Electronic Supporting Information for

Selection between separation alternatives: Membrane Flash Index (MFLI) to compare pervaporation and flash distillation

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Content

I Example calculation for Membrane Flash Index (MFLI)

Baseline data:

- organophilic pervaporation
- EtOH-Water binary mixture
- separation factor: $\alpha = 14$
- feed EtOH weight fraction: $x_{EtOH}^{F} = 0.015$

Mori, Y.; Inaba, T., Ethanol production from starch in a pervaporation membrane bioreactor using Clostridium thermohydrosulfuricum. *Biotechnology and Bioengineering* **1990,** 36, (8), 849-853.

- vapor equilibrium EtOH weight fraction: $y_{EtOH}^{D} NRTL = 0.093$

From ChemCAD program, VLE database: J. Gmehling et al.: Azeotropic data, VCH, 1994; DDB VLE data

1. Calculation of permeate weight fraction $\left(y_i^{PV}\right)$:

$$y_i^{PV} = \frac{\alpha * x_i^F}{(\alpha - 1) * x_i^F + 1} \quad \text{(S1)}$$

$$y_{EtOH}^{PV} = \frac{\alpha * x_{EtOH}^{F}}{(\alpha - 1)x_{EtOH}^{F} + 1} = \frac{14 * 0.015}{(14 - 1) * 0.015 + 1} = \frac{0.21}{1.195} = 0.176$$

Control calculation:

$x_{EtOH}^{F} = 0.015$

$x_{Water}^{F} = 1 - 0.015 = 0.985$

$y_{EtOH}^{PV} = 0.176$

$y_{Water}^{PV} = 1 - 0.176 = 0.824$

$$\alpha = \frac{y_i * x_j}{x_i * y_j} \quad \text{(S2)}$$

$$\alpha = \frac{y_{EtOH}^{PV} * x_{Water}^{F}}{x_{EtOH}^{F} * y_{Water}^{PV}} = \frac{0.176 * 0.985}{0.015 * 0.824} = \frac{0.1734}{0.0124} = 14$$

In the case of hydrophilic pervaporation **Eq. S1** is the following:

$$y_{Water}^{PV} = \frac{\alpha * x_{Water}^{F}}{(\alpha-1) * x_{Water}^{F} + 1} \tag{S3}$$

2. Calculation of Membrane Flash Index $(MFLI)$:

$$MFLI = \frac{y_i^{PV}}{y_i^{D} IT} \tag{S4}$$

$$MFLI = \frac{y_{EtOH}^{PV}}{y_{EtOH}^{D} IT} = \frac{0.176}{0.093} = 1.89$$

II Separation of methanol and water

II/1 Organophilic pervaporation

PDMS and hydrophobic zeolite membranes are evaluated in the case of organophilic methanol–water separation. **Table 1** and **Table 2** contain the MFLIs with regressed vapor equilibria, feed and calculated permeate weight fractions. **Fig. 1** and **Fig. 2** show the calculated permeate methanol weight fractions of OPV.

Table 1 Comparison of Membrane Flash Indexes in methanol–water organophilic pervaporation with PDMS membranes

	PDMS membranes	x_{MeOH}^{PV} [wf]	y_{MeOH}^{PV} [wf]	$y_{MeOH}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	PDMS/silica nanocomposite	0.04	0.49	0.19	2.60	Shirazi et al., 2012 [1]
2	PDMS copolymer	0.05	0.31	0.24	1.32	Guo and Hu, 1998 [2]
3	PDMS-ZIF-71 matrix 10:3	0.05	0.30	0.24	1.26	Y Li et al., 2014 [3]
4	PDMS - CA s.	0.05	0.27	0.24	1.15	Luo et al., 2008 [4]
5	PDMS - CA s. 16 µm	0.05	0.269	0.24	1.15	L Li et al., 2004 [5]
6	PDMS - CA s. 8 µm	0.05	0.25	0.24	1.08	L Li et al., 2004 [5]
7	PERVAP™ 1060	0.05	0.24	0.24	1.02	Kujawski et al., 2000 [6]
8	PERVAP™ 4060	0.20	0.56	0.61	0.92	Toth and Mizsey, 2015 [7]
9	PPMS - CA s.	0.05	0.21	0.24	0.90	Luo et al., 2008 [4]
10	PERVAP™ 1060	0.20	0.43	0.61	0.70	Molina et al., 2002 [8]

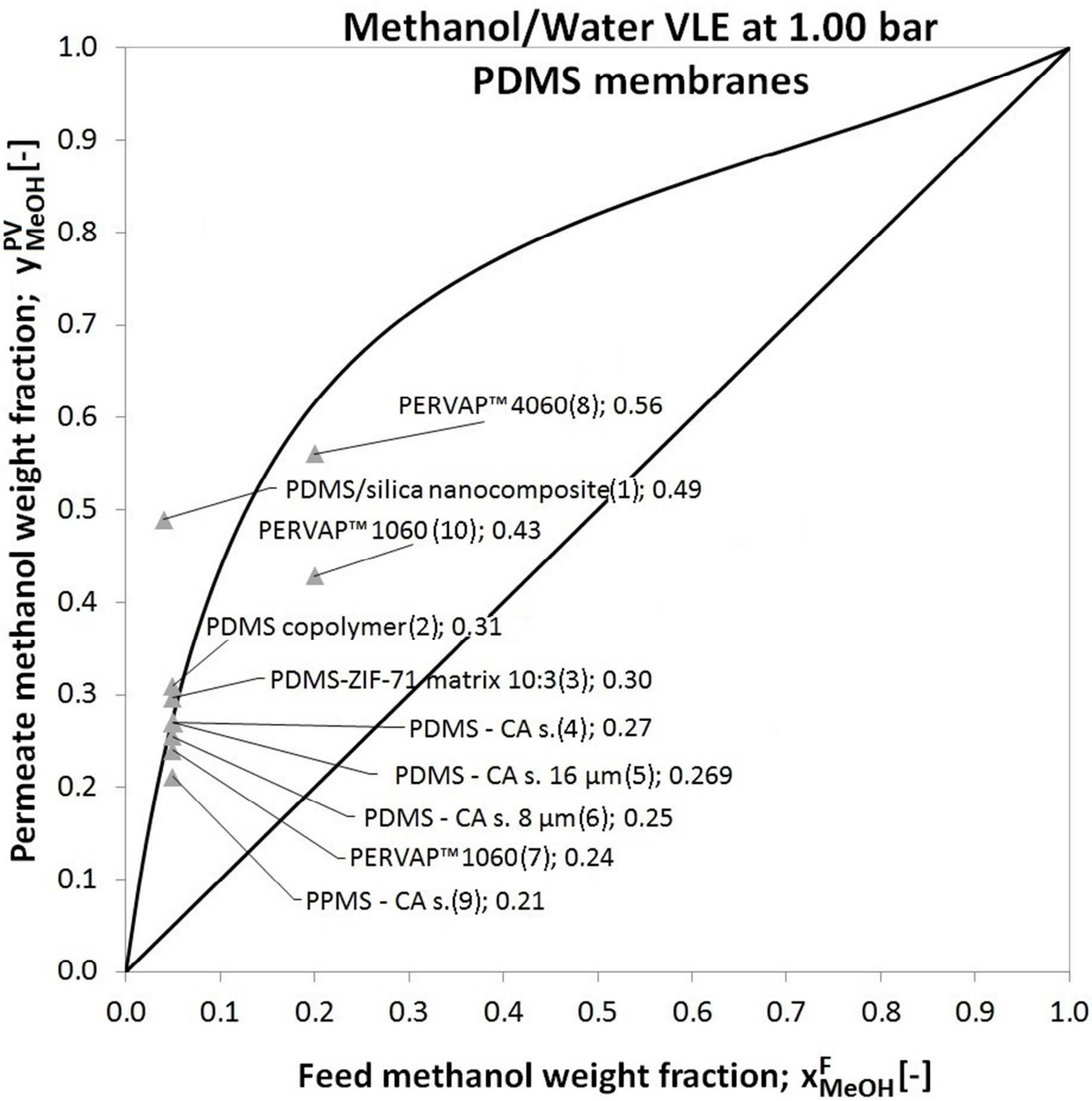

Fig. 1 Calculated permeate methanol weight fractions of organophilic pervaporation with PDMS membranes

As it can be seen, the MFLIs are close to 1, therefore PDMS membranes do not mean the good solution for methanol removal from water mixtures with OPV.

Table 2 Comparison of Membrane Flash Indexes in methanol–water organophilic pervaporation with hydrophobic zeolite membranes

	Hydrophobic zeolite membranes	x_{MeOH}^{PV} [wf]	y_{MeOH}^{PV} [wf]	$y_{MeOH}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	B-ZSM-5, SS s.	0.05	0.84	0.24	3.58	Tuan et al., 2002 [9]
2	Silicalite-1, SS s.	0.05	0.74	0.24	3.16	Tuan et al., 2002 [9]
3	Silicalite-1, SS s.	0.03	0.40	0.14	2.87	Chen et al., 2008 [10]
4	Ge-ZSM-5, SS s.	0.05	0.65	0.24	2.79	S Li et al., 2003 [11]
5	ZIF-71	0.05	0.53	0.24	2.25	Dong and Lin, 2013 [12]
6	Silicalite-1, SS s.	0.04	0.34	0.19	1.82	Liu et al., 1996 [13]
7	Crosslinked PBD	0.03	0.25	0.14	1.77	Yoshikawa et al., 1992 [14]
8	Zeolite-filled (60 m/m% silicone)	0.05	0.41	0.24	1.73	Hennepe et al., 1987 [15]
9	B-ZSM-5, α-alumina s.	0.05	0.39	0.24	1.65	Bowen et al., 2003 [16]
10	Silicalite-1, SS s.	0.05	0.37	0.24	1.56	Sano et al., 1994 [17]

