



# PWMI Newsletter

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Plastic Waste Management Institute  
JAPAN

## At the Frontline of Plastic Recycling

# Latest Trends in Post-use Plastic Recycling in Japan

– New and Innovative Chemical Recycling Technologies –

In 1999 and 2000, the amount of plastic consumed in Japan reached about 11,000 thousand tons/year after running at about 10,000 thousand tons/year from 1990 on.

In relation to this, the total amount of plastic waste discharge was about 5,600 thousand tons in 1990 rising to about 9,800 thousand tons in 1998 and then 10,000 thousand tons in 2000.

Looking only at fiscal year 2000, the amount of plastic consumed in Japan was 10,980 thousand tons. Here, 9,070 thousand tons of this amount will become waste in the form of post-use plastic, or 9,970 thousand tons if we include production and processing waste. The recycling rate for this 9,970 thousand tons of plastic waste is 4,940 thousand tons or about 50% for material, chemical, and energy recovery combined.

Similar to many countries around the world, Japan is seeing a year-on-year increase in the amount of post-use plastic waste. To deal with this trend, the country is testing and deploying a variety of recycling technologies with the aim of making effective use of resources and constructing a recycling-oriented society. Of these technologies, chemical recycling in particular is attracting much attention.

We here report on the current state of chemical recycling technologies in Japan. These include technologies using chemical techniques to “return” post-use PET bottles to raw material the same as that prepared from naphtha, technologies for turning post-use plastic into raw material for use as cement fuel, and technologies for breaking down polyvinyl chloride for recycling purposes.

## 1. Monomerization of PET Bottles

### Recycling technologies for "returning" post-use PET bottles to monomers using new chemical techniques

In material recycling, which is based on physical techniques, quality deteriorates after repeated recycling making it difficult to use the material in question. This prompted the development of recycling technologies that would be the first in the world to "return" PET bottles to raw material the same as that prepared from naphtha.

These technologies, which use chemical techniques to return post-use PET bottles to monomers, represent a totally new concept in recycling.

The developers of these new technologies are Teijin Ltd. and AIES Co., Ltd.

#### 1. New chemical-recycling technology developed by Teijin

##### ● "Returns" most polyester products to high-grade materials

The chemical-recycling technology developed by Teijin Ltd. uses chemical techniques to recover dimethyl terephthalate (DMT) from post-use PET bottles. The DMT recovered in this manner is equivalent to the resin material prepared from naphtha for use in manufacturing PET bottles.

This process is based on a material recycling technology that has been used at Teijin since 1962 following the

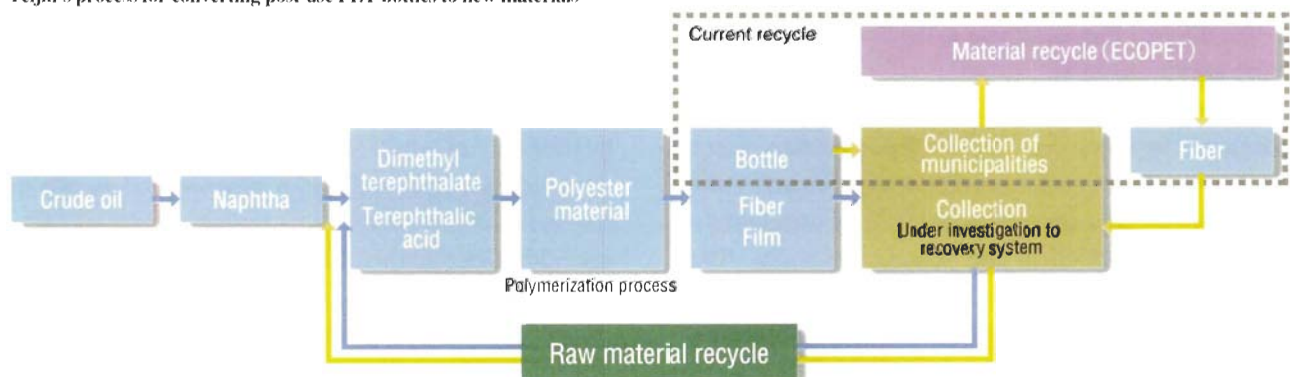
start of polyester production in 1958. That technology chemically breaks down the waste thread generated in manufacturing processes in Teijin plants and turns it back into polyester material. Further improvements to this technique have resulted in a landmark technology that can recover material equivalent to that prepared from naphtha for manufacture of fiber, film, resin, and other products, and that enables almost all polyester products to be used for recycling.

