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초청강연

RECENT DEVELOPMENTS OF MEMBRANE TECHNOLOGY IN JAPAN

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Since the discovery of the Loeb-Sourirajan reverse osmosis membrane, thirty years have passed and many membrane technologies and new membranes for applications have been developed in the world. In the early stage of these developments Japan has not contributed much, but from the middle of 70ties Japan has started its own R&D projects starting from the desalination technology, and now various private industries and government ministries are actively engaging in R & D of membrane technologies in Japan.

In Table 1 the chronological developments of important events of developments and projects relating membrane technologies inside and outside of Japan are introduced and their details will be explained.

The first membrane technology applied in the Japanese industry was a electrodialysis(ED) process using ion-exchange membranes. These membranes were first developed in early 50ties and the Japanese government decided to use this method for concentration of sea-water to produce salt, which was then produced by solar evaporation. This development program started from 1960 by the Japan Monopoly Corp.(at that time). To apply ED process for sea-water concentration it was necessary to develop ion-exchange membranes having very low electric resistance to avoid energy loss due to Joule heat, and those having selectivity to permeate single valent ions only to avoid scale formation in the ED stacks. Three Japanese companies, Asahi Glass, Asahi Chemical and Tokuyama Soda, have succeeded to develop such membranes, and until 1971 all of the seven salt manufacturing companies had adopted ED for production of food salt.

Application of RO for desalination of lake water to produce industrial water for a steel manufacturer(Sumitomo Metal Ind.) at Kashima area started in 1971 using UOP's spiral modules. The initial size was 1,000m^3/day and eventually expanded to 13,400m^3/day within 9 years[1]. Development of domestic modules were started form 1977 in the R & D project of energy saving desalination technology(RO) conducted by WRPC. Modules that could desalt sea-water by a single stage were manufactured by Toyobo using hollow CA fiber, called Hollosep, and by Toray using spiral PEC-1000 composite membrane. Both modules had been tested in 800m^3/day unit located at Chigasaki[2,3]. This result led to produce one of the largest sea-water desalination unit in Saudi Arabia, whose specifications are given in Table 2.

Table 1. Chronological Table of Membrane Technology Developments

year	Desalination Conf.	Topics	Societies
1960		Loeb-Sourirajan	
1961		Binning(Pervap.)	
1962	1st Delyannis(Athens)		
1963			
1964		Spiral Module(UOP)	
1965	1st Symp.(Washington)	Permasep B-5	Soc. Sea-Water Sci., Japan
1966			"Desalination"(Elsevier)
1967	2nd Delyannis(Athens)	Findley(Memb. Dis.)	
1968			
1969			
1970	3rd Dely.(Dubrovnik)	Permasep B-9	
1971		N.S.(NS-100, NS-200)	
1972		DDS module	
1973	4th Dely.(Heidelberg)	Permasep B-10	
1974		Yuma project	Membrane Research Circle
1975			
1976	5th Dely.(Sardinia)	PA-300	"J.Memb.Sci.,"(Elsevier)
	1st IDEA(Mexico)		"Maku(Membrane)"
1977	2nd IDEA(Tokyo)		The Membrane Soc., Japan
1978	6th Dely.(Las Palmas)	Hollosep, PEC-1000	
		WF-21	
1979	3rd IDEA(Nice)	Prism Separator	
1980	7th Dely.(Amsterdam)	FT-30	30 years Anni. of the
			Soc. of Sea-Water Sci.
1981	4th IDEA(Bahrain)		
1982	WSIA(Honolulu)	Gore Memb. Dist.	The European Soc. Memb.
1983	1st IDA(Florence		
1984			1st Europe-Japan Congr.
			Memb.(Stresa)
1985	2nd IDA(Bermuda)		
1986		1st Pervap.	North American Memb.Soc.
1987	3rd IDA(Nice)	2nd Pervap.	ICOM '87(Tokyo)
1988		3rd Pervap.(Nancy)	
1989	4th IDA(Kuwait)	4th Pervap.(Florida)	
1990			ICOM '90(Chicago)
			The Memb. Soc. of Korea
1991	Europe(Malta), IDA (Washington)		Indian Memb.Soc.
1992			
1993			ICOM '93(Berlin)

