TADAMOTO SAKAI
Shizuoka University (visiting Professor)
Campus Innovation Center, 3-3-6 Shibaura, Minato, Tokyo 108-0023 Japan
e-mail: mtsakai@ipc.shizuoka.ac.jp

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Screw extrusion technology — past, present and future*)

Summary — Extrusion using a screw extruder has been playing very important roles in polymers processing as one of the basic processing technologies for producing compounds and various products such as pipes, car parts, films, *etc*. In this paper the history and recent technology progress in screw extrusion are reviewed, mainly focused on Japan. Screw extrusion has a long history starting from food processing to rubber and plastics, and is now applied to various fields, such as compounding, fiber spinning, film making, blow molding and injection molding. Recent progress in extrusion technology allows for precise selection of conditions, providing for production of polymers and nanocomposites with required properties, *e.g*. morphology/structure, as well as significant improvement in production efficiency and energy consumption. Today extrusion technology has been divided into two major streams. The first stream goes towards higher efficiency using larger scale processing lines, and the other towards the production of functional products with special properties, such as nanocomposites and/or polymer blends with specified nanoscale morphology. Starting from the wide range of extrusion technologies against the historical background, this review will cover new advancing extrusion technologies in processing of sustainable/eco-friendly biopolymers such as PLA or nanocomposites.

Keywords: screw extrusion, fiber reinforcement, nanocomposites, polymer blends, reactive processing, film extrusion.

TECHNOLOGIA WYTŁACZANIA ŚLIMAKOWEGO — PRZESZŁOŚĆ, TERAŹNIEJSZOŚĆ I PRZYSZŁOŚĆ

Streszczenie — Wytłaczanie odgrywa bardzo ważną rolę w przetwarzaniu polimerów, jako jedna z podstawowych technologii otrzymywania produktów, takich jak: rury, części samochodowe, folie, itp. W tej pracy przedstawiono historię i najnowsze osiągnięcia technologii wytłaczania, koncentrując się głównie na tematyce dotyczącej Japonii. Wytłaczarki ślimakowe były stosowane od dawna, począwszy od przetwórstwa spożywczego do przetwórstwa gumy i tworzyw sztucznych, a obecnie znajdują również zastosowanie w procesach, takich jak mieszanie składników, przędzenie włókien, wytwarzanie cienkich błon, formowanie z rozdmuchiwaniem i formowanie wtryskowe. Najnowsze technologie wytłaczania pozwalają na precyzyjny dobór warunków umożliwiających otrzymywanie polimerów i nanokompozytów o wymaganych właściwościach, np. morfologii/strukturze, jak również uzyskanie znacznej poprawy wydajności produkcji i oszczędności energii. Obecnie technologie wytłaczania rozwijają się w dwu kierunkach. Jeden kierunek prowadzi do uzyskiwania coraz większych wydajności na coraz większych liniach technologicznych. Drugi natomiast ma na celu wytwarzanie produktów funkcjonalnych o specjalnych właściwościach, takich jak nanokompozyty czy mieszaniny polimerowe charakteryzujące się określoną morfologią w nanoskali. Przegląd ten obejmuje także najnowsze postępy w technologiach wytłaczania zgodnych z zasadami zrównoważonego rozwoju i służących przetwarzaniu przyjaznych dla środowiska biopolimerów, takich jak np. poli(kwas mlekowy) (PLA).

Słowa kluczowe: wytłaczanie ślimakowe, włókno wzmacniające, nanokompozyty, mieszaniny polimerowe, reaktywne przetwarzanie, wytłaczanie folii.

INTRODUCTION

Today we are surrounded with many kinds of polymer products. According to the statistic data from Japan Plastics Industry Federation (JPIF), the world total pro-

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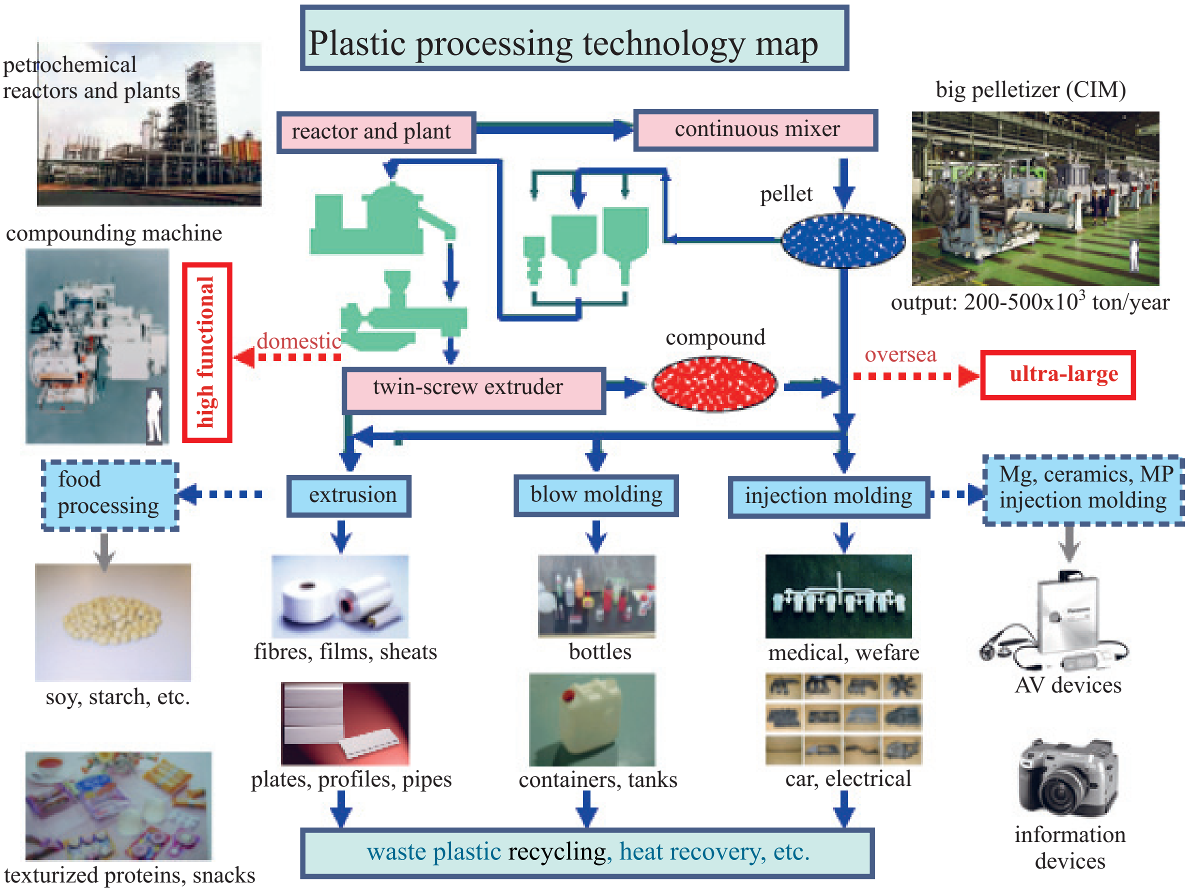


