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Development and application of environmentally degradable plastics in Eastern Asian countries

Summary — General demographic and economical situation in Eastern Asian countries has been characterized. For several of those countries (Japan, South Korea, Taiwan, Singapore, China) the present state of plastic wastes utilization and environmentally degradable plastics (EDP) production has been described.
Key words: Eastern Asian countries, plastics wastes utilization, environmentally degradable plastics.

GENERAL SOCIAL AND ECONOMICAL ASPECTS OF EASTERN ASIAN COUNTRIES

In the Eastern Asian countries the economical development and industrialization are going very fast so the total consumption including plastics is rapidly increasing. However, the social infrastructure of transportation and waste treatment is weak, so it causes various problems. Furthermore, there are many giant cities with over several millions of people. In addition, there are big differences between the countries in: population and its density, economical development and income, petrochemical production and consumption as well as the infrastructure of waste treatment. Table 1 summarizes the demographic and some economical data of the Eastern Asian countries. Majority of countries are pretty dense inhabited, especially Singapore, Taiwan, Korea, and Japan. Japan and Singapore have high GNP/capita followed by Taiwan and Korea. Several countries including Japan, Korea, and China have the big petrochemical industries. The plastics consumption in Japan, Taiwan, Singapore, and Korea is over or near to 100 kg/capita · year, *i.e.* a value similar to the most consuming plastics

Table 1. Demographic and some economical data of Eastern Asian countries

Country	Population mill	Area 10^3 km^2	Population/km^2	GNP/capita '98, US$	Petroch. capacity*) mill. ton, '98 ('04)	Plastics consumption Kg/(y · p)	MSW**) production Kg/(d · p)	Waste treatment***) %	Regulation against plastics
China	1278	9600	133	826	4.4 (5.9)	20			high-medium
Indonesia	212	1919	110	662	0.5 (0.6)	7			
Japan	127	378	336	32350	7.3 (7.1)	115			high
North Korea	22	120	183	—	—	—	—	—	
Korea	47	99	475	8542	4.8 (5.6)	86	1.0	L 47, I 12, R 41	high
Malaysia	23	330	70	3247	0.6 (1.6)	55	0.6	L > 95	
Philippines	76	300	253	898	0 (0)	6			
Singapore	3	0.65	4615	30417	1.0 (1.9)	105	2.6	mainly by I	
Thailand	61	514	119	2036	1.2 (1.9)	39	0.6		
Taiwan	22	36	611	12166	1.0 (2.4)	135			medium
Vietnam	80	330	242	923	0 (0)	11			
Total	1951	13627			20.8 (27)				
World total	8000				103.3				

*) Based on ethylene; the data from MITI, Japan.
**) MSW = Municipal Solid Waste.
***) L = landfill, I = incineration, R = recycling.

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countries in the world. However, the municipal solid wastes (MSW) are mainly landfilled in the majority of countries. Only in Japan and Singapore the MSW are properly treated mainly by incineration.

The content of plastics waste in MWS rapidly increases as in the other countries in the world. The application of Environmentally Degradable Plastics (EDP) is one of the solutions in certain areas in addition to recycling. Although the mechanical properties and the processing abilities of EDPs have been improved recently the high prices of most of EDPs impede their wide extension worldwide.

The EDP demand in each Eastern Asian country can be divided into 3 groups based on the criteria of plastic consumption, GNP, population density, regulations, and composting infrastructure:

Group I: Japan, Singapore, Taiwan, and maybe Korea with its large industry;

Group II: China, Malaysia, Thailand, and maybe Indonesia with its big population;

Group III: Philippines, Vietnam, North Korea.

In the foregoing text the recent developments of EDP in several Eastern Asian countries are summarized. Those are taken from the national reports presented in the "ICS-UNIDO International Workshop on Environmentally Degradable Plastics: Industrial Development and Application" held on 19—22 September 2000 in Seoul, Korea. The authors are listed at the end of this work (see Acknowledgment).

The countries located in the western part of Asia, such as India, Iran and Middle-East Asian countries, were not included into the present article.

