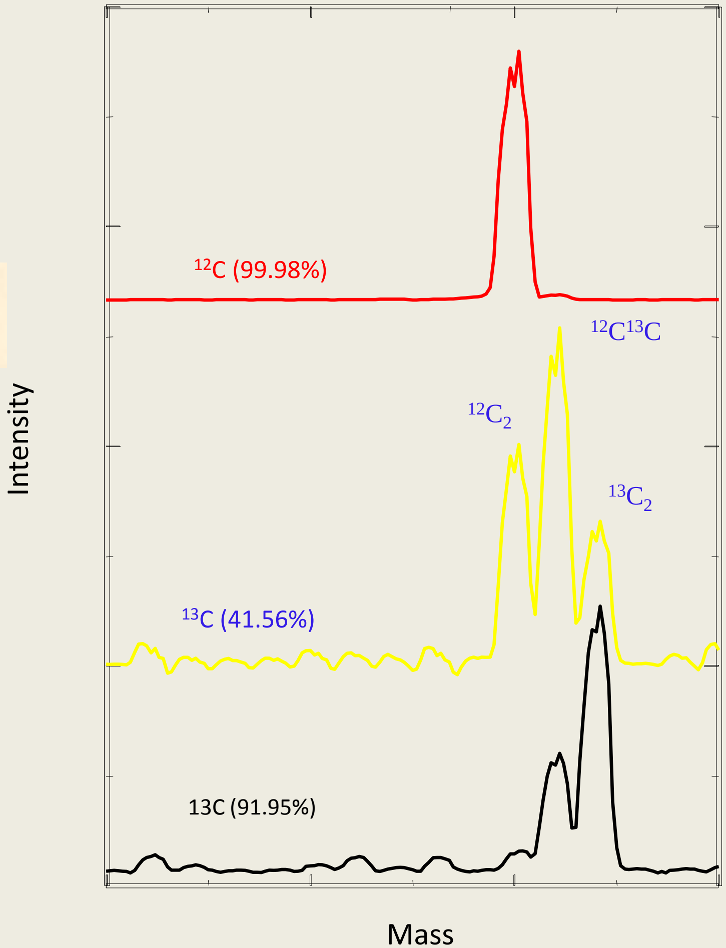
Klydon Ltd.(RSA): Laser Isotope Separation

Laser-based isotope enrichment of Carbon-12/13

- Feed: Freon ($CHClF_2$), Product: C_2F_4

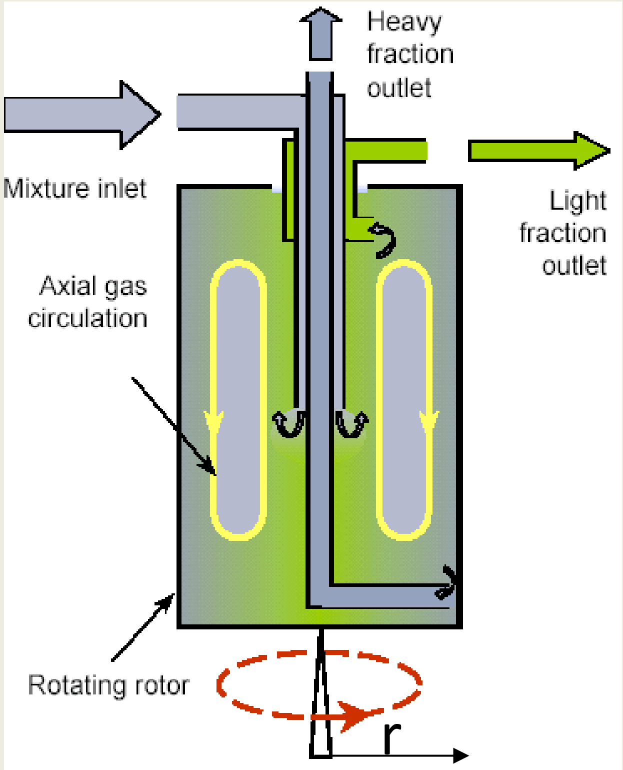
$$CHClF_2 \xrightarrow{h\nu} {}^{13}CHClF_2 + {}^{12}C_2F_4 + HCl$$