Fig. 1. Total polymer processing scheme from the polymerization to final products

duction of plastics was 256 million tons/year in 2010. With the production growth, the average world consumption of plastics per head has jumped up from 10 kg in 1980 to 40 kg in 2010. The growing trends are much steeper in the developing countries such as China or India. On the other hand, Japan has changed the target from mass-consumption to sustainably consuming country, taking into account high recycling ratio.

Figure 1 shows the total map of polymer related industries concerning polymer processing technologies mainly for thermoplastic materials. The trends in the major polymer industry have been advancing in two different directions. One is the mass production with the aim to reduce cost, for example 60—100 tons/h production capacity of polyolefin pellets. The other direction is production of high added value functional products with small/medium production capacity.

The largest polymers consumption (25—30 % of the total production) relates to packaging and containers, and on the other hand the consumption related to automobile parts is 8—10 %. Today, about 1 billion cars are used in the world by so-called „automobile dependent society", and the car number is remarkably increasing every year. Typical plastic car parts are, for example fuel tank, bumper, door trim and instrument panel. According to JPIF, the average polymer use for a passenger car in Japan was only 2 % in 1980, but it increased to 8 % in 2001 and to 10 % in 2010. Regarding various polymer materials for cars, the use of polypropylene has rapidly increased, but meanwhile consumption of poly(vinyl chloride) drastically decreased owing to the environmental recycling issues. Recent remarkable trends are the increase in the use of polypropylene, engineering polymers such as polycarbonate and newly emerging biopolymers.

The technological advancements in processing have been playing very important roles in the massive growth of polymers applications in the wide ranges of industries, in particular, automobile industry [1]. In production of car parts, major molding processes are injection molding and blow molding, but extrusion plays also an important role in compounding prior to the final forming of various car parts.

VARIOUS FUNCTIONS OF A SCREW TYPE EXTRUDER

As shown in Fig. 1, key equipment for polymer processing is a screw type extruder, which is used for compounding, extrusion, blow molding and injection molding. Figure 2 shows various kinds of screw type extruders widely used in the polymer industry. The screw extruders have been utilized as food and ceramic processors for long years. Major extruders are generally classified with a single-screw and a twin-screw type. A batch type mixer and multi-screw extruder (with more than three

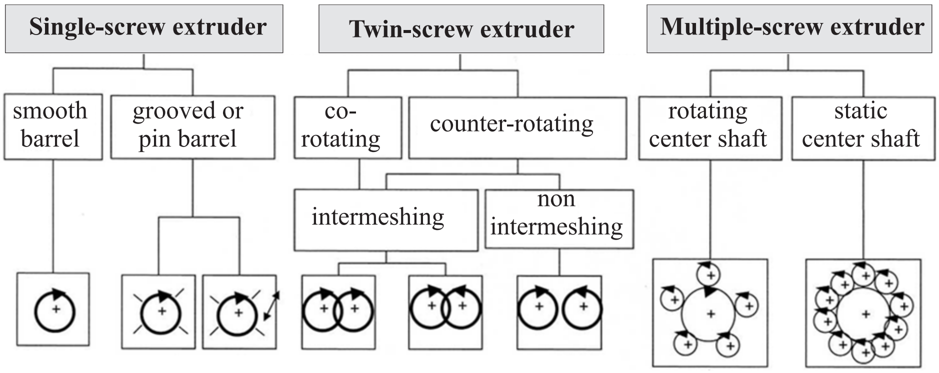


Fig. 2. The classification of continuous type screw extruders

screws) are only limited to use in small scale production or special applications. A single-screw extruder is widely applied to general polymer processing, such as blow molding, film making and injection molding. On the other hand, a twin-screw extruder is mainly used for compounding of various fibers, nano-size fillers and polymer blending prior to final molding. Key functions of screw type extruders are:

— conveyor for particles, granules and viscous fluids,
— melter for organic and inorganic particles,
— pressure riser for highly viscous fluids,
— mixer for granules and highly viscous fluids,
— grinder for granules, fibers and fine particles,
— reactor for particles and highly viscous fluids,
— devolatilizer for highly viscous fluids,
— squeezer for particles, granules and viscous fluids,
— heat-exchanger for particles and viscous fluids,
— crystallizer for organic and inorganic fluids,
— dryer for organic and inorganic particles and fluids,
— continuous pyrolizer for organic materials.

Table 1. **Functional comparisons between a single-screw extruder and a twin-screw one**

Single-screw	Twin-screw
Fair feeding	good feeding
Flood feeding	starve feeding
Fair melting	good melting
Fair mixing (distributive mixing)	good distributive and dispersive mixing
Non-self wiping	self wiping
Non-modular design	modular design (flexible)
Fair degassing	good degassing
Good pressurization	limited pressurization

Using these functions the control of the microstructure and the chemical modification of polymers are performed, as well as filler compounding. Table 1 explains the comparison between a single-screw extruder and a twin-screw one. Generally a twin-screw extruder has higher melting, mixing and devolatilization capability than a single-screw one. Owing to the recent high demand for higher additional-value production and lower consumption of energy used for processing, the application of a twin-screw extruder has been rapidly increasing, mainly in film/sheet extrusion so as to rationalize a conventional single-screw extrusion process line.