JAPAN

Japan is one of the most industrialized countries in the world having 130 millions of people and GNP per capita above 30 000 US$. Japan has got the 2nd largest petrochemical industry in the world and produces 14 million tons of plastics, consumes 115 kg/capita/year. The infrastructure in Japan is very advanced so more than 70% of MSW is properly treated by incineration. According to the 1998 data, the total plastics consumption is 10.2 million ton, among them about 9 million ton is utilized. Among them, over 4 million ton (44%) is recycled: mechanically 1.0 (12%) or with energy recovery either as a solid fuel (15%) or in power plants (17%). The rest, *ca.* 5 million ton (56%) is: incinerated (23%) or land-filled 3.2 million ton (33%). In another word, more than a half (55%) of the plastics waste is incinerated.

In addition, in Japan the regulations concerning the reducing of plastics packaging and extending the recycling become stronger. Japan developed various recycling technologies for MSW and plastics waste. The utilization of the tires used as a fuel for cement kiln or utilization of the plastics waste as a solid fuel are good examples.

Japan as one of the leading countries in the world is also very active in the application of EDPs and R&D on EDP. Such activity bases on Japanese strong academic and industrial R&D and is supported by large enough domestic market. The activity on EDP is lead by Japanese Biodegradable Plastics Society (JBPS). JBPS was founded on 1989 and has near 60 industry members. They have accomplished various projects such as: biodegradability field test of commercial EDPs, composting test of MSW with EDP trash bags and mulching film, development of the method for biodegradability determination and EDPs certification system. In addition, JBPS members have tried to publicize and educate people in the EDPs field (nickname "GreenPla").

Table 2. Commercial EDPs in Japan

Co.	Trade mark	Characteristics and capacity
A. Aliphatic polyesters and polylactide		
Daicel Chemicals	CELLGREEN PH	— Polycaprolactone — 1000 t/y (plan to 5000 t/y)
Showa High Polymers	BIONOLLE	— Poly(butylene succinate/adipate) — 3000 t/y (plan to 20 000 t/y)
Shimadzu Corp.	LACTY	— Polylactide — 100 t/y
Mitsui Chemicals	LACEA	— Poly(lactic acid) — 500 t/y
Mitsubishi Gas	IUPEC	— Poly(butylene succinate/carbonate)
Nihon Shokubai	LUNARE SE	— Poly(ethylene succinate)
Mitsubishi Gas	BIOGREEN	— PHB, microbial polyhydroxybutyrate
B. Starch-based and natural		
Nihon Corn Starch	CORNPOL	— Chemically modified starch
Nihon Gosei	MATER-BI	— from Novamont, Italy — Starch/PCL or cellulose
Daicel Chemicals	CELLGREEN PCA	— Cellulose acetate

In Table 2 EDP manufacturers in Japan are shown. The biggest one is Showa High Polymers Co. producing aliphatic polyester (3000 t/y) followed by Daicel Chemicals Co. producing polycaprolactone (1000 t/y) for years. In Mitsui Chemicals Co. a pilot plant of polylactide is operating by a unique condensation process in a solution/dispersion medium. Shimadzu Corp. is also developing polylactide but by conventional ring opening polymerization process. In addition, Mitsubishi Gas Co. and Nihon Shokubai Chemicals Co. are recently joining with different polyesters. Mitsubishi Gas Co. is also working on microbial polyester, polyhydroxybutyrate (PHB).

There are also several companies producing starch- or cellulose-based EDPs. Nihon Corn Starch Co. has a longer history dealing with starch and Daicel Chemicals added cellulose acetate to the list.

The EDP market in Japan on 1999 was about 2500 t, among them poly(butylenes succinate) 50%, polylactide 30%, starch-based material 10%, and others 10%. These materials are applied as trash bags (30%), in agriculture (30%), packaging (20%), and others (20%). JBPS expects the EDP market to grow to 20 000 t on 2003 and 100 000 t on 2010. Anyway the size of 100 000 t/y means only 1% of the total plastics demand in Japan.