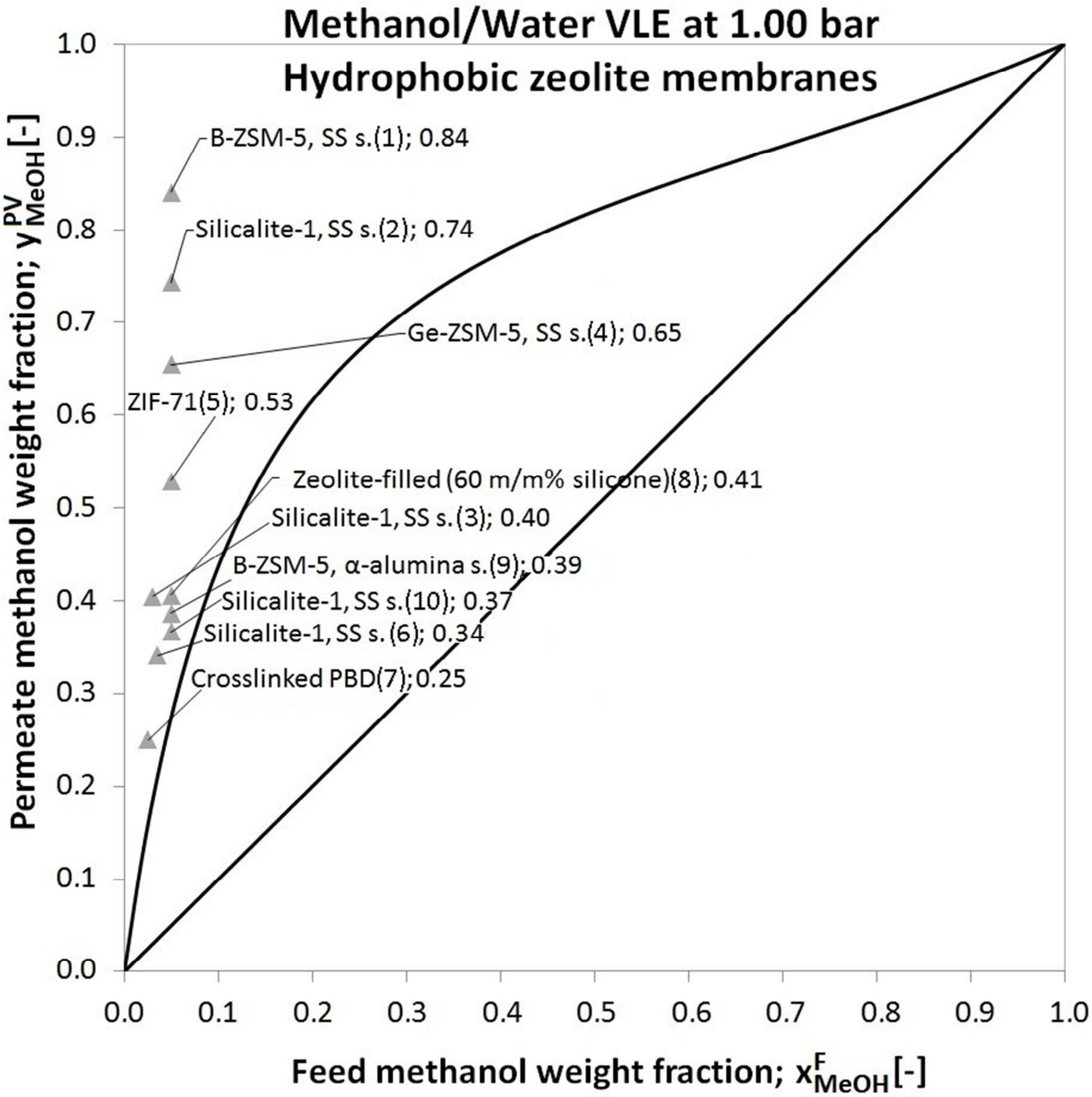

Fig. 2 Calculated permeate methanol weight fractions of organophilic pervaporation with hydrophobic zeolite membranes

Hydrophobic zeolite membranes have already significantly better efficiency than flash distillation (see **Fig. 2**), but considering the MFLIs of this group in **Table 2**, it can be seen that, they cannot reach breakthrough separation capability.

II/2 Hydrophilic pervaporation

Polyvinyl alcohol based membranes are the most utilized membranes in the case of dehydration of methanol mixtures with pervaporation. The Membrane Flash Indexes are found in **Table 3** and **Table 4**. **Fig. 3** shows the comparison of PVA membranes with flash distillation and **Fig. 4** depicts another hydrophilic membranes.

Table 3 Comparison of Membrane Flash Indexes in methanol–water hydrophilic pervaporation with PVA membranes

	Polyvinyl alcohol (PVA) membranes	x_{MeOH}^{PV} [wf]	y_{Water}^{PV} [wf]	$y_{Water}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Composite PVA/P(AA-co-AN/SiO_2)	0.90	0.96	0.04	24.17	Pang et al., 2006 [18]
2	PVA with 0.1% nano SiO_2	0.90	0.94	0.04	23.50	Bano et al., 2013 [19]
3	PVA with 0.125% nano SiO_2	0.98	0.19	0.01	22.55	Liu et al., 2008 [20]
4	PVA with 0.075% nano SiO_2	0.98	0.17	0.01	20.74	Liu et al., 2008 [20]
5	PVA with 0.05% nano SiO_2	0.98	0.15	0.01	18.30	Liu et al., 2008 [20]
6	PVA with 0% nano SiO_2	0.99	0.07	0.004	16.03	Sarkar et al., 2010 [21]
7	PVA with citric acid	0.90	0.51	0.04	12.82	Burshe et al., 1997 [22]
8	PVA by GFT	0.60	0.52	0.15	3.54	Wesslein et al., 1990 [23]
9	PVA grafted with hydrazine reacted SMA	0.75	0.23	0.09	2.44	Chiang and Chen, 1998 [24]
10	PVA-60°C	0.30	0.15	0.29	0.50	Shah et al., 2000 [25]

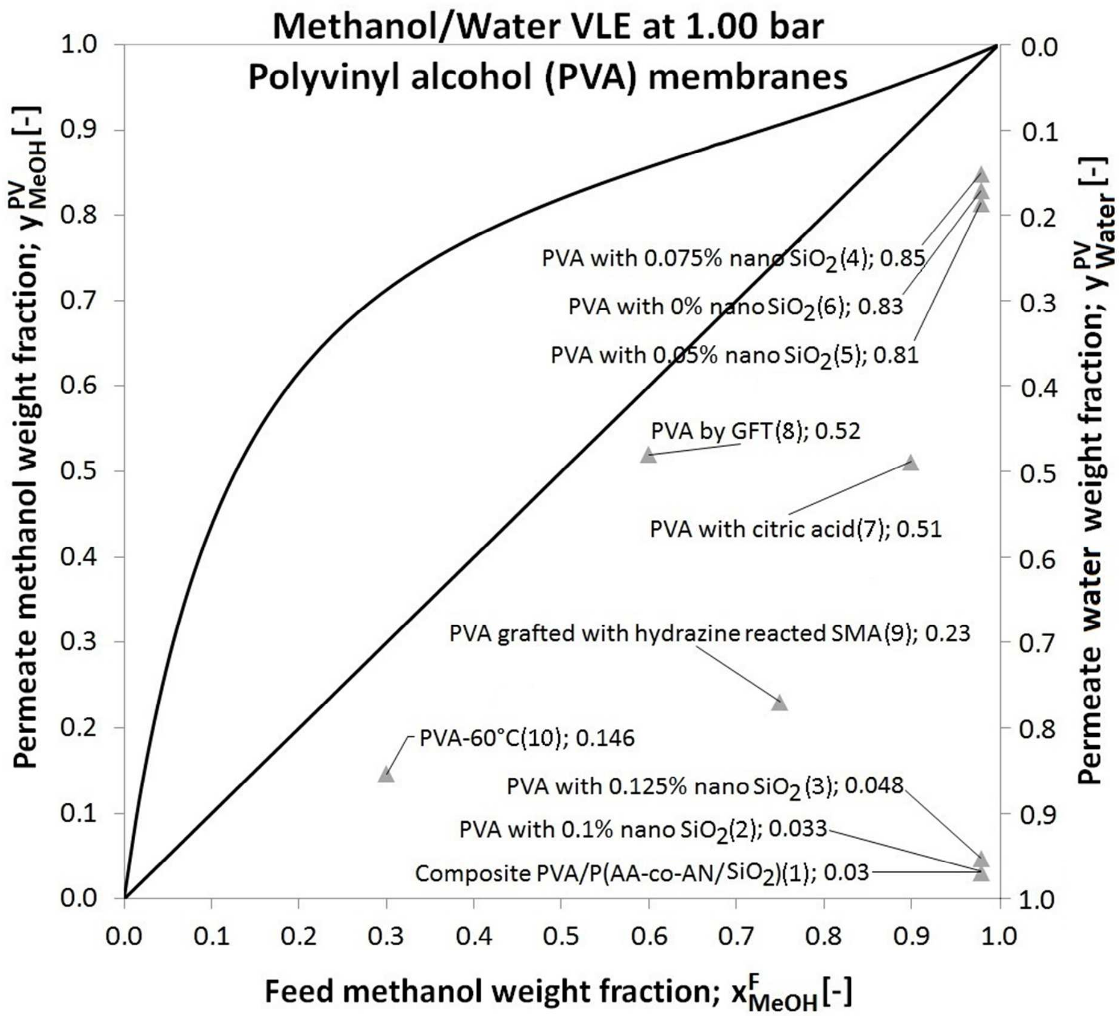

Fig. 3 Calculated permeate methanol weight fractions of hydrophilic pervaporation with PVA membranes

Table 4 Comparison of Membrane Flash Indexes in methanol–water hydrophilic pervaporation with other hydrophilic membranes

