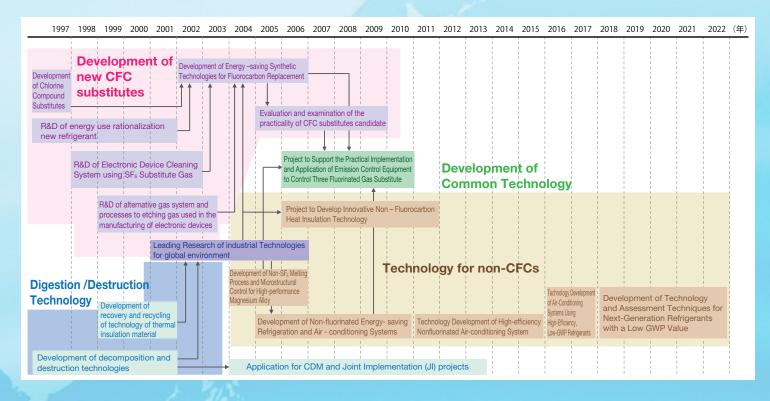


# NEDO Environment Department Progress in the Field of Reducing Fluorocarbon Emissions 2021



#### **INDEX**

I. Background and Purpose of R&D
Fluorocarbon Controls and Measures to Reduce Fluorocarbon Emissions
Trends of International Regulations on Fluorocarbons
Japanese Regulations on Fluorocarbons
II. NEDO R&D Projects
Development of HFC-23 Destruction Technology
Development of Technology for Chemical Recycling of HCFC Refrigerants
Development of Energy-saving Synthetic Technologies for Fluorocarbon Replacements
Development of Chlorine Fluorinated Gas Substitutes
R&D of SF6 Substitute Gas Cleaning System for Electronic Device Manufacturing
Development of Non-SF6 Melting Process and Microstructural Control for High- performance Magnesium Alloy
Project to Support the Practical Implementation and Application of Emission Control Equipment to Control Three Fluorinated Gas Substitutes
Development of Non-fluorinated Energy saving Refrigeration and Air-conditioning Systems
Project to Develop Innovative Non-Fluorocarbon Heat Insulation Technology
Technology Development of High-efficiency Non-fluorinated Air-conditioning Systems
Technology Development of Air-Conditioning Systems Using High-Efficiency, Low-GWP Refrigerants20
Development of Technology and Assessment Techniques for Next-Generation Refrigerants with a Low GWP Value
III. Project List



### I. Background and Purpose of R&D

#### Fluorocarbon Controls and Measures to Reduce Fluorocarbon Emissions

After the Montreal Protocol was adopted in 1987, the shift from using specified CFCs to using HFCs accelerated. Accordingly, there was concern about the sharp rise in greenhouse gas emissions in the fields of refrigeration and air conditioning. In 1992, the United Nations Framework Convention on Climate Change was adopted, followed by the adoption of the Kyoto Protocol in 1997. Japan achieved its target of reducing greenhouse gas emissions by 6% from the 1990 level by the end of the first commitment period (2008-2012).

In 2015, the Paris Agreement was adopted, under which Japan set a target to reduce greenhouse gas emissions by 26% from the fiscal year 2013 level by fiscal year 2030. In addition, an amendment was made to the Montreal Protocol, known as the Kigali Amendment, which stipulates the reduction of HFC production and consumption. Even refrigerants for refrigeration and air conditioning equipment that meet the target GWP values under the Act on Rational Use and Proper Management of Fluorocarbons in Japan may fall short of meeting the standards required by the Kigali Amendment.

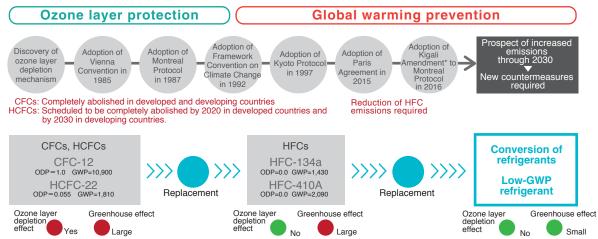
The Ministry of Economy, Trade and Industry revised the fluorocarbons usage forecast in July 2020 to realize the application of lower-GWP refrigerants to achieve the reduction targets. Based on the declaration on carbon neutrality by 2050, further countermeasures to reduce GHG emissions for fluorocarbons such as HFCs are urgently required.