KOREA

Korean petrochemical industry is the 4th largest in the world and produces 8 million tons of plastics while 4 million tons (~96 kg/capita/year) are consumed. Korea is very dense inhabited especially in the metropolitan area around Seoul. Korea has shown very fast economical development in a short time, but the infrastructure of the wastes treatment has not been invested the same time. Most (over 80% till 1994) of the MSW was land--filled. In order to solve the problem, the volume based collection fee (VCF) system was introduced on 1995. In this system, all the MSW should be discharged only in the designated trash bags, which prices include the treatment costs. Therefore, the more MSW is discharged the more is to pay, which is the first and unique system in the world. The VCF system brought a great effect in the various aspects; the amount of the total MSW decreased to 2/3 and twice more of materials are recycled. Korea continuously extends the regulations against plastic disposable and packaging materials. There is operating the Deposit Refund System for drink containers, bottles *etc.* and the Product Charge System for plastics *etc.*

There are substantial academic and industrial R&D activities on EDP in Korea too. Companies have been encouraged by an environmental issue related to VCF system and plastic waste and the R&D direction of Japan. There was a national project on EDP for the period of 1992—1996. Several EDP products based on aliphatic polyesters or starch/polycaprolactone blend were introduced to the market. But generally their prices are high which limits their application. As most of MSW is still landfilled, the government introduced the biodisintegrable trash bags, containing 30% of EDP, used for the VCF system — to reduce the amount of plastic buried and to create the EDP market. In 1999 Korean Biodegradable Plastics Association (KBPA) was founded by the EDP manufacturers. KBPA has carried out several project such as: standardization of EDP, burial test of the VCF trash bags containing 30% EDP, and composting test of food waste in EDP bags. Table 3 presents the EDP manufacturers in Korea. SK Chemicals Co. is a pioneer in developing the aliphatic polyester and is followed by Saehan Industry. Ire Chemical Co. has joined later but is very active and invests at a new plant producing 3000 t/y of aliphatic and aliphatic/aromatic polyesters. Several companies have worked on PHB but their activities seem to be rather declined recently. In addition, biodegradable medical suture made of polyglycolide or its copolymer has been commercialized by Samyang Co. and Korean Institute of Science and Technology. Samyang Genex was the first one introducing a starch foam for the application to loose-fill, but withdrew off this area. Daesang Corp., SK Corp. and Hanwha Petrochemical Co. are also producing the modified starch.

T a b l e 3. **Commercial EDPs in Korea**

Co.	Trade mark	Characteristics and capacity
A. Aliphatic polyesters		
SK Chemicals	SKYGREEN	— PET manufacturer — poly(butylene succinate/adipate) — PET facility, ~200t/y
Saehan Ind.	ESLON GREEN	— similar to above
Ire Chemical	EnPol	— aliphatic/aromatic polyesters — 3000 t/y
Samsung Color Ind.	KOMAGREEN	— aliphatic/aromatic polyesters in development
Kohap Ltd.		— PHB pilot plant study
B. Starch-based or starch/polycaprolactone		
SK Corp.	GREENPOL	— oil, naphtha, plastics maker — PCL/starch blend
Hanwha Petrochemical	ECO-PLAST	— naphtha, plastics maker — PCL/starch blend
Daesang Corp.	BIONYL REGREEN	— starch producer — PCL/starch blend — modified starch foam for loose fill
Samyang Genex	BIOFIL	— starch producer — modified starch foam for loose fill

The present Korean EDP market is around 250 t/y. This is 1/10 of the Japanese one, but it is usual ratio in other cases when comparing Korea with Japan. Most of EDP is applied at the moment to the biodisintegrable trash bags as explained above. If the biodisintegrable trash bag extends in all countries, the EDP market will reach about 6000 ton. In addition, there are other potential new markets for EDP such as the packaging material for lunch-boxes (for keeping rice warm or instant noodle) to replace expanded polystyrene. The lunch-box made of expanded polystyrene has been already banned officially. Food waste content is the highest in MSW and is going to be banned for landfilling since 2003. The separate collection and composting of food waste is already preparing. Therefore, large quantities of the trash bags made of EDP for food waste composting will be needed.