	Other hydrophilic membranes	x^{PV}_{MeOH} [wf]	y^{PV}_{Water} [wf]	$y^{D}_{Water}NRTL$ [wf]	MFLI [-]	Reference
1	T type zeolite (Mitsui)	0.90	0.996	0.04	24.96	Sommer and Melin, 2005 [26]
2	Polyamide-6	0.90	0.99	0.04	24.81	El-Gendi and Abdallah, 2013 [27]
3	Amorphous silica (ECN)	0.90	0.86	0.04	21.54	Sommer and Melin, 2005 [28]
4	Poly(Amidesulfonamide) PASA2	0.90	0.73	0.04	18.24	He et al., 2001 [29]
5	Crosslinked chitosan	0.84	0.97	0.06	15.49	Won et al., 2003 [30]
6	Chitosan	0.95	0.28	0.02	13.54	Won et al., 2002 [31]
7	5% sPPSU	0.85	0.66	0.06	11.14	Tang et al., 2012 [32]
8	Poly(Amidesulfonamide) PASA1	0.90	0.40	0.04	10.12	He et al., 2001 [29]
9	Tubular membr. Pervatech+silica	0.85	0.55	0.06	9.37	ten Elshof et al., 2003 [33]
10	PAI-PEI Hollow fiber	0.85	0.45	0.06	7.69	Wang et al., 2009 [34]

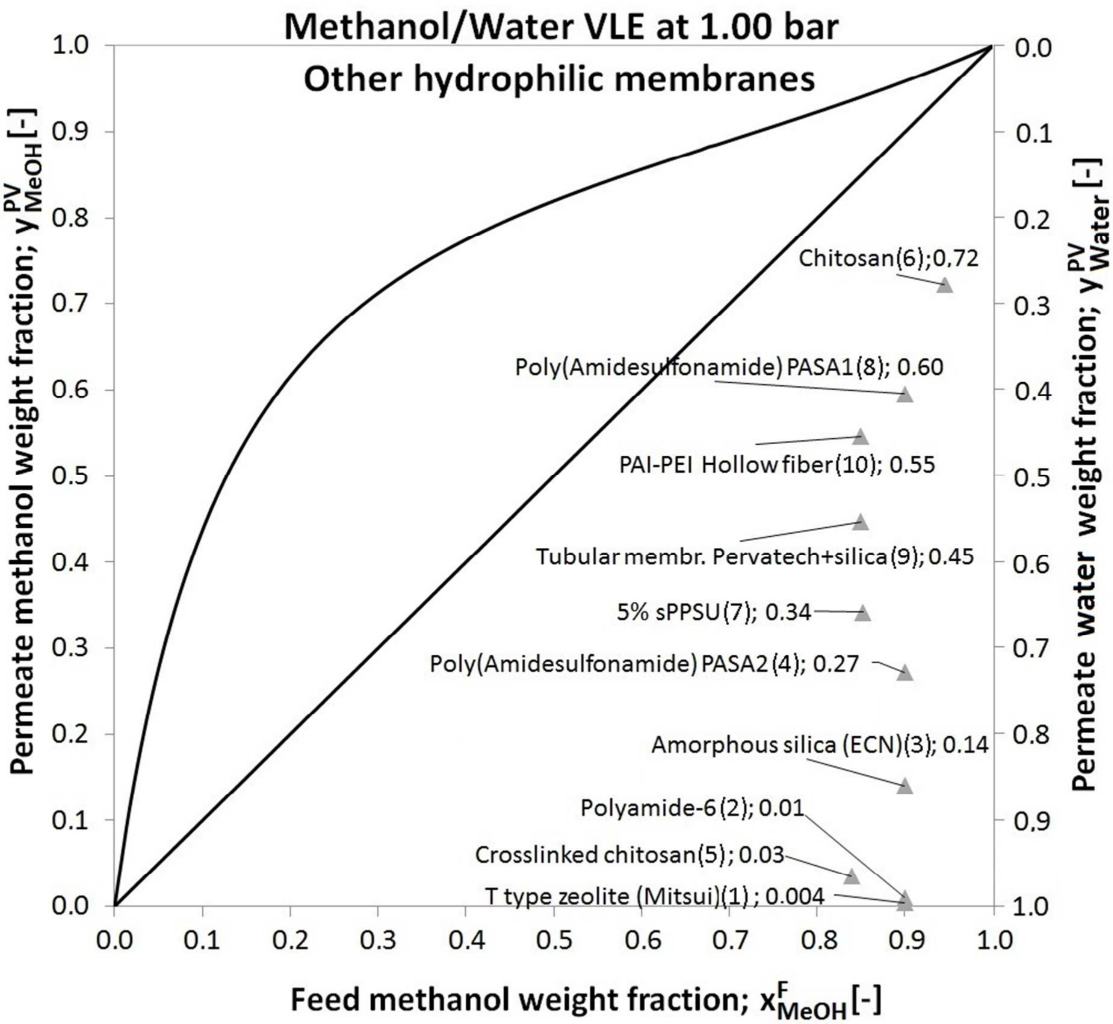

Fig. 4 Calculated permeate methanol weight fractions of hydrophilic pervaporation with other hydrophilic membranes

Table 3 and **Table 4** show that T type zeolite membrane from Mitsui has the highest Membrane Flash Indexes between methanol dehydration membranes.

III Separation of ethanol and water

Ethanol removal and dehydration pervaporation membranes are the most attractive in industrial application and research too.

III/1 Organophilic pervaporation

MFLIs of four different membrane types are evaluated in the case of organophilic separation. **Table 5** and **Fig. 5** show the characteristics of PDMS membranes, **Table 6** and **Fig. 6** interpret other polymeric membranes for ethanol removal from water. Hydrophobic zeolite types are found in **Table 7** and **Fig. 7**. Finally silicalite-silicone rubber mixed membranes are presented in **Table 8** and **Fig. 8**.

Table 5 Comparison of Membrane Flash Indexes in ethanol–water organophilic pervaporation with PDMS membranes

	PDMS membranes	x_{EtOH}^{PV} [wf]	y_{EtOH}^{PV} [wf]	$y_{EtOH}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Porous PTFE	0.02	0.18	0.09	1.89	Mori and Inaba, 1990 [35]
2	PDMS oil in porous s.	0.04	0.34	0.19	1.86	Kashiwagi et al., 1988 [36]
3	PDMS - PTFE s.	0.02	0.17	0.09	1.83	Zhang et al., 2009 [37]
4	PDMS	0.05	0.34	0.23	1.49	Slater et al., 1990 [38]
5	PDMS	0.02	0.14	0.09	1.47	Mori and Inaba, 1990 [35]
6	PDMS - CA s.	0.05	0.33	0.23	1.42	Luo et al., 2008 [4]
7	PDMS - PA s.	0.04	0.26	0.19	1.41	Shi et al., 2007 [39]
8	PDMS - CA s.	0.05	0.31	0.23	1.33	Li et al., 2004 [5]
9	GE 615 PDMS	0.06	0.36	0.28	1.31	Moermans et al., 2000 [40]
10	Cross-linked oligodimethylsiloxane	0.08	0.48	0.37	1.31	Ishihara and Matsui, 1987 [41]
11	MTR PDMS	0.06	0.33	0.28	1.18	Schmidt et al., 1997 [42]
12	PDMS	0.06	0.31	0.28	1.10	Mulder et al., 1986 [43]
13	PDMS	0.07	0.35	0.32	1.09	Jia et al., 1992 [44]
14	Fuji System PDMS	0.09	0.44	0.42	1.06	Nakao et al., 1987 [45]
15	Tisso Co Ltd PDMS	0.17	0.61	0.61	1.00	Takegami et al., 1992 [46]

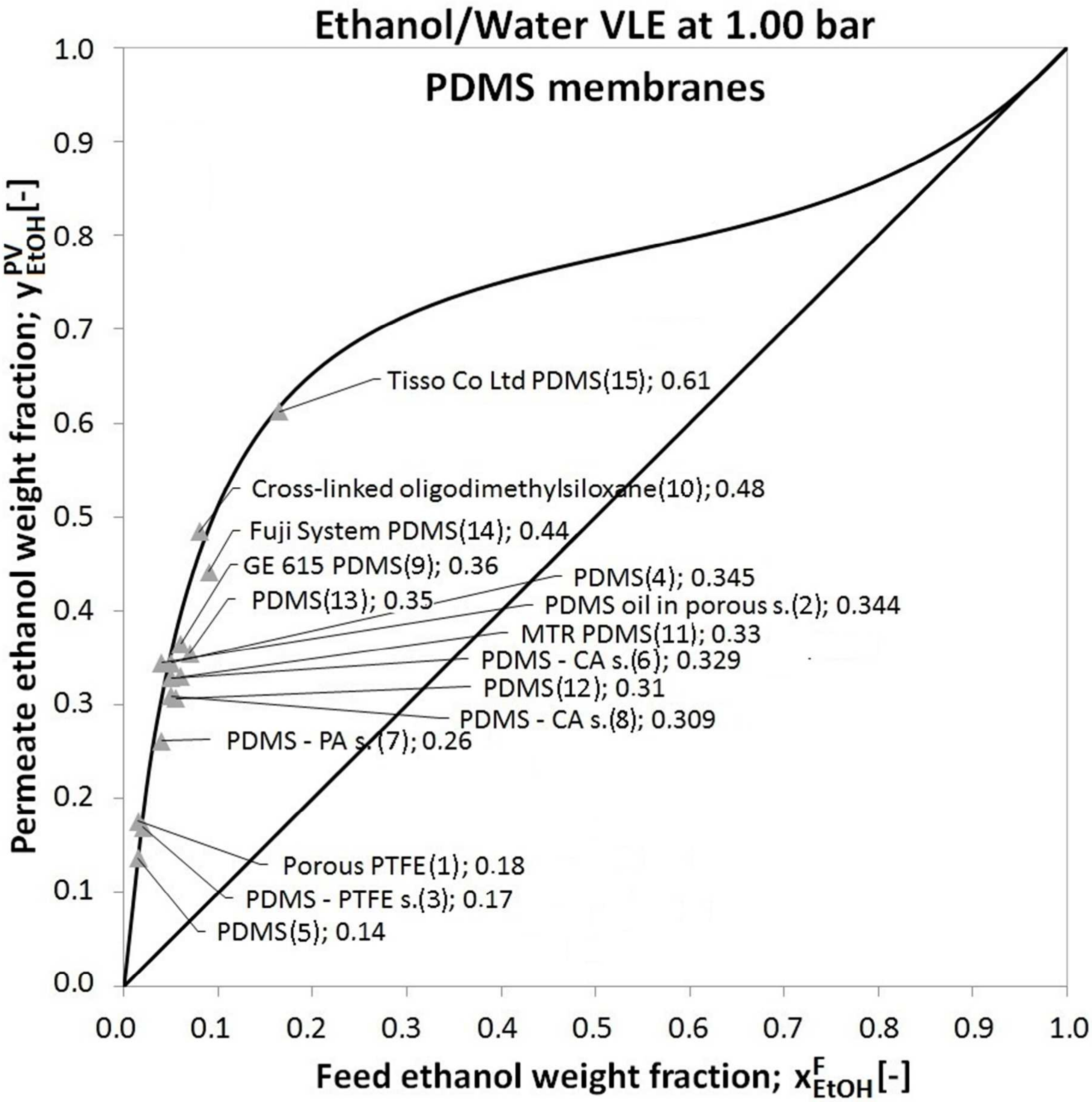

Fig. 5 Calculated permeate ethanol weight fractions of organophilic pervaporation with PDMS membranes