Recycling polyester by chemical techniques as in the above technology means that polyester can be reused any number of times. In this sense, chemical recycling is playing a significant role in Japan's efforts to construct a recycling-oriented society. This form of chemical recycling, moreover, presents no problems in terms of public safety and health, and the company has applied for 57 patents including peripheral technologies.

Teijin has also developed a technology for re-manufacturing PET bottles by refining the DMT recovered from post-use PET bottles into terephthalic acid (TPA).

Operations for re-manufacturing PET bottles in the above way commenced in April 2002 at Teijin's Tokuyama Factory in Yamaguchi Prefecture. These initial operations can recover about 24,000 tons/year of high-grade DMT from about 30,000 tons/year of collected PET bottles (equivalent to about 1 billion 500-ml PET bottles). Facilities will be expanded in 2003 to expand annual DMT recovery to about 50,000 tons/year with the idea of converting this DMT to TPA to achieve "bottle-to-bottle" PET bottle recycling.

Teijin's process for converting post-use PET bottles to new materials





**2. Chemical-recycling technology by AIES for "returning" PET bottles to high-grade PET resin**

**●Rebirth of post-use PET bottles**

At AIES Co., Ltd., extensive research into whether old PET bottles can be turned back into new PET bottles has culminated in a recycling technology called the "AIES process." In this process, old PET bottles are converted to flakes from which resins of different color additives and materials are removed. It was the first chemical technique in the world to return post-use PET bottles to monomers that can then be made into resin for producing PET bottles.

The most outstanding feature of this chemical-recycling technology is that the PET resin generated achieves a level of quality equivalent to that of PET resin manufactured from naphtha.



Process for converting post-use PET bottles to new materials

**●Unlimited recycling by "bottle-to-bottle" technology**

AIES's original chemical-recycling technique breaks down post-use PET bottles into a monomer (bis-2hydroxyethyl-terephthalate (BHET), an intermediate material in the manufacture of PET resin), and then manufactures high-grade resin for PET bottle use by distillation and refinery. AIES says that this "bottle-to-bottle" technology enables PET bottles to be recycled any number of times. To apply this technology, a company called PET Rebirth Co., Ltd. has recently been established, and a business plan has been drawn up for constructing a plant within the premises of Nippon Oil

Corporation (a joint-investing company) in Kawasaki City in Kanagawa Prefecture. Business operations are scheduled to commence in January 2004, and the plan here is to manufacture 24,500 tons of PET resin annually from an annual collection of 27,500 tons of post-use PET bottles.

As for the safety of re-commercialized PET resin (that is, the safety of each process in the manufacture of new products from post-use PET bottles by chemical techniques), the Japan Food Hygiene Association was commissioned by Japan's Ministry of Health, Labour and Welfare to conduct a study. It was found that "absolutely no problems are present with regard to safety." In addition, AIES independently submitted an application to the US Food and Drug Administration for confirmation of safety, and received an acknowledgment letter stating that the AIES process is a "tertiary recycling process and confirmed to be safe."

AIES's original chemical recycling flow



## Recycling rate of PET bottles in Japan exceeds 40% for the first time in FY2001

The amount of PET bottles produced in Japan in 2001 came to about 402,700 tons, and 161,200 tons or 40.1% of this amount were recycled. In addition, the rate of PET-bottle sorted collection for all cities, towns, and villages in Japan was high at 80.6%.