TAIWAN

Taiwan is the one of the most dense inhabited countries in the world. The GNP is high above 12000 US$ and the plastics consumption, maybe including export, exceeds 135 kg/capita/year. The country is also trying to reduce MSW amounts, especially plastic and packaging wastes and to extend their recycling. Plastics and PET bottles are charged with recycling fee. The government has set up a master plan for the whole recycling system called as Four-in-one (1997) in which the producers, distributors, consumers and recycling companies are operating together to recycle the materials. The regulation against plastic and packaging wastes is continuously strengthened but EDP is encouraged.

The R&D on EDP in Taiwan also began with a starch--filled or photodegradable plastics. Such biodisintegrable plastics are produced while R&D on actually biodegradable EDP is not so extended, especially in the industrial sector. However, EDP product based on modified starch is on the market. Weimon Industry Co. is most active to process mulching films and packaging materials made of modified starch from Novamant Co., Italy. They expect the EDP market to grow to 1000 t/year on 2003.

SINGAPORE

Singapore is the most dense inhabited city country in the world — have a population density above 4600 capita/km^2. There is very high GNP there, more than 30 000 US$ and the plastic consumption is over 100 kg/capita/year. Singapore produces 7800 t/day wastes totally, among them 3700 t/day of MSW, which means 1.3 kg/capita/day. MSW is very properly treated almost by incineration. The regulation against plastic wastes and packaging is initiated recently. There are very few R&D organization in Singapore and therefore the activity on EDP is low. However, it can be assumed Singapore has a big potential EDP market in the near future, based on the following reasons: high plastic consumption, high GNP (people can afford expensive EDP products), highly educated and organized society, ease to control the regulations in a small area.

CHINA

China is a huge country with *ca.* 1.3 billion of people and has tens of very big over-crowded cities. GNP is still not high and the petrochemical capacity is not enough for the domestic demand. Lots of plastics are imported from Korea and other countries. The average consumption/capita of plastics is low but actually pretty large in those big cities. Furthermore, China is rapidly developing nowadays and therefore the consumption is increasing very fast. However, the infrastructure of transportation or waste treatment is still primitive. For a couple of years some plastic disposables such as lunch-boxes and cups made of polystyrene foam have been banned in the certain big cities.

Table 4. Commercial EDPs in China

Co.	Trade mark	Characteristics
Tianjin Dawnhalo Appl.	STARATE	— modified starch based
Jilin Goldeagle Ind.		— modified starch/starch--filled, photodegradable
Nanjing Suchi		— similar to above

China has a great number of universities and institutes and belonging scientists active in all the fields. China presents in the academic sectors the substantial R&D activity on EDP, especially aliphatic polyesters, microbial polyesters and modified starch. However, the industrial activity is in the initial stage. Table 4 shows the EDP products in China. Tianjin Dawnhalo Appliance Co. produces a substantial amount of EDP based on modified starch. Several other companies are joined with starch-filled or photodegradable plastics.

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Polyolefin waste and its utilization in Polski Koncern Naftowy ORLEN SA

Summary — The results of the Environmental Compliance Program started in Polski Koncern Naftowy ORLEN SA in 1997 have been described. Realization of the Program allowed to reduce SO_2 annual emission from 38 kt to 16 kt, to decrease wastewater volume from $15 \cdot 10^6$ m^3/y to $6.6 \cdot 10^6$ m^3/y and to decrease the phenol content in wastewater from 8.2 to 3.8 kg/day. By-products and waste products created in polyolefins units have been described, especially atactic polypropylene and other kinds of wastes from various points of the PP and PE-LD plants. As a results of application of a new generation catalysts for propylene polymerization, atactic PP amount decreased from about 10 to 1.5 wt. %. Method of its application as a component of bitumens has been developed.
Key words: Polski Koncern Naftowy Orlen SA, environmental pollution reducing, polyolefin wastes, atactic polypropylene, utilization.

Polski Koncern Naftowy ORLEN SA was created in 1999 in the merger of Petrochemia Płock SA (the greatest Polish refinery and petrochemical complex) and Centrala Produktów Naftowych SA (the greatest Polish wholesale and retail distributor for petroleum fuels and lubricants). As a result, the greatest Polish refinery and petrochemical production as well as distribution company has been created. In 2000, the amount of crude oil processed reached up to 12 millions t/y. ORLEN SA supplies about 70% of domestic automotive fuels production. As for the production potential in Płock, both the refinery and petrochemical parts include up to 70 processing units altogether. Due to wide application of hydrogenation processes, the yield of white products reaches up to 83% of total production scope.