Table 6 Comparison of Membrane Flash Indexes in ethanol–water organophilic pervaporation with other polymeric membranes

	Other polymeric membranes	x_{EtOH}^{PV} [wf]	y_{EtOH}^{PV} [wf]	$y_{EtOH}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Copolymer of polysiloxane and phosphate ester	0.05	0.62	0.23	2.67	Chang et al., 2004 [47]
2	IPAA/FA-PDMS blend	0.03	0.34	0.14	2.41	Aoki et al., 1993 [48]
3	PTMSP	0.02	0.22	0.09	2.39	Mori et al., 1990 [35]
4	Plasma polymerized silane	0.04	0.43	0.19	2.31	Kashiwagi et al., 1988 [36]
5	30 µm thick PTMSP	0.06	0.62	0.28	2.24	Baker et al., 1997 [49]
6	Plasma polymerized silanes	0.04	0.41	0.19	2.23	Kashiwagi et al., 1988 [36]
7	Styrene-fluoroalkyl acrylate graft copolymer	0.08	0.80	0.37	2.16	Ishikara and Matsui, 1987 [41]
8	PTMSP	0.06	0.59	0.28	2.13	Schmidt et al., 1997 [42]
9	PTMSP/PDMS graft copolymer	0.07	0.68	0.32	2.10	Nagase et al., 1990 [50]
10	14–43 µm thick PTMSP	0.06	0.56	0.28	2.01	Volkov et al., 2004 [51]
11	PTMSP	0.06	0.56	0.28	2.01	Fadeev et al., 2003 [52]
12	10–20 µm thick PTMSP	0.06	0.55	0.28	1.98	Volkov et al., 1997 [53]
13	Phenyl propyne/PDMS graft copolymer	0.07	0.64	0.32	1.97	Nagase et al., 1989 [50]
14	n-Decane substituted PTMSP	0.06	0.53	0.28	1.91	Nagase et al., 1991 [54]
15	Trimethylsilyl substituted PTMSP	0.06	0.53	0.28	1.90	Nagase et al., 1991 [54]

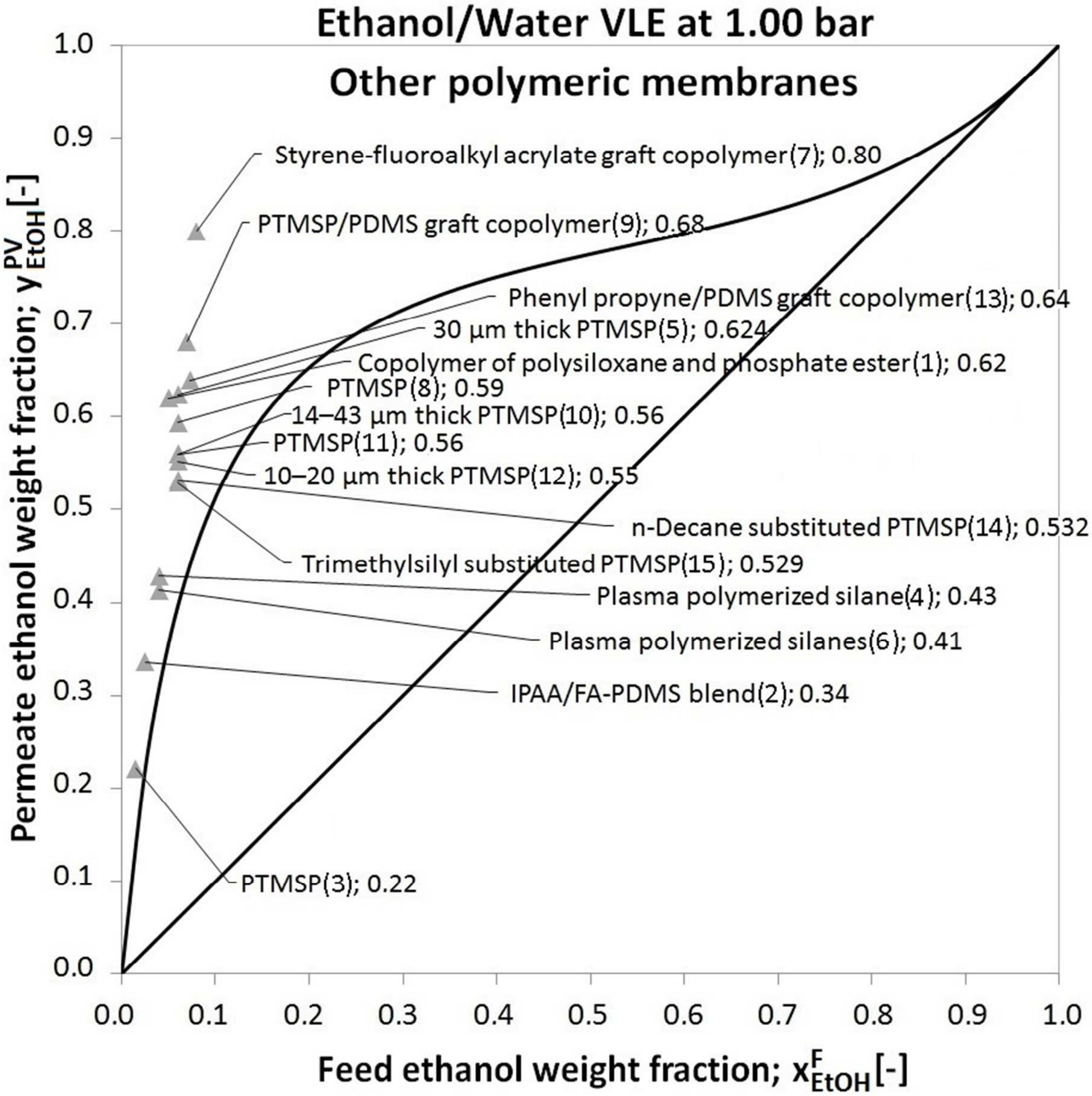

Fig. 6 Calculated permeate ethanol weight fractions of organophilic pervaporation with other polymeric membranes

The PDMS and other polymer membranes show the same picture and conclusion, comparing with methanol removal membranes (cf. **Table 1** and **Table 2** with **Table 5** and **Table 6**). The PTMSP types have the high MFLIs in the group of organophilic polymer membranes (see **Table 6**).

Table 7 Comparison of Membrane Flash Indexes in ethanol–water organophilic pervaporation with hydrophobic zeolite membranes

	Hydrophobic zeolite membranes	x_{EtOH}^{PV} [wf]	y_{EtOH}^{PV} [wf]	$y_{EtOH}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Silicaite-1 with PDMS coating - SS s.	0.04	0.84	0.19	5.52	Matsuda et al., 2002 [55]
2	Silicaite-1 - SS s.	0.04	0.714	0.19	3.85	Sano et al., 1994 [17]
3	Silicaite-1 - SS s.	0.04	0.711	0.19	2.83	Sano et al., 1997 [56]
4	Silicaite-1 - SS s.	0.04	0.68	0.19	3.67	Matsuda et al., 2002 [55]
5	Silicaite-1 - mullite porous s.	0.05	0.85	0.23	3.66	Lin et al., 2003 [57]
6	Silicaite-1 - alumina s.	0.05	0.82	0.19	3.55	Lin et al., 2003 [57]
7	Silicaite-1, silane treated - SS s.	0.04	0.65	0.19	3.52	Sano et al., 1995 [58]
8	Silicaite-1 - SS s.	0.05	0.64	0.23	3.43	Ikegami et al., 1997 [59]
9	Ge-ZSM-5 - SS s., Si/Ge=41	0.05	0.71	0.23	3.07	Li et al., 2003 [11]
10	PDMS - Silicalite-1	0.05	0.69	0.23	2.99	Vane et al., 2008 [60]
11	Silicaite-1 - SS s.	0.05	0.68	0.23	2.95	Nomura et al., 2002 [61]
12	Silicaite-1 - SS s.	0.04	0.54	0.19	2.90	Ikegami et al., 1997 [59]
13	B-ZSM-5 - alumina s.	0.05	0.62	0.23	2.67	Bowen et al., 2003 [16]
14	Silicaite-1 - mullite tubular s.	0.10	0.889	0.41	1.92	Lin et al., 2000 [62]
15	Silicaite-1 - SS s.	0.10	0.888	0.41	1.91	Ikegami et al., 2002 [63]