- High isotope selectivity achieved



Enrichment Factor for Uranium

Diffusion	Centrifuge			
$\beta = \sqrt{\frac{m^{238}UF_6}{m^{235}UF_6}}$	$\beta = \exp\left[\frac{\Delta m v_a^2}{2RT} \cdot \left(1 - \frac{r}{a}\right)^2\right]$			
$\beta_{max} = \sqrt{\frac{352}{349}} = 1.00429$	**r/a**	**400 m/s**	**500 m/s**	**700 m/s**
	0	1.101	**1.162**	**1.343**
	0.5	1.075	**1.119**	**1.247**
	0.8	1.035	1.056	**1.112**
	1.0	1.0	1.0	1.0
$\beta \approx 1.003$	$1.15 < \beta < 1.25$			

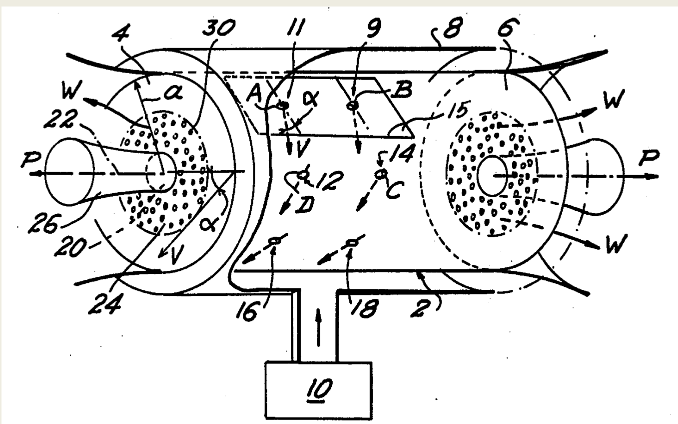


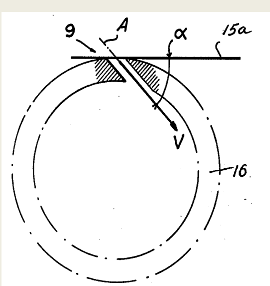
By courtesy of Klydon

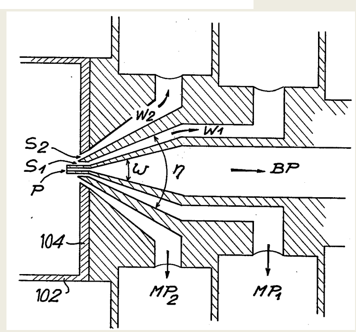
Rosegard Vortex Extraction

October, 1976

Enrichment:	1.056 (Argon)
Cut:	6-8%



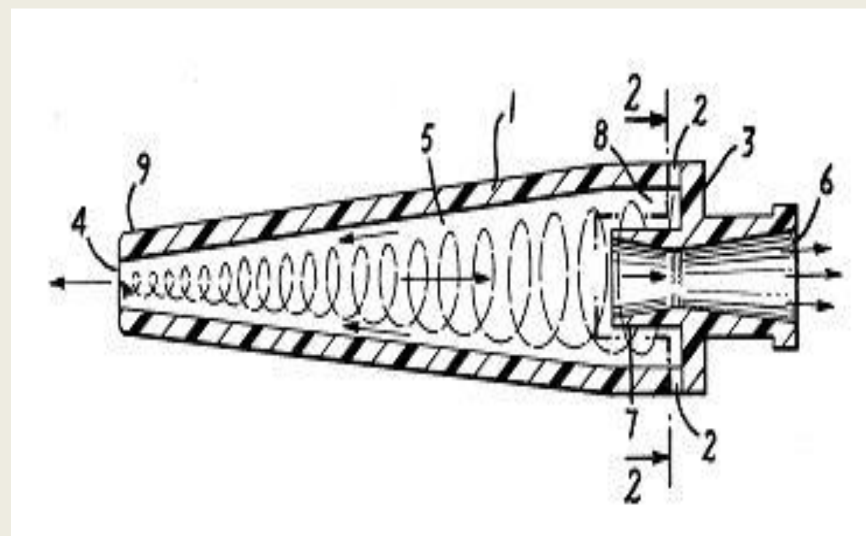




Wikdahl Vortex Separation

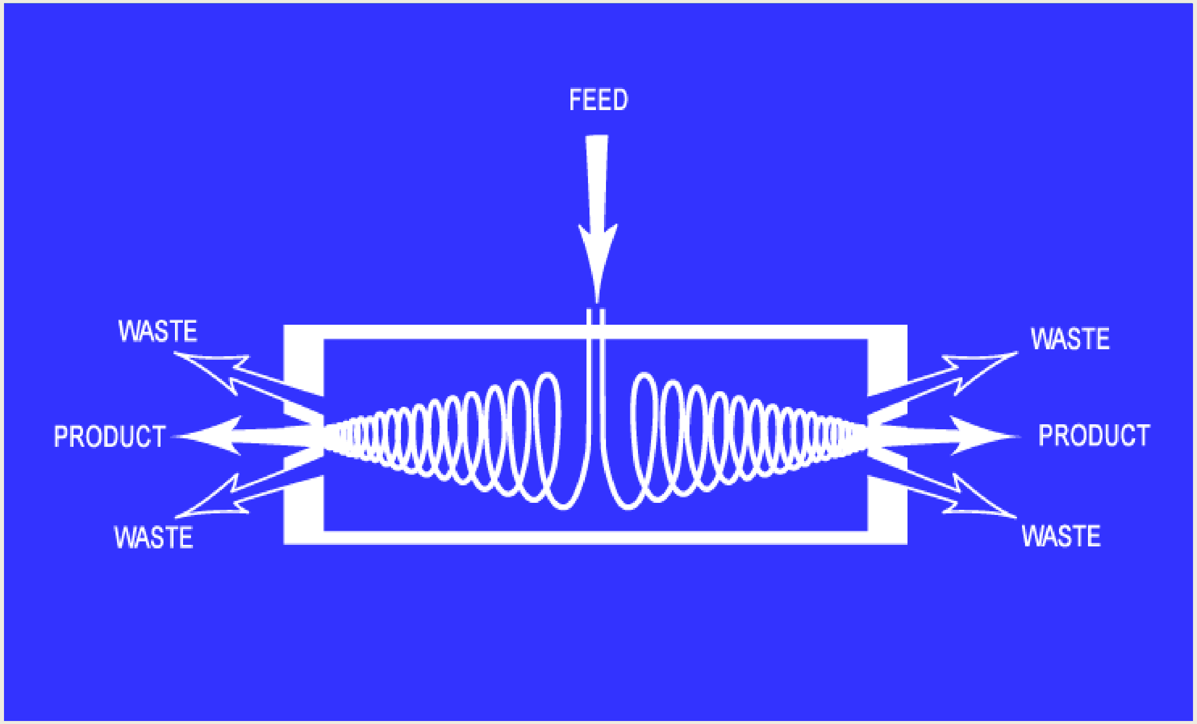