Table 1. Continued

year	MITI and Desalination Projects
1960	(ED for salt production)
1961	
1962	
1963	
1964	
1965	
1966	
1967	
1968	
1969	MSF(Large Scale Proj. Ind.Tech.), Ind. Water in Kashima Area
1970	Tertiary Treatment of Sewage(Soc.Ind.Water)
1971	
1972	(Sumitomo Metal)
1973	Water Re-Use Promotion Center(WRPC)
1974	
1975	Desal. Tech. by Eng. Saving Method(R.O.)
1976	
1977	Started to use domestic module
1978	$13,400 m^3/d$
1979	Started to run $800 m^3/d$
	Sewage recovery by memb.
1980	C_1 Chem.(Large Scale Proj.Ind.Tech)
1981	Jisedai(New Membranes)
1982	Oxygen Sep.Res.Group
1983	phase I
1984	
1985	
1986	Aqua-Renaissance '90
	phase II; Alcohol dehydration(Pervap.)
1987	JIS for ultra-pure water
1988	
1989	phase III
1990	
1991	
1992	

Table 1. Continued

year	Project of Min. of Agri. & Fishery (MAF) and Min. of Pub. Health & Wel. (MPHW)
1960	
1961	
1962	
1963	
1964	
1965	
1966	
1967	
1968	
1969	
1970	
1971	
1972	R & D Projects of Japan Food Industry Center ↓ (1972–1980)
1973	Recovery of potato starch
1974	Conc. of orange juice by RO
1975	Production of maltose using membranes
1976	UF treatment of milk
1977	Desalting of soy sauce
1978	etc
1979	
1980	↓
1981	R & D Project of biomass conversion by MAF ↓ (1981–1990)
1982	Union of R & D of Memb. Tech for Food Processing
1983	phase I (1983–1984); Ass. of R & D of Poly. Memb. for Medical Appl. (1983–1987)
1984	
1985	
1986	phase II (1985–1987)
1987	
1988	Union of R & D of High Sep. Tech. for Food Processing
	Revision of Japan Pharma. reg. water for inject'n
1989	Memb. Res. Circle for Food Tech. ↓ (1989–1992)
1990	↓
1991	
1992	↓

Table 2. Jeddah I Rehabilitation Project, SWCC, KSA

Project Description	
Capacity(ton/day)	56,800
Module	Hollosep
Model	HM10255FI
Array	Parallel
Quantity(pc)	1,480
Salinity(ppm)	43,300
Pressure(kg/cm^2)	65-70
Temperature(℃)	35
Permeate	
TDS(ppm)	2,400
Chloride(ppm)	625
Construction	
Commission	Feb. 1989
Consultant : Bechtel	
Construction : MHI, Japan	

Recently in Japan the largest market of RO is pure water production for IC industry and for this purpose various low pressure membranes have been developed. Their specifications are shown in Table 3. Demands for qualities of ultrapure water are getting harder and harder, and following requirements are given from IC manufacturers.

1) Rejection of organic compounds should be large.
2) No dissolved substances from membranes and modules.
3) Sterilization by hydrogen peroxide can be applied.
4) Operation pressure should be low.

The first requirement is usually expressed by the rejection of IPA. It is seen from Table 3 IPA rejection is as high as 96%. The second requirement is expressed by time to reach low TOC after start-up.

Recent developments of various RO and UF membranes in Japan are surveyed in the literature.[4,5]

These low pressure RO membranes have also been used for R & D project of water recovery from municipal sewage effluents, which are other large water resources than the sea-water in water shortage areas and seasons. After testing various treatment schemes including UF and RO, WRPC selected low pressure hollow fiber and spiral modules and they had been tested combined with pretreatments, such as coagulation, sedimentation dual-media filtration, pH adjustment and safety filtration. Six years results show that these schemes can work steadily by adopting once-a-month chemical washing, and water cost estimate is lower than 100 yen/m^3.