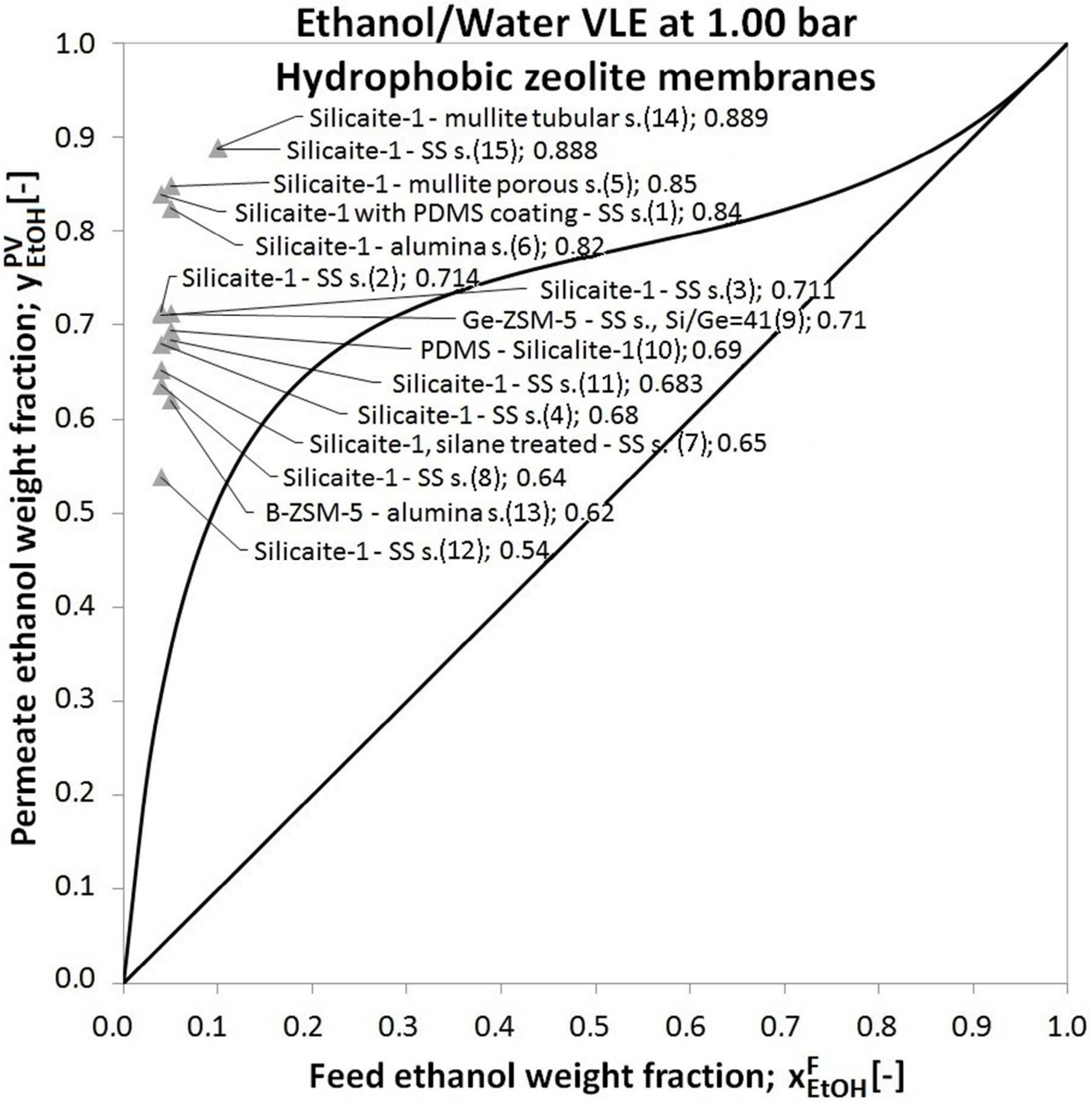

Fig. 7 Calculated permeate ethanol weight fractions of organophilic pervaporation with hydrophobic zeolite membranes

It can be seen that the hydrophobic zeolite membranes are slightly better than PTMSP types.

Table 8 Comparison of Membrane Flash Indexes in ethanol–water organophilic pervaporation with silicalite-silicone rubber mixed matrix membranes

	Silicalite-silicone rubber mixed matrix membranes	x_{EtOH}^{PV} [wf]	y_{EtOH}^{PV} [wf]	$y_{EtOH}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Silicalite particles treated with acid and steam	0.04	0.57	0.19	3.10	Chen et al., 1998 [64]
2	20 µm thick with microporous s.	0.05	0.64	0.23	2.77	Jia et al., 1992 [44]
3	125 µm thick	0.07	0.82	0.32	2.51	Jia et al., 1992 [44]
4	GE RTV615 PDMS	0.05	0.47	0.23	2.04	Adnadjevic et al., 1997 [65]
5	GE 615 PDMS	0.05	0.46	0.23	2.00	te Hennepe et al., 1987 [15]
6	GE 615 PDMS	0.05	0.44	0.23	1.90	te Hennepe et al., 1987 [15]
7	Nanoscale silicalite	0.06	0.50	0.28	1.80	Moermans et al., 2000 [40]
8	4–12 µm thick with microporous s.	0.07	0.55	0.32	1.68	Jia et al., 1992 [44]
9	Supported membrane	0.05	0.27	0.23	1.18	Liu et al., 1996 [13]
10	GFT composite membrane	0.06	0.31	0.28	1.11	Vankelecom et al., 1995 [66]

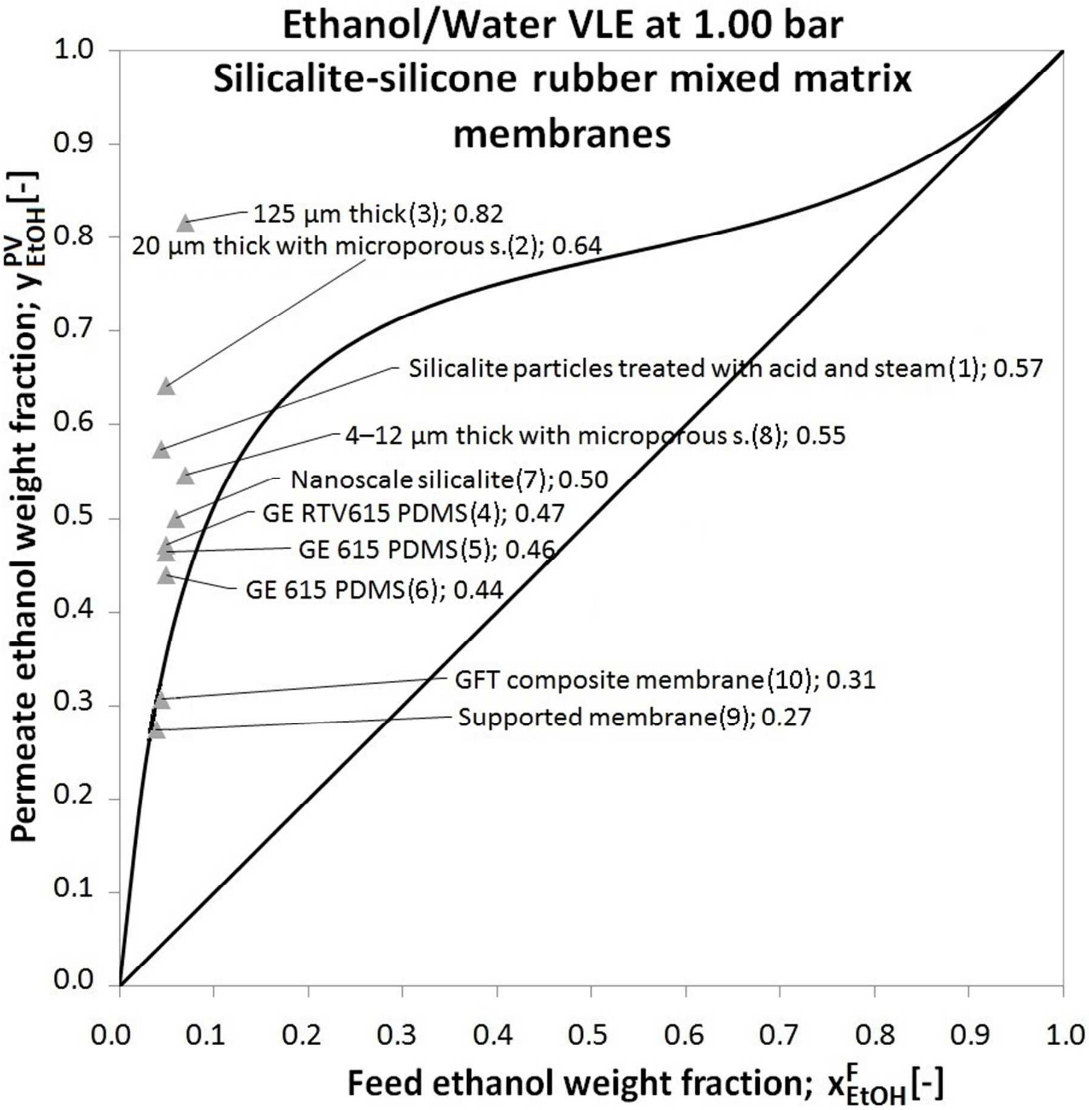

Fig. 8 Calculated permeate ethanol weight fractions of organophilic pervaporation with silicalite-silicone rubber mixed matrix membranes

III/2 Hydrophilic pervaporation

In the case of HPV, four different membrane groups are also represented. The characteristics of PVA membranes are found in **Table 9** and **Fig. 9**. **Table 10** and **Fig. 10** summarize specificities of the chitosan-based membranes. The further two classes are the Membranes containing charged groups (see **Table 11** and **Fig. 11**) and Membranes formed from polysalts (**Table 12** and **Fig. 12**) in our study.