March, 1976:

Enrichment:	1.023
Cut:	50%



UCOR Vortex Process

- Enrichment is achieved under pressurized conditions by centrifugal means in a stationary-wall centrifuge



1975 – 1990:

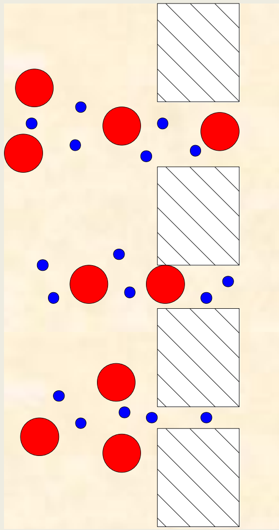
Enrichment: 1.03

Cut: 5%

NO PATENT

Diffusion theory

- Diffusion



$$\frac{\partial c}{\partial t} = D_x \frac{\partial^2 c}{\partial x^2}$$

- Diffusion through a porous medium:

$$v\frac{\partial c}{\partial x} + \frac{\partial c}{\partial t} = D_x \frac{\partial^2 c}{\partial x^2}$$

$$v\frac{\partial c}{\partial x} + F\frac{\partial Q}{\partial x} + \frac{\partial c}{\partial t} = D_x \frac{\partial^2 c}{\partial x^2}$$

$$c(x,t) = \frac{Ax}{2\sqrt{\pi D}(t-t_0)^{3/2}} \exp\left\{-\left(\frac{v}{2\sqrt{D}}\sqrt{t-t_0} - \frac{x}{2\sqrt{D}}\frac{1}{\sqrt{t-t_0}}\right)^2\right\}$$