Table 3. Rejection performance of thin film composite membranes[3]

Manufac.	Nittoh Elec. Ind.			Toray Ind.		
	759HR	739HF	729HF	SU700	SU600	SU200S
solutes						
NaCl	99.5	98	92	99.5	80	65
Na_2SO_4	99.9	99	99	99.9	—	99.7
$MgCl_2$	99.8	97	90	99.8	—	99.4
$MgSO_4$	99.9	99	99	99.9	99	99.7
Ethanol	53	40	25	54*	10	—
IPA	96	85	70	96*	35	17
Glucose	99.8	98	97	—	—	—
Sucrose	>99.9	99	99	99.8*	99	99
Test Condi.						
Conc.(%)	0.15	0.15	0.15	0.15	0.10	0.10
Press.(MPa)	1.5	1.5	1.5	1.5	1.5	0.75
Temp(℃)	25	25	25	25	25	25

* : Test Conc. is 0.10%

Another water re-use system is now being developed, and in called Aqua-Renaissance '90. This is a six year R & D project for water re-use and energy recovery supported by MITI. The objective is to develop low cost treatment produce reusable water from various industrial effluents and sewage.

Polymeric and ceramic membranes in capillary, hollow fiber, tubular and plate and frame modules have been tested in conjunction with bioreactors on a number of actual waste water and sewage streams. The total scheme and target is shown in Fig. 1 and Table 4[6].

Table 4. Target of Membrane Module Development

Operating Condition of Membrane (Micro-organism Conc., mg/l)	Power Consumption for Permeation Flux (kWH/m³)
10,000	1.5max
100	0.3max

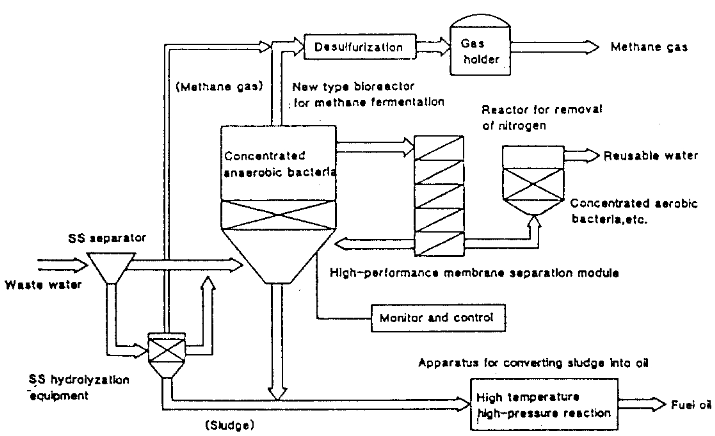

Fig 1. Conceptual Flow Chart of Aqua-Renaissance '90 Project

Jisedai is also MITI project and the objective is to develop basic technologies for future industries, such as new materials, biotechnology and electronic devices. The development of synthetic membranes for new separation technology is one of them. It's 10 years project will be terminated in 1990. Among various permselective membranes developed in this project, two pervaporation membranes that have very large separation factor for water permeation in ethanol-water system have been developed, and they are now transferred to another MITI project for alcohol dehydration. Here hollow fiber modules are now being developed and tested.

C_1 chemistry is a project to develop technology to use CO and H_2 as starting material to synthesize various hydrocarbons, such as ethylene and ethanol. Here membranes separating CO and H_2 were developed. One of them is a polyamide membranes separating CO and H_2 were developed. One of them is a polyamide membrane developed by Ube Ind. and now being commercialized. A similar project is now under planning stage. It is a CO_2 conversion project to reduce CO_2 emission to the atmosphere and separating membranes and conversing catalysts will be sought.

Applications of membrane technologies for food industries have been important subjects also in Japan, and R & D projects had been organized by the Japan Food Industry Center under the Ministry of Agriculture and Fishery. But speed of development was rather slow, because enough information of membranes was not given to food manufacturers, while membrane manufacturers could not understand real demands for memberane qualities in food industries. To solve this problem a union of R & D of membrane technology for food processing was organized and 18 pairs of membrane and food manufacturing co. engaged to work on each subject relating various food processing, and exchange their information on behaviors of membranes and modules. This union lasted for six years and now expanded to a "Membrane Research Circle for Food Technology", consisting about 100 people.

Regarding water for medical and pharmaceutical industries, RO, UF and MF membranes have been used for pure water production, which was also encouraged by the Ministry of Public Health and Welfare to reduce energy consumption. But it was not permitted to use them for water for injection. To study this problem the Ass. of R & D of Poly. Memb. for Medical Appl. started to work with the ministry and established validation methods. Following these lines the Japan Pharmacopoeia was revised and water treated by membranes could be used for injection.

During these discussions the Japanese Industrial Standard(JIS) was considered to be established and now membrane manufactures, various users and neutral scientists and engineers are working together for this purpose.

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