As for material recycling, the total amount for 2000 was 68,575 tons. Re-commercialization by product type is shown in the graph below.

Recycling rate for re-commercialization by product type in 2000



Incidentally, if we compare the PET-bottle recycling rate in 2000 among Japan, US, and EU, we see that Japan's rate of 34.5% was significantly higher than the 22.3% and 20.0% rates of the US and EU, respectively. This figure was actually the best rate in the world for that year.

Recycling rates of Japan, US, and EU

		1997	1998	1999	2000
Japan	Production	21.9	28.2	33.2	36.2
	Volume of collected bottles	2.1	4.8	7.6	12.5
	Rate of collection %	9.8	16.9	22.8	34.5
USA	Production	115.7	136.4	147.4	159.7
	Volume of collected bottles	31.3	33.8	35.0	35.6
	Rate of collection %	27.1	24.8	23.7	22.3
EU	Production	98.0	105.0	122.0	155.0
	Volume of collected bottles	10.8	17.2	21.9	27.0
	Rate of collection %	11.0	16.4	18.0	20.0

[thousand tons]  
 Japan The Council for PET Bottle Recycling  
 USA NAPCOR (The National Association for PET Container Resources)  
 EU PETCORE (PET Container Recycling Europe)

The chemical-recycling technologies of Teijin and AIES are progressing within the framework of Japan's Containers and Packaging Recycling Law. We can expect the overall recycling rate to rise dramatically when combining the volume of chemical recycling with that of material recycling.

## 2. Making Plastic into Cement Fuel and Material

### From post-use plastic to fuel and material for the cement industry

Cement is a familiar material in everyone's life, and it is an important material in the construction of social capital. In the cement manufacturing process, large quantities of industrial waste plastic are being recycled as fuel making a great contribution to the construction of a recycling-oriented society. Technology has also been developed for recycling polyvinyl chloride as fuel and material (which has not been previously done) after separating the chlorine.

#### ● Using post-use plastic as fuel and material

The cement industry accepts a wide variety of materials as "raw material resources." These include coal ash discharged from power stations, blast furnace slug from iron works, sewage sludge, industrial sludge, ash from incineration plants for household trash, and discarded tires.

In recent years, moreover, the industry has also come to accept industrial waste plastic discharged from plastic and plastic-product manufacturers. This recycling of waste plastic as fuel and material should bring about major changes in the cement manufacturing process.

Thermal energy is indispensable to the manufacture of cement.

Inside the firing equipment (kiln) at the heart of the cement manufacturing process, high temperatures from 1,100 to 1,800°C can be achieved by burning coal, discarded tires, industrial waste plastic, etc. Under these conditions, the system continuously pours cement material into the kiln and transfers chemically altered cement material to cooling equipment. This latter material is called "clinker" (an intermediate cement product).

The thermal energy needed for the cement manufacturing process has traditionally been obtained through the introduction of coal (pulverized coal), discarded tires, etc., as described above. As of yet, the saving of resources by using (recycling) high-calorie industrial waste plastic as a substitute for some coal has not had a significant impact on the cement manufacturing process.

According to a survey performed by the Japan Cement Association, the amount of recycled industrial waste



plastic has been increasing steadily from 29,000 tons in 1998 to 58,000 tons in 1999 and 102,000 tons in 2000.

In addition, the fact that cement quality using the conventional manufacturing method does not suffer at all when using recycled industrial waste plastic is a significant feature, and we can expect the cement industry to be expanding their use of industrial waste plastic in the years to come.

Up to now, thermal energy in the cement manufacturing process has depended mainly on coal. The recycling of industrial waste plastic as cement fuel and material, however, should play a major role in advancing the construction of a recycling-oriented society by decreasing environmental loads and preserving resources.