Technological progress of single-screw extrusion

The first single-screw extruder for polymers was produced in 1951 in Japan for extrusion of wire/cable coating from low density polyethylene. Since then, many kinds of single-screw extruders have been manufactured and used for example, for fiber-spinning of polyamide and polyester and for film-making of poly(vinyl chloride) and polyolefin.

A single-screw extruder is roughly composed of three zones, that is, solid conveying, melting and melt conveying (metering). Theoretical simulation on single-screw extrusion was started for molten polymer flow in the metering section by Carley *et al.* in 1953 [2]. Furthermore the first melting theory was published by Tadmor in 1966 [3]. However, practical extrusion behavior is normally more complicated than such theoretical models. Therefore various theories concerning molten polymer flow, melting and solid conveying in single-screw extruder have been reported until now [4].

Thanks to the technological progress in extrusion, a lot of mixing elements for a single-screw extruder have been developed and recent theoretical simulation allows analyzing three-dimensional and non-isothermal flow and mixing behaviors. In addition to recent progress in theoretical simulation techniques, the utilization of extensional flow mixing is becoming popular as well as shear flow mixing.

Technological progress of twin-screw extrusion

Following the increase in plastics consumption, the demand for larger extrusion capacity and better mixing performance drastically raised and therefore a twin-screw extruder has been playing more important role in capacity increase than a single-screw extruder in the field

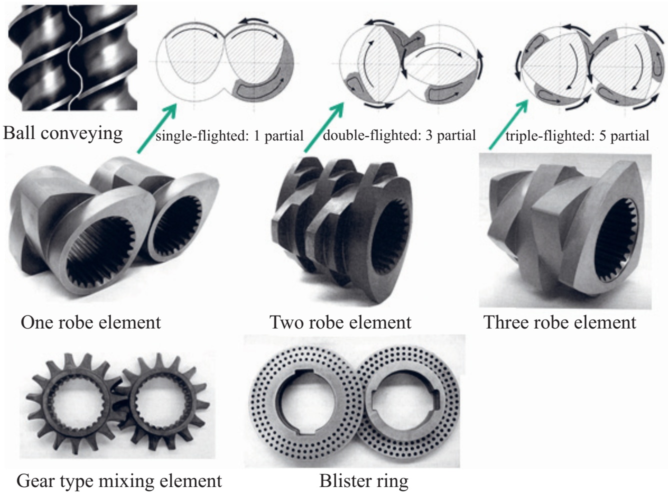


Fig. 3. Various mixing elements for an intermeshing co-rotating twin-screw extruder

of compounding/pelletizing since early 1970s. The most important differences between a twin-screw extruder and a single-screw extruder are higher melting/mixing capability and self-cleaning effect, as well as precisely segmented screw/barrel structure. For a twin-screw extruder, various kinds of mixing elements have been also developed since 1960s, as shown in Figure 3. In this figure the most popular mixing element is the combination of diversified kneading disks with left-handed/right-handed configuration and/or the adjustable thickness. As a result, these mixing elements are applicable to different operational purposes with wide variety of materials.

In case of an intermeshing co-rotating twin-screw extruder, molten polymer flow behavior was analyzed in detail and several flow simulation programs were commercialized in 1990s. Furthermore, recent theoretical developments enable to apply sophisticated 3D analyses for the complex kneading disk alignment and to contribute greatly to mixing effect improvement. Figure 4 shows grooved barrel structure for an intermeshed co-rotating twin-screw extruder as an example of complex screw/barrel geometries. The flow behaviors around grooved barrels and kneading disks were calculated by a particle tracing method [4]. From these results the combination of grooved barrels with kneading disks was optimized so as to obtain more homogeneous flow behavior at lower polymer temperature. Lately the optimum combination of grooved barrels and kneading disks has been successfully applied, for example in practical use of polypropylene-carbon nanotube compounding.

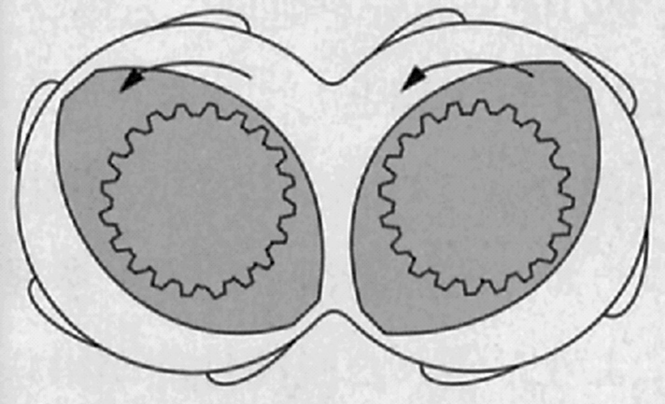


Fig. 4. Grooved barrel structure for an intermeshing twin-screw extruder

HISTORY OF SCREW EXTRUSION RELATED TECHNOLOGIES

Table 2 explains the historical progress in polymer processing technology in the past. In this table double underline items show the progress in technology related to polymer materials, single underline items concern to developments in processing machinery and other items relate to the activities of the academic or industrial society for plastics and rubber. The history of extrusion has started from rubber processing and then applied to plastics processing in the early 1930 after the commercialization of polystyrene, poly(vinyl chloride) and polyethylene.