Technology of Isosilicon – Reasons for silane, SiH_4

- The lighter the compounds to be separated, the larger the differences in diffusion coefficients. No stable molecule lighter than SiH_4 among those involving Si. (MW is 32 for SiH_4 vs. 104 for SiF_4). The relative isotopic difference of mass between ^{28}Si and ^{29}Si is 1/32 = 0.03125 in case of SiH_4 and 1/104 = 0.009615 in case of SiF_4.
- SiH_4 is now widely used in the electronics and ceramics industry.

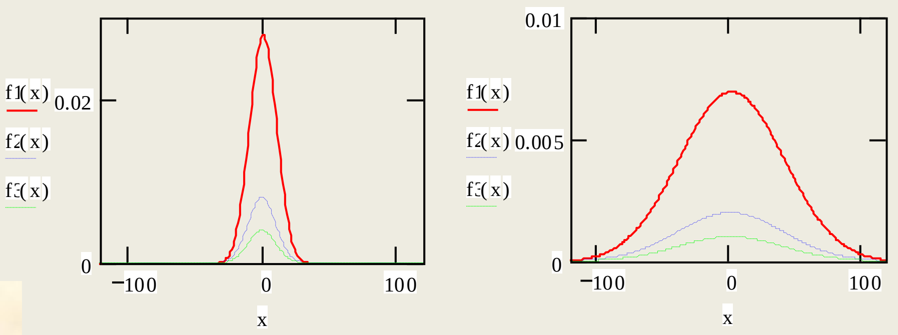
Technology of Isosilicon – Reasons for silane, SiH_4

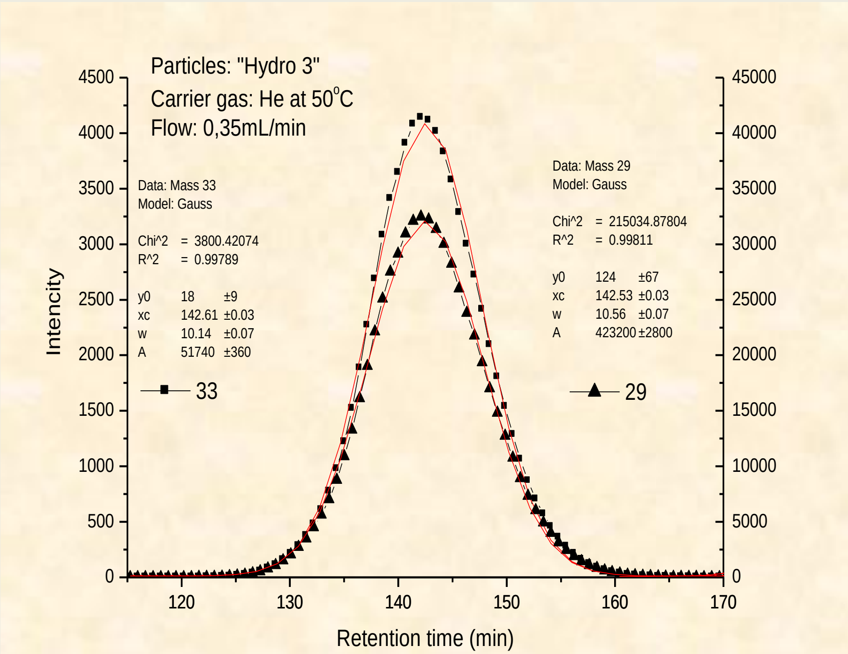
- The suitable isotope ^{28}Si is by far the most abundant (92,23%); ^{29}Si (4,67%) and ^{30}Si (3,10%). We may afford to "spoil" a large part of the feed of SiH_4 to increase the isotopic ratio. The remaining silane may be used for purposes requiring silane with enrichment of the two heavier isotopes.
- The impact of ^{2}H in natural hydrogen, i.e. 0.015%, will just add a small portion to the fractions of the heavier Si-isotopes.