Giant rotary kiln (at the Tokuyama Factory)

#### **What is the significance of "fuel and material" here?**

Industrial waste plastic, whose main components are hydrogen and carbon, has high-calorie properties. This makes it suitable as auxiliary fuel material, while the ash left behind after burning this fuel can also be recycled as a portion of cement raw material.

"Fuel and material" therefore refers to two key characteristics possessed by industrial waste plastic. In other words, industrial waste plastic plays a role in both material and energy recovery.

### **Tokuyama Corporation aims to accept 150,000 tons/year of industrial waste plastic**

#### **● Tokuyama has been developing recycling technology for industrial waste plastic since 1992**

The use of discarded tires in addition to coal in the cement manufacturing process is a mature technology that was first deployed some time ago. In 1992, however, amid growing social concern about the need for recycling industrial waste plastic, the Tokuyama Factory of Tokuyama Corporation began development of recycling technology to address this need.

First, as an initial development concept, they addressed

the question of what the plastic crush size should be to make recycling most efficient and to optimize economy and cost competitiveness.

The conclusion reached was that hard waste plastic should be 2 cm or less in size and that soft waste plastic should be 3 cm or less in size. In 1995, the company performed joint trials with the Plastic Waste Management Institute (PWMI) to verify the validity of these requirements, and the results confirmed expectations.

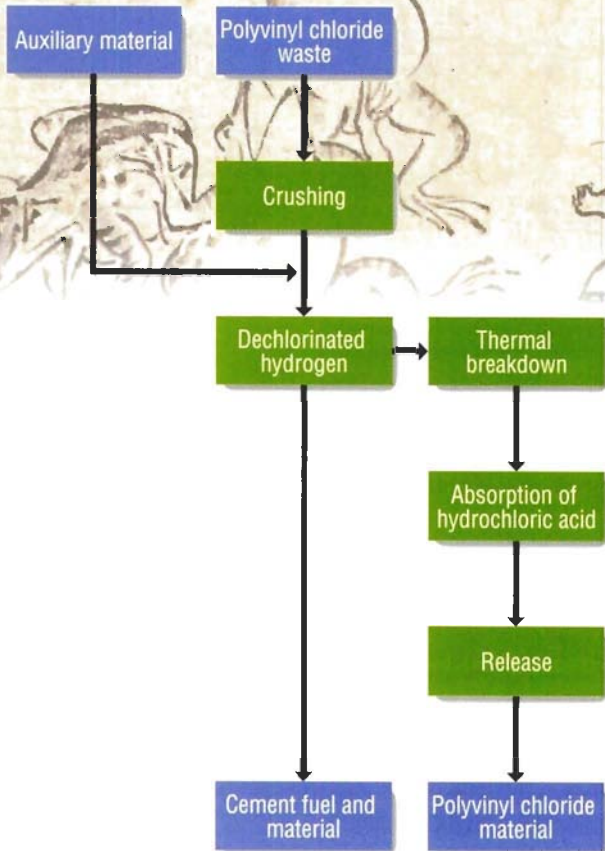
#### **● A fuel and material capacity of 45,000 tons/year achieved**

Then, in August 1999, a plant with an annual capacity of 15,000 tons/year was constructed followed by another plant with an annual capacity of 30,000 tons/year in June 2001. A total capacity of 45,000 tons/year was therefore achieved for converting industrial waste plastic into fuel and raw material.

This system has a number of key features. For example, the energy needed for converting (crushing) waste plastic to fuel and material is relatively small, the resource-recovery rate of industrial waste plastic is high, and large quantities of recycled fuel can be obtained. There are many expectations for this recycling technology in the years to come.

The industrial waste plastic crushed at these plants is blown from the front of a rotary kiln (total length: 110 m; diameter: 5.7 m) into its interior as fuel and raw material.

Here, 90% of the industrial waste plastic introduced into the kiln is of the soft type.