Table 2. History of polymer processing technologies

Year	Technology
1909	Synthetic rubber — methyl-isoprene (Hoffman, Germany)
1916	Banbury mixer for rubber (Farrel, USA)
1930	Polystyrene production (BASF, Germany)
1931	Poly(vinyl chloride) production (BASF, Germany)
1933	Injection molding machine for plastics (Franz, Germany)
1938	High pressure type polyethylene production (ICI, UK)
1939	Intermeshing type twin-screw extruder (LMP, Italy)
1939	Single-screw extruder for plastics (Troester, Germany)
1943	1st Annual Meeting of Society of Plastics Engineers (Detroit, USA)
1949	Extrusion blow molding machine (Kautex, Germany)
1951	Single-screw extruder in Japan (JSW, Japan)
1952	Foundation of Japan Society of Polymer Science (Japan)
1953	Single-screw extrusion theory (DuPont, USA)
1955	Modular type twin-screw extruder (W&P, Germany)
1955	Twin-screw extruder for PVC in Japan (Toshiba, Japan)
1959	Single-screw melting behavior (Maddock, USA)
1962	Devolatilization theory in extrusion (Latinen, USA)
1965	1st International Plastics Fair (Tokyo, Japan)
1966	Single-screw melting theory (Tadmor, USA)
1968	Ziegler-Natta catalyzed polyolefin (Germany, Italy)
1972	Reactive extrusion for graft copolymer (Ide, Japan)
1975	Injection molding process simulation (Wang, USA)
1978	Injection molding simulation commercialization (MF, Australia)
1981	Thermoplastic elastomer by dynamic cure (USA)

1981	Twin-screw extruder melting theory (Sakai, Japan)
1983	More than 10 000 injection molding machine production (Japan)
1985	1st Polymer Processing Society Annual Meeting (Akron, USA)
1987	Over 10 million tons plastic production/year (Japan)
1989	5th Polymer Processing Society Annual Meeting (Kyoto, Japan)
1990	Foundation of Japan Society of Polymer Processing (Japan)
1990	Nylon-Clay, nanocomposite for car parts (Toyota, Japan)
1991	Metallocene catalyzed polyethylene (Japan, USA)
1991	Thermoplastic elastomer/polypropylene blends (Toyota, Japan)
1993	Supercritical fluid foaming injection molding (Trexel, USA)
1998	4th Polymer Processing Society Annual Meeting (Yokohama, Japan)
2002	Poly(lactic acid) commercial production (Cargill/Dow, USA)
2004	22nd Polymer Processing Annual Meeting (Yamagata, Japan)

The full-scale developments in the plastics industry and polymer processing technologies in Japan have started since 1950s. Japan Society of Polymer Science (SPSJ), which mainly deals with polymer materials, was founded in 1952, but Japan Polymer Processing Society (JSPP) was established 48 years after this foundation. JSPP has three kinds of periodical meetings to be held every year (Annual Meeting, Regional Meeting and Asian Workshop). On the other hand, International Polymer Processing Society (PPS) was established as a global academic society at Akron (USA) in 1985. And till now, Annual PPS Meetings were held three times in Japan (1989 in Kyoto, 1998 in Yokohama and 2004 in Yamagata).

Several polymer processing machines such as extruders have started to be manufactured in Japan in early 1950s [5]. However, a very interesting thing is that the elementary components of these machines have not essentially changed, although their size, control/monitoring equipment, output capacities drastically changed. For example, in case of single-screw extruders, the output of 65 mm screw diameter was only 30 kg/h in 1950's (at present, more than 500 kg/h) and the cable coating speed was 66 m/min (now, more than 2000 m/min).

Recently, the capability of processing machines was much upgraded in accordance with the expansion of the polymer industry in the world. For example, a large scale single-screw extruder with the diameter D = 700 mm, a large scale intermeshing twin-screw extruder of the diameter D = 450 mm, a long intermeshing twin-screw extruder with screw and barrel length ratio L/D = 100 and a large scale devolatilization twin-screw extruder with six vent holes. These machines were manufactured in Japan during 1990—2000 corresponding to higher demand for productivity and functionality in the polymer industry.

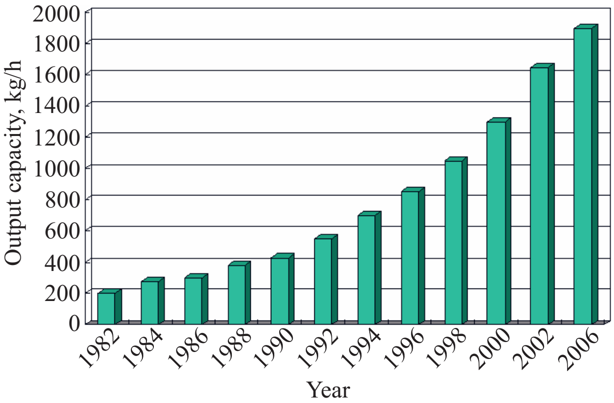


Fig. 5. Productivity improvement in typical compounding for an intermeshing twin-screw extruder 65 mm in diameter (according to [6])

Furthermore, the output capacity of both of a single- and twin-screw extruder has been dramatically improving every year since 1960s, thanks to the improvement in sophisticated screw mixing segments, longer L/D (such as L/D = 40—100), greater screw channel depth (D_{outer}/D_{inner} = 1.60—1.85), higher screw speed (600—2500 rpm), larger screw-drive torque, *etc*. Figure 5 shows the historical throughput progress in an intermeshing co-rotating twin-screw extruder for typical compounding with inorganic fillers (calcium carbonate) and polypropylene, as an example [6]. The throughput of a twin-screw extruder has jumped up about 10 times in 20 years, mainly owing to progress in both machine design and material improvements.

RECENT PROGRESS IN SCREW EXTRUSION

Fiber reinforcement

Carbon fiber reinforcement

Various kinds of plastic products reinforced with glass fibers are widely applied in injection molding mainly in the area of automobile parts and electric appliances. Recent developments are focused on the applications of carbon fibers (CF) to highly processed products such as airplanes, trains and passenger cars. One of the topical examples is the application of CF reinforced plastics (epoxy resin matrix) to a newly developed jet-liner, Boeing 787. Thanks to the technological progress in carbon fiber reinforcement, the total weight reduction attained nearly 50 % and the fuel consumption was reduced by 20 % compared to conventional jet liners. The other advantage of the CF use is that the safety can be improved. A carbon fiber reinforced plastic propeller shaft for a car shows higher impact absorption effect. In this application, the CF reinforced shaft indicates considerably higher impact energy absorption and vibration stability, which means superior performance in safety compared to a steel shaft.