PKN ORLEN SA is the manufacturer of PE-LD and PP, moreover supplies the Polish market with such petrochemical products as: ethylene, propylene, ethylene oxide, glycol, butadiene, benzene, phenol and acetone. They form the feedstock base for both domestic chemicals and plastics production industry. PKN ORLEN covers almost the whole range of polyolefin production in Poland.

This production activity contributes to environmental impact of the whole refinery and petrochemical complex, therefore in 1997 there was introduced Environmental Compliance Program, including 28 modernization and investment tasks aiming the protection of all the components of natural environment.

So far, 16 tasks have been completed bringing significant environmental benefits, 10 tasks are in progress, and 2 are going to commence in 2002. One of the most important was the erection of the new Vacuum Residue Hydrodesulfurization Plant for sulfur removal from the fuel to our power plant. It enabled major SO_2 emission decrease from 38 kt/y in 1999 to only 16 kt/y in 2000.

Other indicators showing reduction of the environmental impact are as follows:

— percentage of sulfur removal during crude oil processing: increase from 33% to 58%,

— wastewater quantity: decrease from 15 mil. m^3/y to 6.6 mil. m^3/y,

— phenol content in wastewater: reduction from 8.2 kg/d to 3.8 kg/d,

— water intake from the Vistula River in 2000: increase by 5.6%,

— cooling water circulation: increase by 20%,

— wastewater discharged to the Vistula River: decrease by 29%.

The ambient emission monitoring system consists of four automatic monitoring stations for hydrocarbons, CO, SO_2, NO_x, H_2S, O_3. No excessive pollutant concentration has been detected at any of these stations. In most of the points no measurable concentration of aromatic hydrocarbons was detected.

Among the investments for environmental protection the following are of greatest importance:

— catalytic hydrogen vapors combustion units installed at wastewater treatment plant;

— plant for contaminated soil biodegradation;

— ground water quality monitoring system.

Also, all the new and modernized petrol stations in ORLEN SA network are equipped with all the latest technologies warranting high level of environment protection. The solutions applied include double walled fuel storage tanks with the permanent leak control or the control wells system for constant monitoring of ground pollution.

From the organizational activities for the environment protection in ORLEN SA, the most important is the implementation of environment management system according to ISO 14001 standard and participation in the Responsible Care program, understood as form of our activities for environment protection in our neighborhood. In 1999, after the certifying audit, ORLEN SA was awarded a certificate from the program co-ordinating office.

The environmental impact resulting from polyolefin production is not as significant as from other petrochemical products and automotive fuels. At polyethylene and polypropylene production units there are not operated furnaces, polluting the atmosphere with dust and sulfur or nitrogen oxides. Both the feedstock and the products are not liquid hydrocarbons as in the refinery part of the facility, which can pose a potential danger for ground water contamination with hydrocarbon products. However, both in the production stage and the packaging stage there appear some material wastes which are the subjects of activities aiming their reduction and ensuring their proper utilization.

There are two polypropylene units operating in Płock built according to the Japanese license of Mitsui Petrochemical Industries. The production bases on the slurry process of propylene polymerization in hexane using the Ziegler-Natta catalytic system. Polymerization reaction takes place in hexane environment at 70°C and under pressure up to 1 MPa. One of the basic conditions of proper process course is very high purity of monomer, solvent and hydrogen, which must be free from water compounds, sulfur, acetylene and other substances destroying the catalyst.

The major product of in the polymerization process in this unit is namely isotactic polypropylene, characterized with high molecular weight, high degree of stereoregularity and excellent mechanical properties. As the by-product atactic polypropylene of amorphic structure and the average molecular weight of about 10 000 is received, which is separated from isotactic polypropylene in hexane solution, then separated in special film condensers. Afterwards, this semi-liquid product is put into barrels and sold to customers.