### 3. Polyvinyl chloride separation technology

#### Chemical recycling of polyvinyl chloride progresses-separates polyvinyl chloride into hydrocarbon and hydrogen chloride

About 2,400 thousand tons of polyvinyl chloride are produced annually in Japan with about 1,700 thousand tons of this amount used domestically and the rest exported. In addition, slightly more than 1,000 thousand tons of post-use polyvinyl chloride are discharged with about 34% of this being recycled.

Polyvinyl chloride has been recycled for some time. This includes the material recycling of post-use agricultural polyvinyl chloride (film), which has been performed for over 30 years.

Polyvinyl chloride recycling technology has been progressing significantly in recent years. Major steel and chemical companies and other enterprises are undertaking the development of chemical recycling technology, and it appears that the recycling of polyvinyl chloride is entering a new stage.

#### ● An indispensable element in the progress of chemical recycling: polyvinyl chloride separation technology

The chemical recycling of polyvinyl chloride is progressing in the form of various methods including blast furnace reduction, conversion to cement fuel and material, liquefaction, and gasification.

In blast furnace reduction, the process separates polyvinyl chloride into hydrocarbon and hydrogen chloride and utilizes the hydrocarbon as a reducing agent in a blast furnace.

In the conversion to cement fuel and material, the process separates polyvinyl chloride into hydrocarbon and hydrogen chloride as in blast furnace reduction, but here uses these products for some of the fuel and material required in the cement manufacturing process.

Liquefaction as well separates polyvinyl chloride into hydrocarbon and hydrogen chloride but then refines the hydrocarbon into heavy oil. The above three technologies produce hydrochloric acid from the separated hydrogen chloride.

Chemical recycling of polyvinyl chloride

#### ● Polyvinyl chloride also recycled as fuel and material after separating chlorine

Tokuyama Corporation, Vinyl Environmental Council (VEC), Japan PVC Environmental Affairs Council (JPEC), and PWMI have performed joint trials on polyvinyl chloride recycling and have established a viable technology for it.

At the trial plant, the process separates chlorine from 1,500 tons/day of polyvinyl chloride in a chlorine-separation furnace (350°C) and recycles the residue (char) in the furnace as fuel and material for cement manufacturing. As the system is still at a trial-plant stage, the volume of recycled polyvinyl chloride is not that large, but the results achieved have demonstrated that the recycling itself has no problems.

#### ● 50% target set for the recycling rate of industrial waste plastic as fuel and material

By consolidating the above technologies, the target is to increase the amount of recycled industrial waste plastic used as fuel to about 50% in the near future.

If this can be achieved, the recycling of industrial waste plastic will take a giant leap forward, and this in itself is eagerly awaited by society.

The Tokuyama Factory, by the way, produces 6 million tons of cement annually, the second largest capacity in Japan.



As for gasification, its special feature is the thermal breakdown of polyvinyl chloride into a gas, which is then quenched to separate the chlorine component and produce hydrogen as a product. The recovered chlorine can be used as raw material for fertilizer.

In any of these methods, the process of separating polyvinyl chloride into hydrocarbon and hydrogen chloride is indispensable. As described below, a technology for separating polyvinyl chloride under a nitrogen atmosphere has been added to these methods.

## New concept in polyvinyl chloride separation technology

### NKK Keihin Works establishes technology for separating polyvinyl chloride in nitrogen gas

The NKK Keihin Works developed this technology in cooperation with VEC and PWMI with the support of the New Energy and Industrial Technology Development Organization (NEDO). This technology separates highly concentrated polyvinyl chloride into hydrocarbon and hydrogen chloride in a rotary kiln under a nitrogen atmosphere.

NKK began development of this technology in 1997 and conducted trials in March of this year. The results obtained confirmed expectations.