In 2011 Toyota Motor and some other car manufacturers put on sales light-weight sport cars, which fully utilized CF reinforcement. Furthermore, many car manufacturers are now focusing on developing a small electric vehicle mainly used in cities, in which thermoplastic polymers are reinforced with CF. The utilization of CF to a passenger car parts easily applicable to injection molding is the next development target for Japan. For this purpose, long CF reinforced molding compounds, based on polypropylene and polyamide as a polymer matrix are coming on the market.

As other examples can be mentioned particularly polymer nanocomposites (PNC) containing carbon nanotubes (CNT). Single wall type CNT, have been now highlighted [7]. The applications of CNT composites are expected in lithium ion batteries, car/aerospace micro-precise parts such as micro-devices and small medical parts such as micro-catheter, as well as high-performance sealing for oil-drilling equipment.

Natural fiber reinforcement

Various fibers such as aramid and aromatic polyester are widely applied to the reinforcement in various products such as car tires. Today, the applications of natural fibers obtained from, *e.g.* sisal, jute, silk, bamboo and oil palm, are becoming popular, because they have great advantages in both sustainability and high specific strength, as shown in Table 3 [8]. To meet the present requirement in CO_2 emission reduction, many car manufacturers are trying use a combination of natural fibers and biopolymers such as kenaf or bamboo fibers. Toyota Motor recently put on sales a hybrid car, commercially named „SAI", which total inner surface is composed in 80 % of biopolymers.

Table 3. Characteristics of various natural fibrous materials (according to [8])

Fibrous material	Density g/cm^3	Tensile modulus MPa	Tensile strength MPa	Specific modulus GPa/gcm^{-3}	Cost/modulus
Hemp	1.50	13	—	—	—
Flax	1.30	13—26	344	71	1
Jute	1.50	19—35	393	—	—
Kenaf	1.50	15—37	—	—	—
Sisal	1.45	16—37	510	3—82	1.5
Bamboo	0.9—1.2	21—38	310—720	—	—
Glass fiber	2.50	70	3400	28	3
Carbon fiber	1.75—2.0	250—600	5000	134	30

Additionally, the compounding of nano-sized natural fibers obtained from cellulose fibrils is also becoming a hot topic lately, because high mechanical strength and transparency are expected, when nano-fibers are used for polymer composites [9]. Promising products in the near future are flexible organic electronic materials (EL) and car parts. Nano-fibers from natural cellulose pulp are obtainable when a high speed twin-screw extruder is used for grinding under high shearing conditions.

Polymer nanocomposites

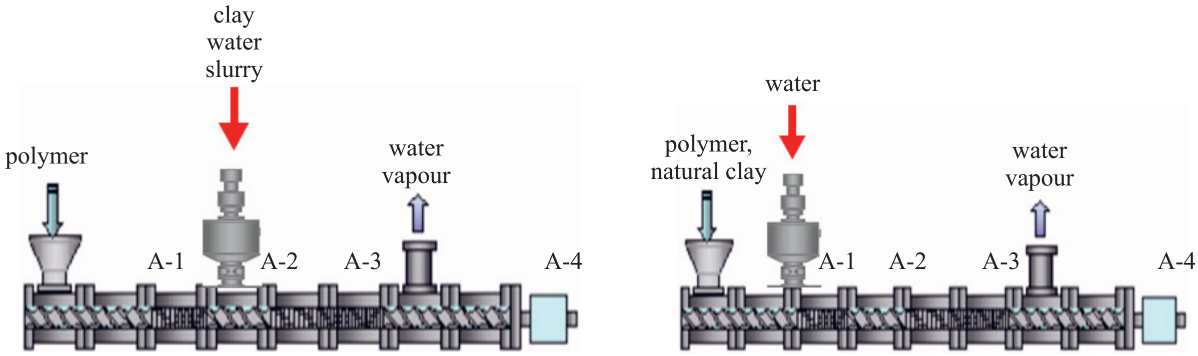
Polymer nanocomposite (PNC) contained in nano-size inorganic filler is attracting much attention as an innovative composite since early 1990s. The first PNC, which is composed of polyamide and nano-size clay, was applied to production of car parts in 1990 in Japan [10]. It was developed in 1983 in a polymerization process. However, major developments in the world have been focused on melt-compounding processes, aiming to lower the manufacturing cost. In the case of car parts, furthermore, polypropylene is targeted as the most promising polymer matrix because of its light weight and high performance. The most serious problem is how to obtain polypropylene-based PNC with prospective physical properties. In the conventional melt-compounding process, multi-compounding steps were essential in the chemical modification both for nanoclay and polypropylene, because of their poor miscibility or affinity between two components. However a new compounding process was developed in Japan in 2003 in order to reduce the cost [11, 12]. This innovative extrusion process of PNC compounding is shown schematically in Figure 6. In this process, a large amount of water is injected to enforce higher shearing stress and better dispersion of nanoclay particles into a molten polymer in a twin-screw extruder. As a result of the water injection, the simplified extrusion process was applied to the polymer matrix and nanoclay without any kind of chemical modification. In Figure 6 PP is polypropylene and PETG is glycol modified polyethylene terephthalate, respectively.

In other examples, biopolymer PNC based on polylactide (PLA) was developed at Unitika in Japan [13]. The PLA-PNC offers greatly improved performance in high crystallization speed, heat-resistance and mechanical strength for injection-molded products.

Polymer blending

Using twin-screw extrusion/compounding technology, lots of polymer alloys were produced in the polymer industry since late 1960s. At the early stage of polymer blending the main target were miscible polymer systems, but since late 1970s polymer blending for immiscible components has been actively developed.

In the early stage of 1960—1980 there was no clear mechanism to control the morphology during twin-screw extrusion/compounding. Therefore, many visualization experiments and analyses on the morphology formation during extrusion have been actively carried out since late 1980s [14]. As an example, the complex



PP-clay nanocomposite PETG-clay nanocomposite

Fig. 6. Newly developed PNC compounding systems (according to [11, 12])

morphology formation in a twin-screw extruder was observed concerning a ternary polymer blending system (polyphenylene ether/elastomer/polyamide) for car parts [15]. During polymer blending operations from feeding to pelletizing, various kinds of physical and/or chemical changes were taking place in a twin-screw extruder, such as melting, dispersion, chemical reaction around the boundary layers of each phase and subsequently phase inversion [16]. For the morphological analyses of the product formed during blending, on-line sampling valves were utilized to extract polymer on-line from a twin-screw extruder. The molten polymer extracted at each zone was cooled down with liquid nitrogen as quickly as possible, prior to the progress chemical reaction and change the morphology.