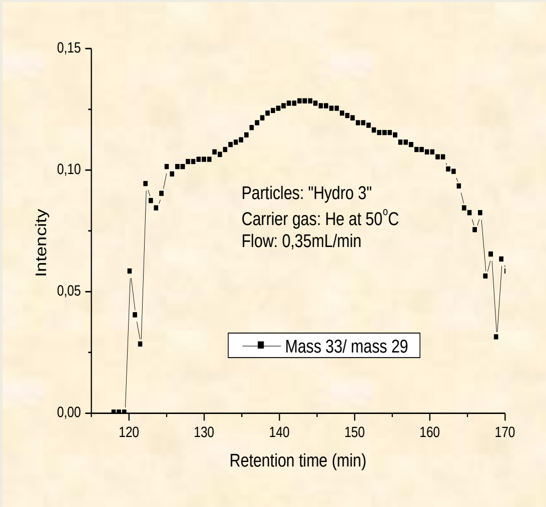
Isosilicon’s separation technology

- The basic idea that led to the first patent:

Experimental proof, based on diffusion through a column filled with monodisperse porous polymer particles:

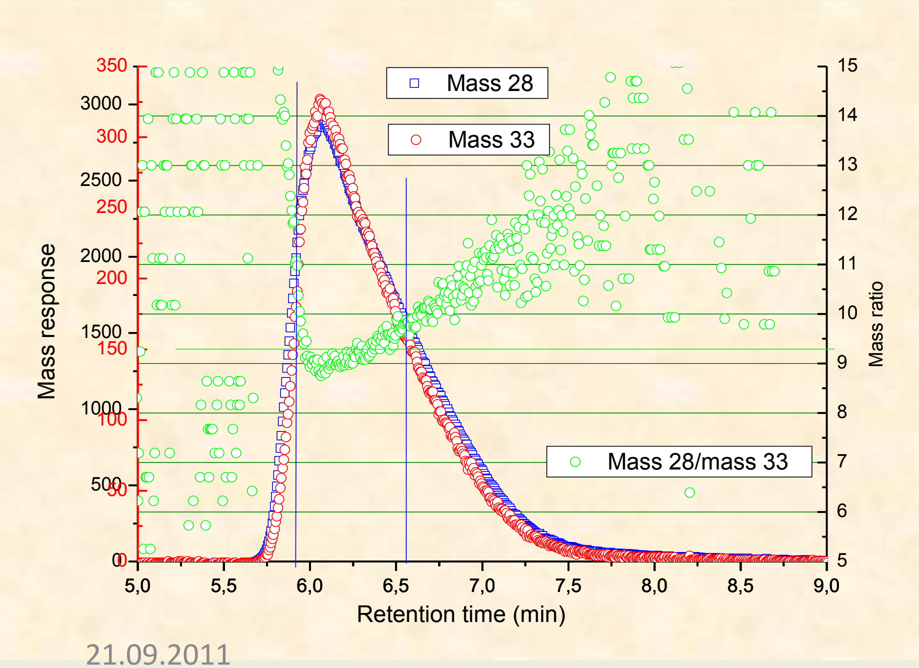


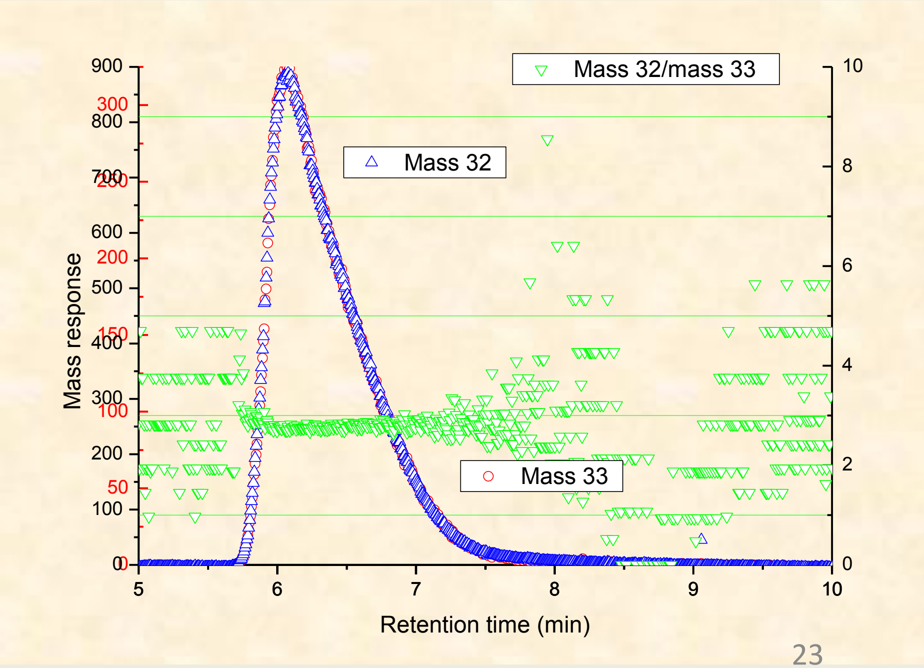




Isosilicon's separation technology

- The present technology is improved by Novasep SA, Nancy, France
- Smaller columns, higher flowrates, and less expensive absorbent, i.e. zeolite
- Patent appl.: WO 2010/018422 A1 "PROCESS FOR THE ENRICHMENT OF ISOTOPES"





Isosilicon's separation technology

(12) **United States Patent**
Eriksen et al.

(10) **Patent No.:** **US 7,309,377 B2**
(45) **Date of Patent:** **Dec. 18, 2007**

(54) **METHOD FOR SEPARATION OF ISOTOPES**

(75) Inventors: **Dag Ølstein Eriksen**, Oslo (NO); **Bruno Ceccaroli**, Kristiansand (NO)

(73) Assignee: **Isosilicon AS**, Kristiansand (NO)

4,780,116	A	*	10/1988	Cheh et al.	96/102
6,146,601	A	*	11/2000	Abesadze et al.	423/89
2003/0039865	A1		2/2003	Alexander et al.	

FOREIGN PATENT DOCUMENTS

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
18 February 2010 (18.02.2010)

PCT

(10) International Publication Number
WO 2010/018423 A1

(51) International Patent Classification:
B01D 59/26 (2006.01)

(21) International Application Number:
PCT/IB2008/003448

(22) International Filing Date:
28 October 2008 (28.10.2008)