#### ● Rotary kiln born of a novel idea

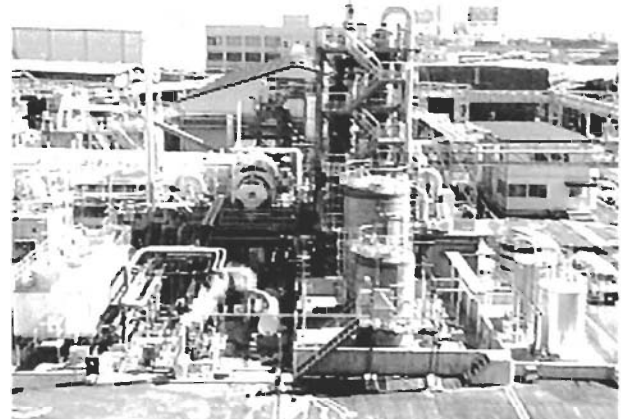
In NKK's process, post-use polyvinyl chloride is first placed in a crushing machine and then dried by hot air at temperatures from 100 to 120°C. This dried polyvinyl chloride is then directly introduced into a compactor and made into fist-size pellets using the heat generated by the friction of dual-axes screws.

This pelletized polyvinyl chloride is now transported by conveyer into a rotary kiln having an original NKK design.

The originality of this rotary kiln has several aspects. First, to achieve efficient thermal breakdown of polyvinyl chloride, the pelletized polyvinyl chloride is introduced into the rotary kiln together with coke, a raw material of steel, and subjected to thermal processing. In

addition, a rotary kiln that is heated from the outside is adopted, and a completely nitrogen atmosphere is employed inside the rotary kiln.

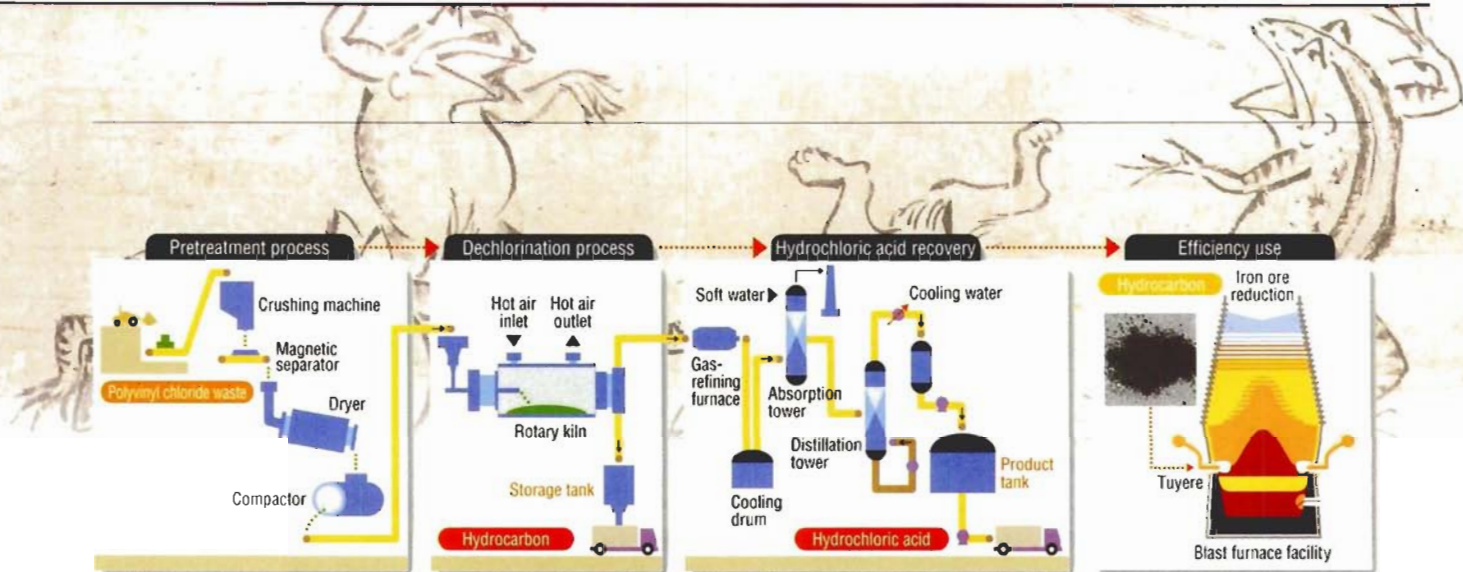
Thus, in the rotary kiln, the pelletized polyvinyl chloride is heated in a nitrogen atmosphere by a hot blast of air from the outside to temperatures from 300 to 350°C, and is then thermally broken down in this baked state into hydrocarbon and hydrogen chloride. An efficient system is achieved by re-circulating the hot air used for heating and using surplus exhaust gas to heat nitrogen.