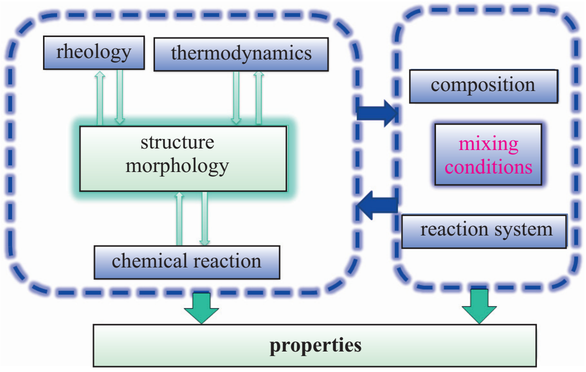


Fig. 7. Relationship between morphology, properties and mixing conditions (according to [17], reprinted under the permission of Plastics Age)

As a result of these studies on the morphology, the blending mechanisms in a twin-screw extruder have gradually been made clear in 1990s. Figure 7 shows the schematic relationship concerning the morphology changes during mixing, processing and component properties such as thermodynamic and/or rheological characteristics [17]. The conformation of phase morphology in the polymer blends by reactive melt blending is very complex, depending on the processing conditions and the material properties. The parameters related to material properties are molecular *e.g.* structure, melt viscosity, melt elasticity and surface tension. Additionally the processing conditions, such as temperature, pressure, shear rate, residence time and feeding sequence, play very important roles in morphology formation during twin-screw extrusion [14, 17].

Today, the focal point of interest concerning studies on morphology of a polymer alloy moved from micron- or sub-micron scale to nanoscale. That is because the nanoscale morphology offers the improvement in various physical and chemical properties. In particular, the improvement in optical and/or crystallization properties of molded products is greatly achieved in case of PLA polymer blends [18].

In late 2000s the sophisticated nanoscale morphology was reported by Atami *et al.* [19]. Utilizing a special compounding procedure with an extremely long twin-screw extruder (L/D = 100), the nanoalloy obtained from this procedure exhibited distinctive mechanical properties, that is, very high elongation value under very high extrusion speed of 1000 mm/min, just the same as under low extrusion speed of 100 mm/min [19]. When the fracture behavior was compared with conventional high-impact polyamide and the nanoalloy (polyamide/polyolefin blends), the nanoscale polymer blends did not break down, but just deformed without cracks even under very severe impact conditions, under which the conventional tough polyamide was completely damaged. Taking advantages of these excellent characteristics, this polymer has been applied to various impact absorbing products such as car fender parts, sporting goods and head protection gears.

Reactive processing

„Reactive processing" or „reactive extrusion" using a single- or twin-screw extruder has been long applied to chemical polymerization of high-viscosity polymers and devolatilization. Today the terminology of „reactive processing" widely includes reactive polymer blending and chemical modification of polymers, as well as chemical polymerization accompanied with devolatilization operations [20—22]. Table 4 shows various items — candidates for reactive processing. This technology is rapidly progressing as important method to create highly functional polymers until now and from now on.

Table 4. Classification of reactive processing and commercialization items

Type of reactive processing	Product obtained
Bulk polymerization	ε-caprolactone, PMMA, PU, PLA, PS, PA, PC, *etc.*
Graft copolymerization	MA grafted PP, alkoxysilane grafted PE, *etc.*
Reactive blending	polymer blends, toughened PP, dynamic vulcanization, TPE, TPV, *etc.*
Chemical modification	functional EVA, chain-extended PET, long-chain branched PP, *etc.*
Depolymerization	MW modified PP, devulcanized rubber, *etc.*
Chemical recycling	L-lactide, styrene monomer, MMA monomer, *etc.*
Dewatering	post-treated rubber, ABS, *etc.*

Waste rubber reclaiming

Today our society requires all plastic products to be treated with proper recycling procedures. Reactive processing technology was applied to an innovative recycling process to reuse waste rubber from used cars and produce directly dynamic vulcanization thermoplastic elastomer (TPE) [23]. This process contains two kinds of recycling procedures. In the first procedure, waste ethylene-propylene rubber (EPDM) was mechanically broken down to lower molecular weight by high shearing forces generated from screw rotation in a twin-screw extruder. The mechanism of this reclaiming procedure is the selective scission of cross-links between weak S-S bonds by mechanical shear stress as shown in Figure 8. The reclaimed EPDM was reused for car parts again, because reclaimed EPDM vulcanizates obtained from this procedure exhibited the equivalent mechanical properties compared to virgin ones in quality. The second recycling procedure is to obtain dynamically vulcanized thermoplastic elastomer (TPE or TPV) added with polypropylene into a twin-screw extruder, when reclaimed EPDM is obtained at the first reactive processing procedure. This TPE obtained from the second reactive processing is used for sealing rubber in cars. In these reactive processes the water injection was utilized for both generation of higher shearing stress to shear cross-links and the efficient extraction of nasty odor from thermally degraded rubber.

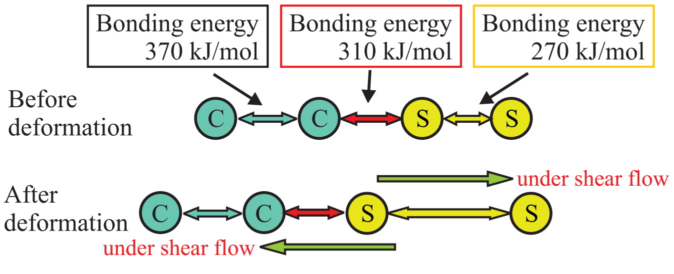


Fig. 8. Reactive processing for waste EPDM reclaiming (according to [23])

Chemical recycling of waste PLA

A pyrolysis process to obtain a monomer with a screw type extruder is common, particularly in case of polymethylmethacrylate [24]. Recently, this technology was applied to biopolymer chemical recycling. In the case of PLA products used for electric appliances, flame retardant agents are normally contained in the products. The flame retardant agents such as $Al(OH)_3$ and MgO accelerate chemical reactions to decompose polymer chain into monomer as a catalyst. Kyushu Institute of Technology has been developing this process in Japan as one of Government Projects since early 2000s. As a result, as shown in Table 5, the high L-lactide recovery rates were obtained when reactive processing technology was successfully applied using a twin-screw extruder [25]. In this system the moisture venting operation allowed more efficient recovery of L-lactide from waste PLA products.