Table 9 Comparison of Membrane Flash Indexes in ethanol–water hydrophilic pervaporation with PVA membranes

	Polyvinyl alcohol (PVA) membranes	x_{EtOH}^{PV} [wf]	y_{Water}^{PV} [wf]	$y_{Water}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	PVA-75°C	0.95	0.97	0.05	20.17	Sun and Zou, 2003 [67]
2	γ-aminopropyl-triethoxysilane	0.95	0.97	0.05	20.04	Zhang et al., 2007 [68]
3	Sulphated zirconia	0.95	0.93	0.05	19.35	Kim et al., 2001 [69]
4	PVA/GA containing PAA/EG IPNs	0.95	0.72	0.05	15.03	Ruckenstein and Liang, 1996 [70]
5	PVA with glutaraldehyde	0.90	0.95	0.09	11.01	Yeom et al., 2001 [71]
6	PVA by GFT	0.90	0.94	0.09	10.91	Wesslein et al., 1990 [72]
7	TEOS (130°C)	0.85	0.99	0.12	8.49	Uragami et al., 2002 [73]
8	TEOS (160°C)	0.85	0.98	0.12	8.40	Uragami et al., 2002 [73]
9	PEG blend and TEOS	0.85	0.98	0.12	8.39	Ye et al., 2007 [74]
10	Poly(acrylic acid) copolymer and TEOS	0.85	0.98	0.12	8.36	Uragami et al., 2005 [75]

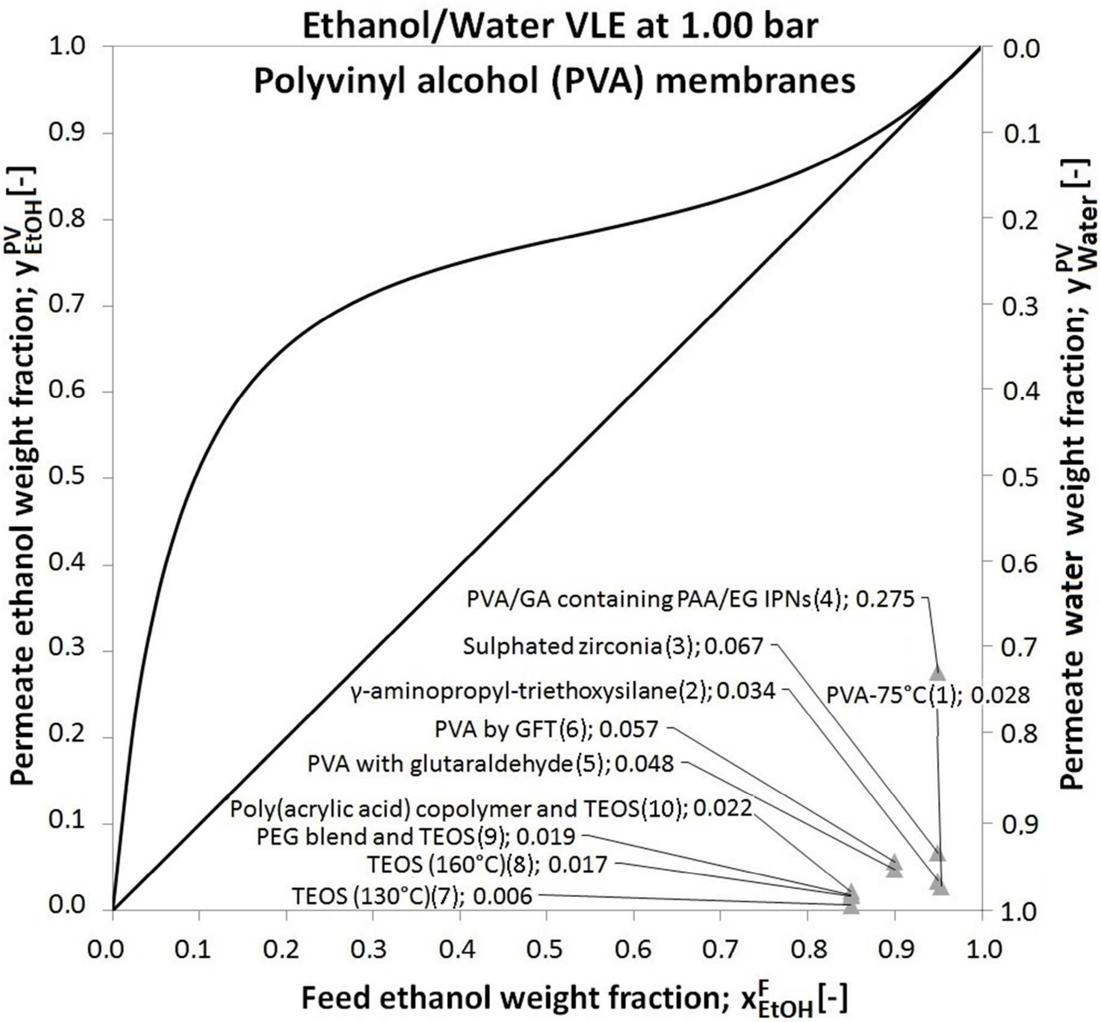

Fig. 9 Calculated permeate ethanol weight fractions of hydrophilic pervaporation with PVA membranes

Table 10 Comparison of Membrane Flash Indexes in ethanol–water hydrophilic pervaporation with chitosan-based membranes

	Chitosan-based membranes	x_{EtOH}^{PV} [wf]	y_{Water}^{PV} [wf]	$y_{Water}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Acetate salt	0.96	0.99	0.04	25.02	Uragami and Takigawa, 1990 [76]
2	GA crosslinked	0.96	0.99	0.04	25.01	Uragami and Takigawa, 1990 [76]
3	Uncrosslinked	0.96	0.89	0.04	22.60	Uragami and Takigawa, 1990 [76]
4	73% deacetylated	0.92	0.99	0.07	13.72	Maeda and Kai, 1991 [77]
5	Hydroxyethylcellulose 50% blend	0.90	0.999	0.09	11.55	Chanachai et al., 2000 [78]
6	Sulphonated & GA	0.90	0.99	0.09	11.49	Lee and Shin, 1991 [79]
7	Carboxymethylated	0.90	0.99	0.09	11.48	Lee and Shin, 1991 [79]
8	98% deacetylated-H_2SO_4	0.90	0.99	0.09	11.46	Maeda and Kai, 1991 [77]
9	98% deacetylated-HCl	0.90	0.99	0.09	11.39	Maeda and Kai, 1991 [77]
10	Phosphorylated	0.90	0.98	0.09	11.37	Lee and Shin, 1991 [79]

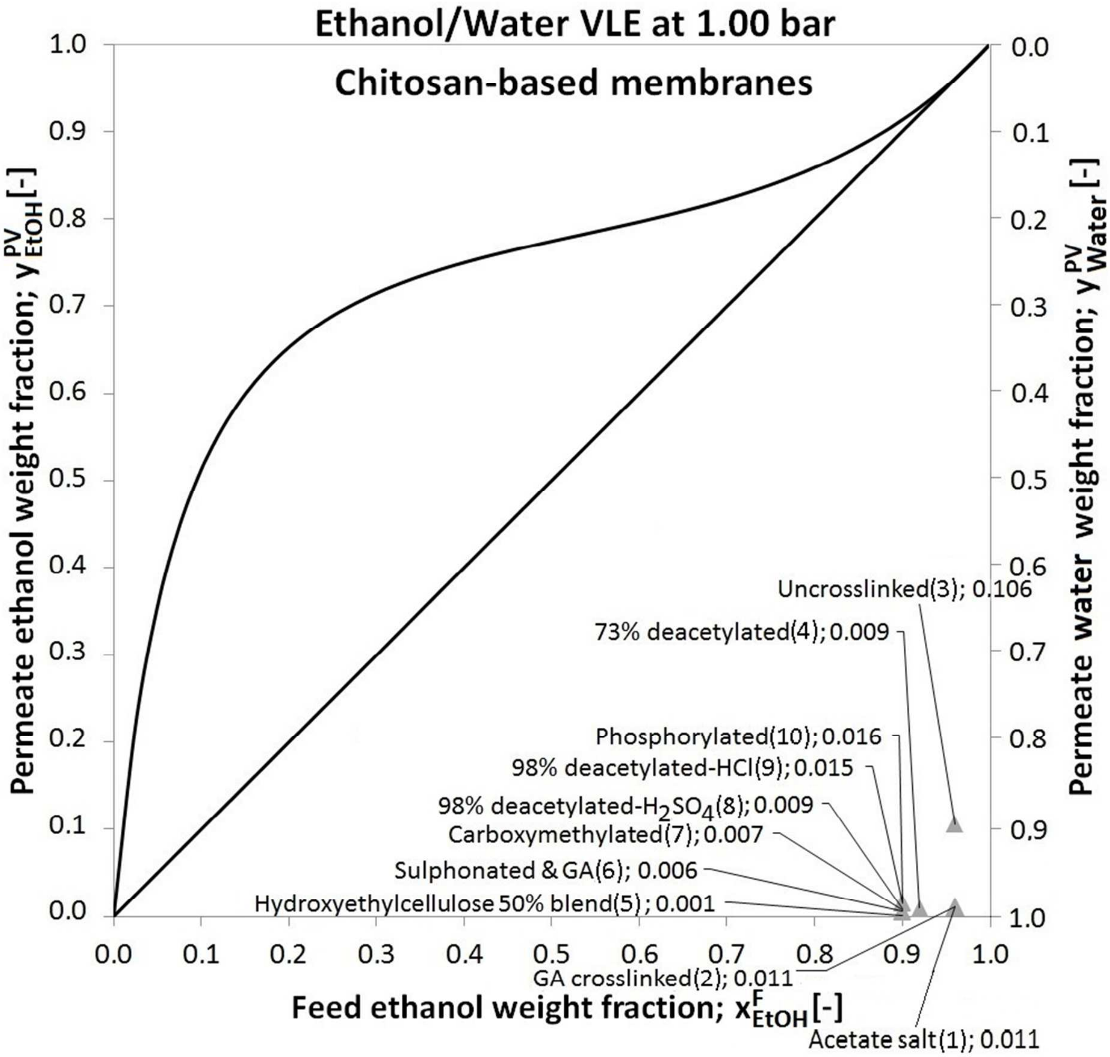

Fig. 10 Calculated permeate ethanol weight fractions of hydrophilic pervaporation with chitosan-based membranes

Table 11 Comparison of Membrane Flash Indexes in ethanol–water hydrophilic pervaporation with membranes containing charged groups