<Major substances whose usage needs to be reduced to prevent global warming>

General	Specific	od CECs	Four fluorinated gas substitutes			
term	Specified CFCs		Three fluorinated gas substitutes			-
Туре	CFC	HCFC	HFC	PFC	SF <sub>6</sub>	NF <sub>3</sub>
International regulations Substances subject to control under the Montreal Protocol (regulations of production and import), not subject to control under the Kyoto Protocol		Substances subject to control under the Kyoto Protocol and the Paris Agreement (Nf <sub>3</sub> was added in 2013)				
Ozone layer depletion effect	Large	Relatively small	Do not deplete the ozone layer at all			
Greenhouse effect (GWP*1)	Extremely large (approx.10,000)	Large (several hundred- approx. 2,000)	Large (several hundred-approx. 4,000) *2	Extremely large (approx. 6,000- 9,000)	Extremely large (approx. 23,900)	Extremely large (approx.17,200)
Main applications	Refrigerants for refrigeration and air conditioning equipment     Detergents, solvents, and the like (Completely abolished after 1995)	Refrigerants for refrigeration and air conditioning equipment     Detergents, solvents, and the like (Scheduled to be abolished by 2020)	Refrigerants for refrigeration and air conditioning equipment     Foaming agents for heat insulation materials, and so on	Semiconductors, liquid crystal manufacturing     Detergents, solvents	Electrical insulating equipment     Semiconductors, liquid crystal manufacturing     Magnesium manufacturing	Semiconductors, liquid crystal manufacturing, and the like

<sup>\*1</sup> GWP: Global Warming Potential (a value representing the number of times the greenhouse effect of CO<sub>2</sub>)

<sup>\*2</sup> Value as a main refrigerant type

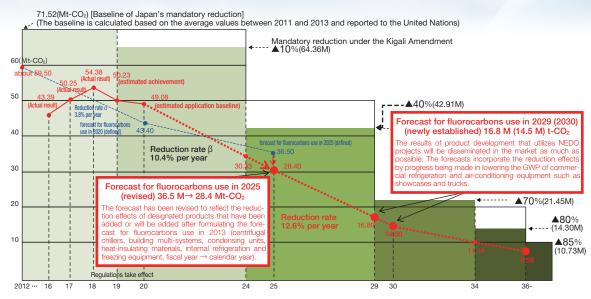


Ozone-depleting potential (ODP): This term refers to the relative strength of the depletion effect affecting the ozone layer when the strength of CFC-11 is fixed at 1.0. \*Kigali is the name of the capital city of Rwanda where the 28th Meeting of the Parties to the Montreal Protocol (MOP28) took place.

The agreement is called the Kigali Amendment since it was concluded at this meeting.

(NEDO

1



Forecast for fluorocarbons use (revised in 2025, and newly established in 2030)

Source: Data by WG on measures to deal with Fluorocarbons, Manufacturing Industry Committee, Industrial Structure Council (March 3, 2021)

#### Trends of International Regulations on Fluorocarbons

#### The Montreal Protocol and the Kigali Amendment

Based on the Vienna Convention for the Protection of the Ozone Layer (1985), the Montreal Protocol was adopted in 1987 as an international framework to control the usage of fluorocarbons. Since the adoption of the protocol, the production and import/export of controlled substances (CFCs and HCFCs) have been regulated in stages in developed countries. In addition, it was decided in September 2007 at the 19th Conference of the Contracting Parties to the Vienna Convention to reduce consumption of those gases in stages. The use of CFCs was completely phased-out in 1996, and the use of HCFCs will be substantially phased-out by 2020. For this reason, the development of substitutes has been steadily progressing. In Japan, the production and import/export of ozone-depleting substances have been regulated since 1989, and measures to steadily reduce production have been implemented.

However, as there is conversion from specified chlorofluorocarbons to fluorinated gas substitutes, the problem of the greenhouse effects of fluorinated gas substitutes has arisen, and measures concerning fluorocarbons have transferred from ozone layer protection to global warming prevention. As a result, the Kigali Amendment to the Montreal Protocol, which establishes duties and the like for the gradual reduction of HFC production and consumption, was adopted at the 28th Meeting of the Parties to the Montreal Protocol (MOP28), held in Kigali, the capital of Rwanda, in October 2016. This amendment provides regulations concerning the production and consumption of HFCs, and aims for gradual reduction according to the schedule shown in Table 1.

Table 1 Schedule of Gradual Reduction of Production/Consumption of HFCs in the Kigali Amendment to the Montreal Protocol

	Developed countries*1	Developing countriesgroup 1*2	Developing countries group 2*3
Baseline years	2011–2013	2020–2022	2024–2026
Baseline formula (HFC + HCFC)  Average HFC production/consumption for 2011-2013 + 15% of HCFC baseline		Average HFC production/consumption for 2020-2022 + 65% of HCFC baseline	Average HFC production/consumption for 2024-2026 + 65% of HCFC baseline
Freeze None		2024	2028*4
Phase-down schedule* <sup>5</sup>	2019: ▲ 10% 2024: ▲ 40% 2029: ▲ 70% 2034: ▲ 80% 2036: ▲ 85%	2019: ▲10% 2024: ▲40% 2029: ▲70% 2034: ▲80% 2036: ▲85%	2032: ▲10% 2037: ▲20% 2042: ▲30% 2047: ▲85%

<sup>\*1:</sup> Belarus, the Russian Federation, Kazakhstan, Tajikistan, and Uzbekistan, which are categorized as developed countries, are provided with different control measures (as to the baseline calculation, 25% of HCFCs are a component of the baseline; as to the phase-down schedule, the first step is a 5% reduction in 2020, and the second step is a 35% reduction in 2025).