Table 5. Chemical recycling results to obtain lactide from waste PLA (according to [25])

Test number	Moisture trap, %	Lactide recovery %	Composition of raw material PLA
1	36 % (venting from No. 1 vent port)	74.7	PLA contained with $Al(OH)_3$ as flame retardant agent
2	50 % (venting from No. 1 vent port)	93.0	PLA contained with $Al(OH)_3$ as flame retardant agent plus MsO as catalyst 0.16 %
3	— (without venting from No. 1 vent)	56.8	PLA contained with $Al(OH)_3$ as flame retardant agent

Since the establishment of The Kyoto Protocol to the United Nations Framework Convention on Climate Change in 1997, our society began to express a deep interest in developing bio-based polymers, so-called „biopolymers" and natural reinforcement additives. If practical ways to produce biopolymers from natural resources are developed, an eco-friendly or sustainable system of plastic products is achieved as shown in Figure 9 [1]. Recent technological development targets of biopolymer technology are listed in Table 6. Commercial production

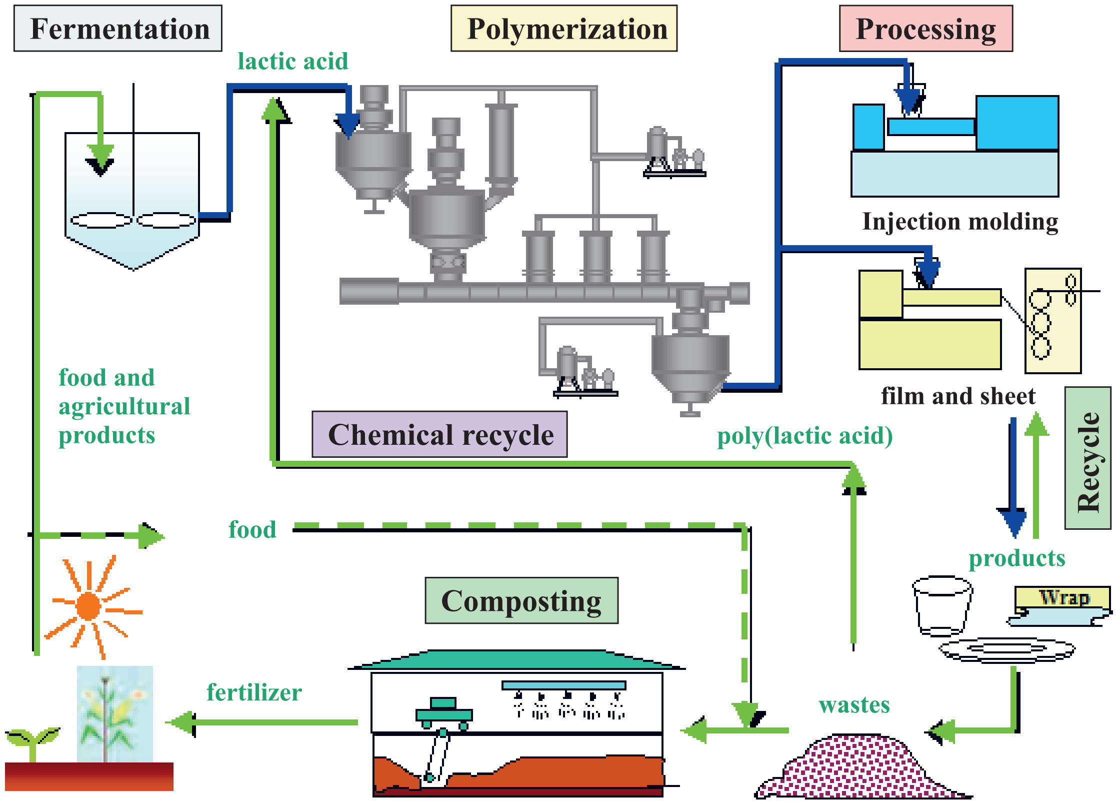


Fig. 9. Eco-friendly patterns for a bio-polymer such as PLA

of several kinds of biopolymers is now being developed in the world, although PLA is the most popular biopolymer at present. The biopolymer growth in the world is expected to reach more than three million tons per year in 2020 [26].

Table 6. Various bio-polymers developed in Japan

Biopolymers produced from 100 % biomass	poly(lactic acid) (PLA) polyhydroxyalkanoate (PHA) polyolefin (Bio-PE, Bio-PP) nylon 11 and nylon 10-10
Biopolymers produced from parts of biomass	cellulose acetate (CA) trimethylene-terephthalate (Bio-PTT) polyethylene-terephthalate (Bio-PET) polyurethane, unsaturated polyester polycarbonate (Bio-PC), Nylon 610 polybutylene-succinate (PBS)

Film/sheet extrusion

Until now, film/sheet extrusion has been developing as one of the most important polymer processing technologies since the embryonic time of 1950s for plastics, in parallel with fiber extrusion. Accordingly, the present film/sheet production reached about 30 % market share of the total plastics consumption in the world. Furthermore, recently new applications of film/sheet products have been rapidly advancing in various fields of electronic and optical industries, as well as in conventional packaging and agriculture fields. As a result of these trends, film/sheet extrusion technology is developing in two major directions. One is higher productivity with lower manufacturing cost, for example package films 6.5—10 meters in width, and the other is higher functionality, such as light guide films produced by sophisticated biaxial stretching extrusion. High performance polymer films/sheets produced by extrusion can be classified into the following types:

— polarizing film, phase contrast film, anti-reflection film, optical diffusion film;

— reflection film, electro-conductive film, condenser film, anti-static film, dicing film, photosensitive film, release film;

— solar battery sealing film, lithium battery separator film, fuel cell membrane;

— optical barrier film, vapor deposited film;

— window seal film, easy peal film, anti-corrosion film, anti-fogging film, *etc*.