	Membranes containing charged groups	x_{EtOH}^{PV} [wf]	y_{Water}^{PV} [wf]	$y_{Water}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	Alg/DNA-Mg^{2+}	0.97	0.996	0.03	33.19	Uragami et al., 2015 [80]
2	Anionic PVA/GA	0.96	0.97	0.04	24.58	Praptowidodo, 2005 [81]
3	Cationic PVA/GA	0.96	0.97	0.04	24.46	Praptowidodo, 2005 [81]
4	PVA/GA	0.96	0.93	0.04	23.59	Praptowidodo, 2005 [81]
5	Cationic PVA	0.95	0.95	0.05	19.78	Sun and Zou et al., 2003 [67]
6	Anionic PVA	0.95	0.95	0.05	19.72	Sun and Zou et al., 2003 [67]
7	PVA/sericin blend	0.92	0.89	0.07	12.37	Gimenes et al., 2007 [82]
8	Rb^+ alginate	0.90	0.9992	0.09	11.55	Mochizuki et al., 1990 [83]
9	Li^+ alginate	0.90	0.9992	0.09	11.55	Mochizuki et al., 1990 [83]
10	Cs^+ alginate	0.90	0.9991	0.09	11.55	Mochizuki et al., 1990 [83]
11	PVA/9% acrylic acid graft	0.90	0.99	0.09	11.43	Semenova et al., 1997 [84]
12	2% NaA-Modified PASA2	0.90	0.99	0.09	11.43	He et al., 2001 [29]
13	Na^+ alginate-PVA blend	0.90	0.98	0.09	11.29	Dong et al., 2006 [85]
14	PVA/7 m/m% sulphosuccinic acid	0.90	0.95	0.09	11.00	Rhim et al., 1998 [86]
15	5% NaA-Modified PASA1	0.90	0.89	0.09	10.26	He et al., 2001 [29]

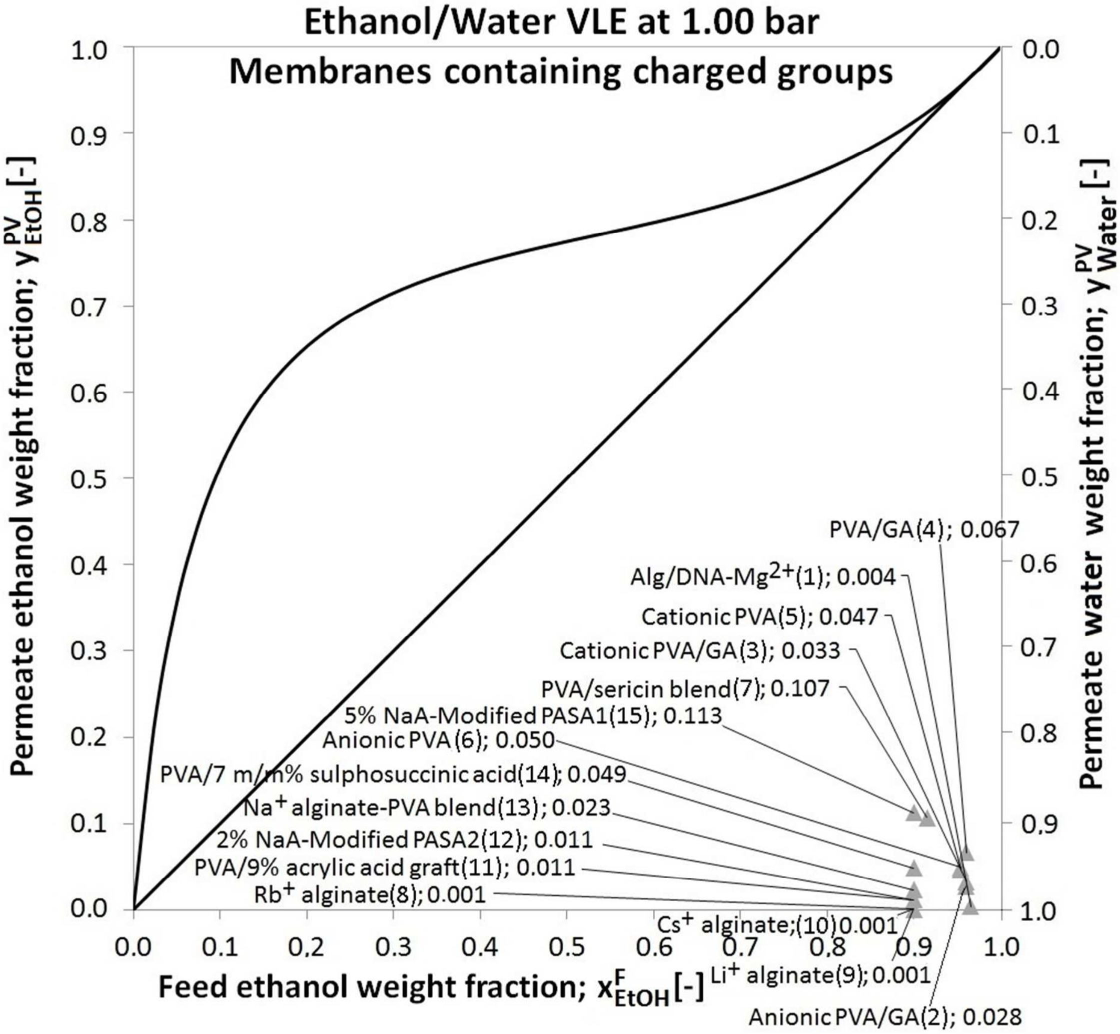

Fig. 11 Calculated permeate ethanol weight fractions of hydrophilic pervaporation with membranes containing charged groups

Table 12 Comparison of Membrane Flash Indexes in ethanol–water hydrophilic pervaporation with membranes formed from polysalts

	Membranes formed from polysalts	x^{PV}_{EtOH} [wf]	y^{PV}_{Water} [wf]	$y^{D}_{Water} NRTL$ [wf]	MFLI [-]	Reference
1	A: Anionic PVA, DS 2.3% - C: Cationic PVA, DS 2.9%	0.95	0.99	0.05	20.56	Sun and Zou, 2003 [67]
2	A: Na^+ polystyrene sulphonate - C: Polyallylamine	0.94	0.82	0.06	14.52	Krasemann and Tieke, 1998 [87]
3	A: Poly(acrylonitrile-co-acrylic acid) - C: Poly(acrylonitrile-co-vinyl pyridine)	0.90	0.998	0.09	11.54	Won et al., 1993 [88]
4	A: Na^+ CMC - C: Chitosan	0.90	0.99	0.09	11.46	Zhao et al., 2009 [89]
5	A: Na^+ CMC - C: N-ethyl-4-vinyl-pyridinium bromide	0.90	0.99	0.09	11.43	Jin et al., 2010 [90]
6	A: Cellulose-SO_3-Na^+ - C: Polyethyleneimine	0.84	0.98	0.12	8.05	Zhao et al., 2009 [89]
7	A: Cellulose-SO_3-Na^+ - C: PolyDADMAC, linear	0.84	0.96	0.12	7.90	Zhao et al., 2009 [89]
8	A: Cellulose-SO_3-Na^+ - C: PolyDADMAC, branched	0.84	0.96	0.12	7.86	Zhao et al., 2009 [89]
9	A: Aromatic polyamide sulphonate - C: Polyethyleneimine	0.80	0.79	0.14	5.60	Kirsh et al., 1996 [91]

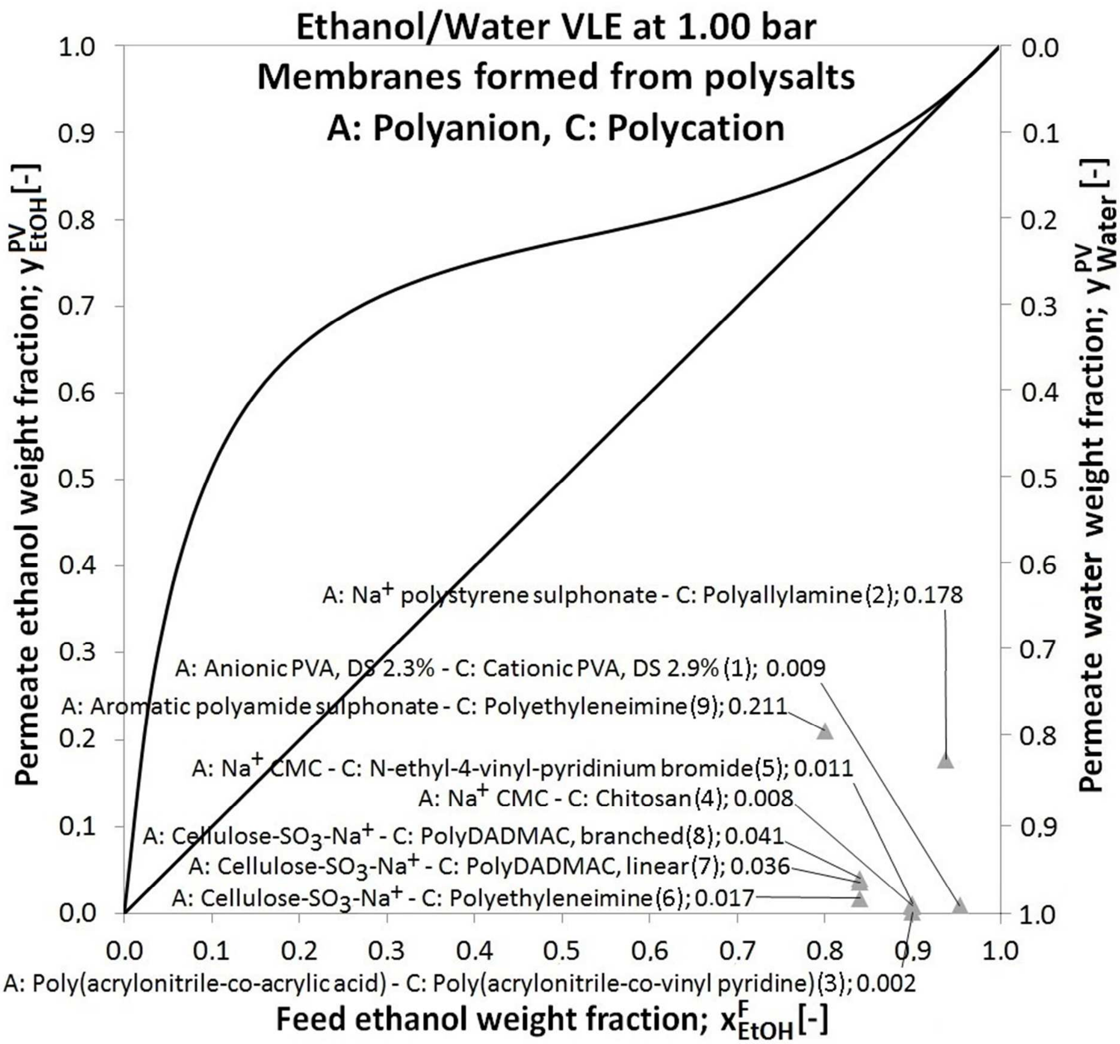

Fig. 12 Calculated permeate ethanol weight fractions of hydrophilic pervaporation with membranes formed from polysalts

Studying ethanol dehydration pervaporation membranes, it can be determined, there is no major difference in the MFLIs. The highest values from these hydrophilic membranes are calculated in the case of membranes containing charged groups.