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: **HILAIREAU, Pierre** [FR/FR]; Le Clos du Point du Jour, 23 chemin des Brigeottes, F-54130 Saint Max (FR). **MAJEWSKI, Wieslaw** [PL/FR]; 4, Terrasse des Vosges, F-54520 Laxou (FR).

(74) Agents: **POCHART, François** et al.; Cabinet Hirsh-

Isosilicon's separation technology

	$^{30}Si/^{28}Si$	2σ	$^{29}Si/^{28}Si$	2σ	Cut off (%)
Front	1.01513	0.00025	1.00784	0.00017	7.1
Tail	1.00573	0.00024	1.00291	0.00014	21.8

Proposed mechanism:
Isotopic exchange between gas and SiH_4 absorbed on the packing material

Number of stages needed:	60
Purity of ^{28}Si:	99.5%
Investment:	15 MUSD
Expected annual production:	5 000 kg
Expected price of $^{28}SiH_4$:	2 000 USD/kg

Possible other elements and isotopes that can be enriched by the technology

- Covered by patent: B_2H_6, NH_3, CH_4, C_2H_4, H_2O, H_2S, HCl, GaH_3, GeH_4 and Ge_2H_6, H_2Se, HBr, H_3Sb, SiH_4 , H_2Te, and UF_6
- Today markets exist for the following isotopes:
  - Non-nuclear applications: D, ^{13}C, ^{15}N, ^{18}O, $^{28,29,30}Si$
  - Nuclear applications: ^{2}H, ^{3}H, ^{6}Li, ^{7}Li, ^{10}B, ^{15}N, ^{28}Si, ^{91}Zr-depleted, ^{64}Zn-depleted, ^{235}U

Acknowledgements:

- Thanks to Research Council of Norway for financial support
- Thanks to the Franco-Norwegian Foundation for financial support
- Thanks to Novasep SA, Nancy, France for experimental support
- Thanks to Institute for Energy Technology for experimental support

Thank you for your attention!