Recently the applications of these sophisticated film/sheet products have been rapidly expanding towards various fields.

Highly multi-layered film extrusion

Film extrusion processes for thermoplastics are mainly classified into two groups, that is, blown film extrusion and melt-cast film extrusion [27]. Nowadays, multi-layered film extrusion technology has been rapidly advancing, for example, in packaging application. Furthermore, the number of multi-layers is greatly increasing, depending on the application. In a special case, about 1000 layered film is applied. It has metallic gloss and is used for a car part such as tachometer display. These multi-layered films are fabricated using sophisticated multi-feed block dies connected with several screw extruders.

Optical film extrusion

Highly functional film applications for optical devices such as LCD-television, mobile phone, personal computer, car navigation system, *etc*. Standard manufacturing process of these films was usually a solvent casting method until now, because of the difficulties in control of the optical anisotropy and thickness precision. Recently an extrusion casting process has been developed as a more efficient production method, thanks to recent progress in the extrusion technology.

Today a conventional sequential biaxial stretching film production line is widely applied to various thermoplastic polymers such as PET, polyethylene, polypropylene, polycarbonate, cyclopolyolefin and PLA. Up to now, the major stream of film extrusion technology was how to obtain higher production speed and wider film with better thickness precision. As a result, nowadays the take-up speed attained more than 500 m/min, stretched film width is 8—10 meters (50 years ago, the film speed was only 10 m/min in Japan as mentioned before).

The emerging technology in film extrusion deals with obtaining higher-value-added film/sheet for electronic and/or optical applications. As an example of the sophisticated technology in film extrusion, a special production line of biaxially stretched oblique films (with 45 degree optical orientation) was developed at Nippon Zeon [28] in Japan. In this film-stretching line, the film is stretched in the direction of 45 degree in the center so as to obtain specially oriented film with target angle. Owing to this development, a continuous roll-to-roll production process is directly available for fabricating multi-layered optical films. Prior to this technical development, patched phase difference film was used for the lamination at a batch production line.

Micro or nano surface transcription

According to the progress in utilizing fine structural devices in the fields of biology and communication equipment, innovative technologies have been strongly demanded so as to precisely transcript fine 3D structures of micro- or nanoscale on the film surface. For this purposes, special transcription technology such as „roll-to-roll" process or „nano-in-printing" method has been actively developed since early 2000s. A continuous transcription method was applied to non-reflection film with complicated surface of moth-eye structure for LCD TV, as an example [29]. By use of moth-eye structure, TV screen with few reflections could be achieved.

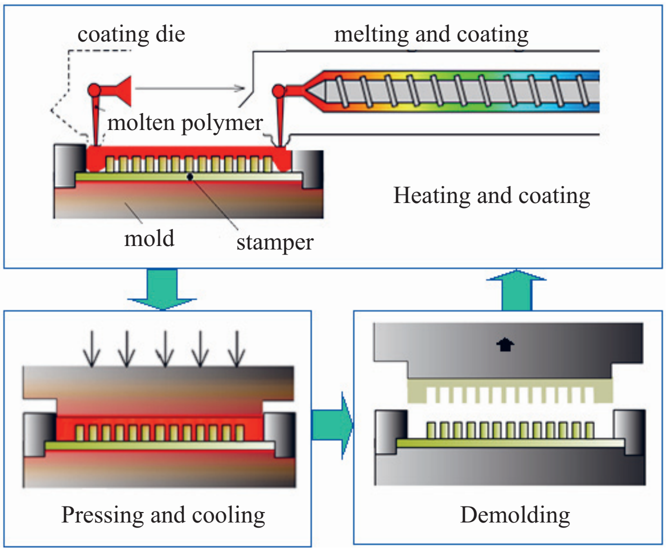


Fig. 10. „Melt transcription molding" system for micro- or nano-devices (according to [30, 31], figure courtesy of JSW)

In case of the transcription with micro- and/or nanoscale surface structure, the „nano-in-printing" method is popular at present, but the disadvantages of this process are long processing cycle time such as 15—30 min and the transcription area limitation. An innovative transcription method has been developed in early 2000s in Japan, which is so-called „melt-transcription molding" process mainly in the electronic and/or medical fields [30, 31]. This process utilizes a combination of extrusion (molten polymer coating by extrusion) and hot embossing technology. Figure 10 shows the outline of a „melt-transcription molding" system. Three dimensional structures of 5 micron scale with 120 mm square were fabricated within 30 s, for example, using polymethylmetacrylate, polycarbonate and cyclopolyolefin. The advantages of the „melt-transcription molding" are the reduction of cycle time and the applicability of larger transcription area and higher aspect ratio.

CONCLUSION

Major modern plastics industry in the world has started since 1950 in Japan, and today the total plastics production of the world has attained about 300 million tons per year. Extrusion technology has greatly supported this advancement as the most important and fundamental tool for polymer processing.

Up to now the polymer industry has been focusing on higher productivity, smaller energy consumption and easy recycling. From now, however, high quality/performance of diversified products and the sustainability are indispensable in our society. To reconcile these conflicting issues, we have to fully utilize many kinds of processing technologies to control and modify the polymer morphologies and structures, combined with total quality management systems and sophisticated operational and simulation software. Sophisticated extrusion technology is essential to contribute towards these future targets more than ever.

Table 7. New trends/counter-measures in polymer processing technology

New trends		Counter-measures
— Diversified and higher production — Thinner and sophisticated surface/inner structuring — Lower energy consumption — Highly multi-layering and structuring — Sustainable polymer use	⇨	— development of various hybridized processes — efficient mixing and cooling for screw, dies and rollers — precise and stable machine parts and units — utilization of CAE for design — line monitoring and control — polymer modification to fit processability improvement

In conclusion of this review, new trends and counter-measures in today's polymer processing technologies are summarized in Table 7, focused mainly on screw extrusion technology.

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