Atactic polypropylene (PPA) is a major waste at the polypropylene plant due to relatively high quantity, which according to the license reach up to 10 wt. % of the isotactic polymer. Because of high scale production process, wastes utilization creates problems.

Apart from the atactic polymer there is also another waste polypropylene material, in the form of powder, quantity of 185 t/y, appearing in the process of samples taking and apparatus cleaning or from the filters. This waste material is packed into bags and sold to customers who use it for the production of extrusion elements not requiring good mechanical properties.

During polypropylene granulation and packing, other types of material waste appear as following:

— Wet polypropylene, quantity of 170 t/y, which appears during extruders start up and shut down as well as oversize grain and undersize grain from classification sieves. Its quantity depends on the frequency of extruders shut down and start up operations as well as the precision of the granule cutting.

— Fine grain polypropylene (powder), quantity of 20 t/y, being the product of PP dust extraction.

— Polymer spills, quantity of 140 t/y, produced during extruders start up in the granulation section.

— Polypropylene sweeps, quantity of about 200 t/y, from transport pipelines and storage tanks cleaning as well as from sweeps away.

— Polypropylene fibers created in the pneumatic transport from the production unit to the shipment; due to high velocity of granulate transport in the pipelines there occurred, especially in bends, lamination and creation of fibers. This waste was difficult to remove, a large portion of it was blown into the atmosphere and the remaining quantity was sent with the product to the customers.

— Waste film, quantity of 30 t/y, created in the laboratories in film tests, and left after packaging.

Total quantity of the waste material mentioned above is about 740 t/y, which is 0.6% of the total PP production.

The similar situation with waste material production is also at the two polyethylene units. First unit, built according to the ICI license, consists of two lines of 20 kt/y each. Ethylene is compressed in two-stage process up to 150 MPa. Polymerization reaction is realized in autoclave with peroxides as initiators; conversion degree is about 17%. Post-reaction mixture is separated, polyethylene is sent to extruder and pelletizer while ethylene is directed back to the process. Granulate is pneumatically sent to the homogenization section and to shipment.

The other unit licensed by ATO Chemie consists of two lines of 50 kt/y production capacity. The reaction takes place in the tubular reactor of length 810 m under pressure of 250 MPa. Oxygen is used as an initiator. The rest of the sections work according to the same technological scheme as autoclave unit.

In both of the units a waste material is created in the form of wax characterized with molecular weight 1000 to 2000 in the quantity 80 t/y. It is separated in the medium pressure recycle and put into barrels.

As in the polypropylene plant also in the polyethylene plant there is waste material created in the total quantity of about 800 t/y, which is of about 0.55% of the total PE production.

Activities aimed to reduce the quantity of material waste include:

— Reduction of atactic polypropylene produced by the application of new generation catalysts. As a result of two subsequent upgrades of the polypropylene unit depending on introduction of the catalysts of II and III generation, PPA production was reduced to about 1.5 wt. % and in 2000 was equal to *ca.* 1700 t with PP production of 125 000 t/y. As a result of this modernization, a few separate operations resulting in heavy environmental impact were eliminated. Also, research work was started on commercial PPA, which was burned before. As a result, technologies for bitumen production with PPA were prepared. Atactic polymer addition improved their thermal and rheological properties, as well as improved homogenic structure. Atactic polypropylene is also used as additive for bitumen products such as: roofing pitch, insulation and grounding bitumen products.

— Removal of fiber created during pneumatic transport was realized by the application at transport pipelines new bends of special construction "GAMA Bend" and application of de-dusting system.

The other polymer material waste created in the production process, blending operations and pneumatic transport, namely polypropylene powder, polypropylene and polyethylene dust, spills, sweeps and waste film from both units, are utilized by our customers by re-granulation, milling or direct application as feedstock for further processing.

The non-production waste from our factory as for example post maintenance waste as wood, metal, concrete, but also paper, batteries, bottles and films, are segregated at the automatic line.

Summarizing, polyolefin production process in PKN ORLEN SA produce waste materials which are completely utilized and do not pollute the environment. Activities with the aim to reduce waste quantity by the changes in process technology and improvements in the construction have been realized.