IV Separation of isobutanol and water

Table 13 shows the comparison of MFLIs in the case of OPV and the hydrophilic membranes are interpreted in **Table 14**. Finally, **Fig. 13** depicts the permeate isobutanol weight fractions in the function of VLE.

Table 13 Comparison of Membrane Flash Indexes in isobutanol–water with organophilic membranes

	Organophilic membranes	x_{IBU}^{PV} [wf]	y_{IBU}^{PV} [wf]	$y_{IBU}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	$(TX\text{-}PDMS)_n$	0.01	0.36	0.04	9.75	Schnabel et al., 1998 [92]
2	$(T\text{-}PDMS)_n$	0.01	0.35	0.04	9.45	Schnabel et al., 1998 [92]
3	$(T\text{-}PDMS\text{-}T\text{-}BFH)_n$	0.01	0.34	0.04	9.18	Schnabel et al., 1998 [92]
4	$(IP\text{-}PDMS)_n$	0.01	0.32	0.04	8.85	Schnabel et al., 1998 [92]
5	$(TX\text{-}PDMS\text{-}T\text{-}BFCH)_n$	0.01	0.32	0.04	8.63	Schnabel et al., 1998 [92]
6	$(IP\text{-}PDMS\text{-}IP\text{-}BFCH)_n$	0.01	0.31	0.04	8.47	Schnabel et al., 1998 [92]
7	PDMS	0.01	0.29	0.04	7.94	Böddeker et al., 1990 [93]
8	silicalite-filled GFT-PDMS	0.05	0.73	0.18	4.00	Jonquieres and Fane, 1997 [94]
9	PERVAP™ 4060	0.01	0.12	0.04	3.15	Toth et al., 2015 [95]
10	PERVAP™ 4060	0.07	0.71	0.26	2.79	Toth et al., 2015 [95]

Table 14 Comparison of Membrane Flash Indexes in isobutanol–water with hydrophilic membranes

	Hydrophilic membranes	x_{IBU}^{PV} [wf]	y_{Water}^{PV} [wf]	$y_{Water}^{D} NRTL$ [wf]	MFLI [-]	Reference
1	PERVAP™ 1510 (PVA)	0.99	0.998	0.05	21.70	Toth et al., 2015 [95]
2	PERVAP™ 1510 (PVA)	0.99	0.97	0.05	21.15	Valentinyi et al., 2014 [96]
3	zeolite LTA, porous Al_2O_3	0.95	0.99	0.17	5.71	Huang et al., 2014 [97]
4	PERVAP™ 2510 (PVA)	0.95	0.95	0.17	5.47	Guo et al., 2004 [98]
5	Pervasiv hollow-fiber	0.96	0.78	0.15	5.21	Kujawski and Krajewski, 2004 [99]
6	zeolite TFN, PAN support	0.90	0.97	0.26	3.76	Fathizadeh et al., 2013 [100]
7	PAI/PEI dual-layer hollow fiber	0.85	0.9999	0.30	3.31	Wang et al., 2009 [34]
8	PERVAP™ 1510 (PVA)	0.85	0.98	0.30	3.24	Toth et al., 2015 [95]
9	PERVAP™ 2210 (PVA)	0.90	0.57	0.26	2.21	Omidali et al., 2014 [101]

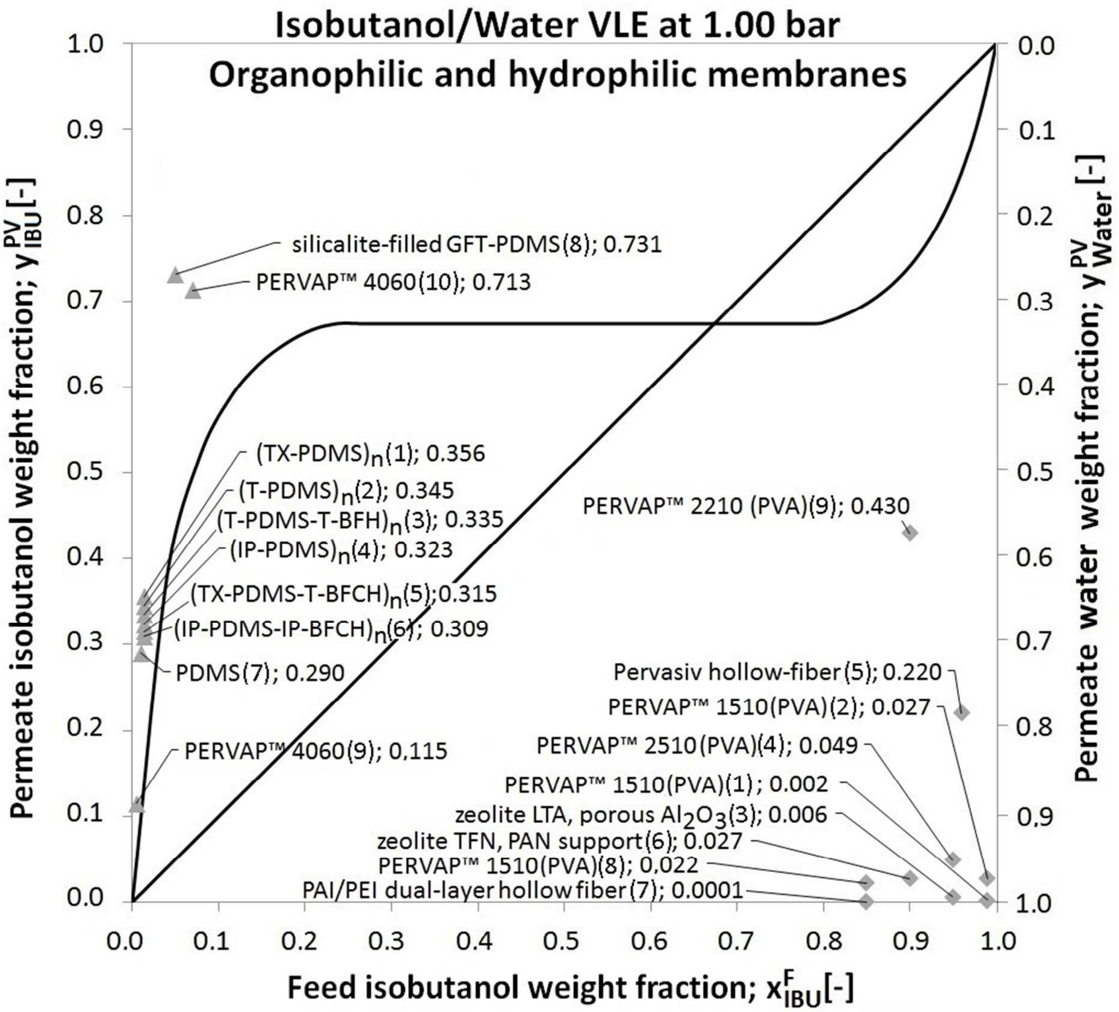

Fig. 13 Calculated permeate isobutanol weight fractions of organophilic and hydrophilic pervaporations

Nomenclature

F	Feed	
i	Component number	
j	Component number	
V	Vapour equilibrium	
x_i^F	Feed alcohol or water weight fraction	$[-]$
x_i^D	Equilibrium liquid alcohol or water weight fraction	$[-]$
y_i^D	Equilibrium vapour alcohol or water weight fraction	$[-]$
x_i^{PV}	Retentate alcohol or water weight fraction	$[-]$
y_i^{PV}	Permeate alcohol or water weight fraction	$[-]$

Abbreviations

CA	Cellulose acetate
EtOH	Ethanol
HPV	Hydrophilic pervaporation
hydr	hydrophilic
IBU	Isobutanol
IPAA/FA	Copoly(N-isopropylacrylamide/1H,1H,2H,2H-perfluorododecyl acrylate)
LTA	Linde Type A
MDMS	1,3-bis(3-aminopropyl) tetramethyldisiloxane
MeOH	Methanol
MFLI	Membrane Flash Index
NRTL	Mon-random two-liquid model
ODMS	α, ω -(bisaminopropyl) dimethylsiloxane oligomer
OPV	Organophilic pervaporation
org	organophilic

PAE	Polyamide-imide
PAN	Polyacrylonitrile
PASA	Poly(Amidesulfonamide)
PBD	Polybutadiene
PEBA	Polyether-block-amide
PEI	Polyetherimide
PDMS	Polydimethylsiloxane
PMDA	1,2,4,5-benzenetetracarboxylic dianhydride
PTFE	Polytetrafluoroethylene
PTMSP	Poly[1-(trimethylsilyl)-1-propyne]
PUR	Polyurethane
PVA	Polyvinyl alcohol
PV	Pervaporation
sPPSU	Sulfonated polyphenylsulfone
SS	stainless steel
TEOS	Tetraethoxysilane
TFN	Thin film Nanocomposite
VLE	Vapor-Liquid Equilibrium
wf	weight fraction

Greek letters

α	Separation factor

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