View of polyvinyl chloride recycling plant



Rotary kiln for thermal breakdown of polyvinyl chloride into hydrocarbon and hydrogen chloride



NKK process flow for converting polyvinyl chloride into blast furnace material

### ● Why nitrogen, external heating, and coke?

This rotary kiln is cylindrical in shape with a diameter of 2 m and a total length of about 20 m, and has an annual processing capacity of 5,000 tons. With this kiln, why is a nitrogen atmosphere, an external heating system, and coke used? The reasons are given below.

- ① Nitrogen gas is inert (i.e., stable; it does not react with other substances), which means that secondary reactions do not occur.
- ② An external heating system enables the decomposed gas and heating gas to be separated making for more compact facilities.
- ③ On the other hand, an external heating system is not good in terms of heat transfer efficiency. To therefore improve heat-transfer characteristics, coke is also introduced. Coke, which is coal in a baked state, has high heat-transfer efficiency.

Furthermore, with the aim of maintaining stable operation, coke plays the role of a dispersing material that prevents the generation of large lumps of polyvinyl chloride during thermal breakdown. It also helps to scrape off polyvinyl chloride that has become attached to the inner wall of the rotary kiln.

### ● Ratio of polyvinyl chloride and coke in kiln is optimized in the range from 1-to-1 to 3-to-1

The thermal breakdown of polyvinyl chloride runs smoothly as long as the mixing ratio lies within this range. This ratio varies somewhat according to whether the polyvinyl chloride is of the hard or soft type. In addition, a polyvinyl chloride ratio greater than 50% presents no particular problem even if thermoplastic resin other than polyvinyl chloride becomes slightly

mixed in.

As the system has moved from the idea stage to a thermal breakdown plant for recycling polyvinyl chloride, it goes without saying that efficiency will be greater the less there is of resin other than polyvinyl chloride.

Within the kiln, the hydrocarbon thermally decomposed from polyvinyl chloride is separated from the coke and used as blast furnace material (reducing agent) together with reservoir coke. This plant also features the reuse of material. For example, the coke used in the separation of hydrocarbon is re-introduced into the kiln together with recycled polyvinyl chloride.

### ● Hydrogen chloride is quenched and recovered in a process flowing from a high-temperature gas-refining furnace to a cooling drum

In addition, gas that has been sent to a gas-refining furnace and thermally broken down within the kiln will be quenched in a cooling drum at temperatures below 80°C after combustible material included in the gas has been burned for at least two seconds at 1,300°C.

The hydrogen chloride in the gas is then recovered by an absorption tower at 99% efficiency or greater. The gas remaining after recovering the hydrogen chloride is then neutralized and made harmless by a caustic soda or similar and released into the atmosphere.

In this absorption tower, the concentration of hydrogen chloride absorbed in soft water is from 20 to 25%, which means that a 35% solution of hydrochloric acid can be obtained through distillation. This hydrochloric acid is now being tested for use on an acid-cleaning line in a hot strip mill at the NKK Keihin Works.