 $<sup>^{\</sup>star}$ 5: All parties to the protocol will conduct technical assessments in 2022 and every 5 years thereafter



<sup>\*2:</sup> Developing countries - group 1 are developing countries that are not categorized as group 2.

<sup>\*3:</sup> Developing countries - group 2 are India, Pakistan, Iran, Iraq, and the Gulf Countries.

<sup>\*4:</sup> Group 2 countries are to conduct technical assessments 4 to 5 years before 2028 to consider the compliance deferral of two years from the freeze of 2028.

#### The Paris Agreement and Japan's approach to the Agreement

The 21st Session of the Conference of the Parties to the United Nations Framework Convention on Climate Change (COP21) by members was held in Paris in November 2015, at which all countries including the primary emitting countries attended and agreed to the Paris Agreement. Global long-term objectives were established, including the suppression of global increases in the average temperature since before the Industrial Revolution to a temperature substantially below 2°C (2°C target), the submission and update of reduction targets every five years by all countries, and the investigation of the status of initiatives globally every five years.

Furthermore, ahead of COP21, the Japanese government submitted to the United Nations Framework Convention on Climate Change Secretariat a treaty draft aiming for a globally superior and ambitious 26.0% reduction from FY2013 to FY2030 (a 25.4% reduction from FY2005). Based on this, the Japanese Cabinet decided the Plan for Global Warming Countermeasures for promoting global warming countermeasures comprehensively and strategically on May 13, 2016. This plan states that Japan, which is at the top of advanced countries with high emissions to GDP, aims at a future improvement up to 40% ( $0.29 \rightarrow 0.16$  kg-CO<sub>2</sub>/US dollars). On June 11, 2019, the Japanese Cabinet approved the Long-Term Strategy under the Paris Agreement as Growth Strategy as a long-term strategy for low GHG emission development under the Paris Agreement. By setting the ultimate goal of a decarbonized society (\*This is the first time a target of net-zero GHG emissions in the G7 has been set.) and ambitiously aiming to realize that as early as possible in the second half of this century, Japan has decided to make bold efforts toward the realization of reducing GHG emissions by 80% by 2050.

Table 2 Target Reductions of Emissions of Four Fluorinated Gas Substitutes in Plan for Global Warming Countermeasures

	2013 (millions of tons of CO <sub>2</sub> )	2030 (millions of tons of CO <sub>2</sub> )	Ratio of total emissions (%) compared to 2013
CO <sub>2</sub> from energy sources	1235	927	21.9% reduction
CO <sub>2</sub> , CH <sub>4</sub> , and N <sub>2</sub> O from non-energy sources	134.4	123.5	0.8% reduction
Four fluorinated gas substitutes (HFC, PFC, SF <sub>6</sub> , NF <sub>3</sub> )	38.6	28.9	0.7% reduction
Greenhouse gas absorption sources (absorption by forests, etc.)	_	(37.0% reduction)	2.6% reduction
Total	1408	1042	26.0% reduction

#### Japanese Regulations on Fluorocarbons

#### Act on the Protection of the Ozone Layer Through the Control of Specified Substances and Other Measures

Japan has been advancing regulations on specified chlorofluorocarbons under the Act on the Protection of the Ozone Layer Through the Control of Specified Substances and Other Measures as domestic security measures of the Montreal Protocol. Specifically, it is a mechanism to achieve the reduction objective of the protocol by establishing standard limits on production and consumption by allotting production and consumption amounts to individual businesses with yearly manufacturing licenses and import approval. However, HFCs, which are fluorinated gas substitutes, became subject to the protocol due to the Kigali Amendment to the Montreal Protocol in 2016, in addition to specified chlorofluorocarbons. So, under the revision of this law in 2018, the production and consumption of HFCs are now regulated in Japan.

#### Act on Rational Use and Proper Management of Fluorocarbons

Fluorocarbons cause depletion of the ozone layer and global warming, so it is necessary to control emissions to the atmosphere. Therefore, the Act for Securing, etc. the Implementation of Recovery and Destruction of Fluorocarbons Contained in Specified Products was established in 2001, and it has advanced the recovery of fluorocarbons during the provision and disposal of commercial refrigeration and air conditioning equipment, as well as the destruction of fluorocarbons. However, in addition to problems such as the sudden increase in refrigerant HFCs, the stagnation of the refrigerant recovery rate, and ascertaining large-scale leaks during use of equipment, response based on changes in the status surrounding fluorocarbons, known as trends toward global regulations on HFCs, was required. To that end, the law was amended in June 2013 in order to include comprehensive measures throughout the entire lifecycle, from production to disposal of fluorocarbons, in addition to the recovery and destruction of fluorocarbons, and the name was changed to Act on Rational Use and Proper Management of Fluorocarbons (entered into force April 1, 2015). Furthermore, to improve the recovery rate at the time of disposal, which has remained sluggish at slightly less than 40% for over a decade, the act was revised in June 2019 to allow for drastic measures to be taken such as the introduction of a direct punishment for violations in which users do not collect fluorocarbons at the time of disposal of equipment (effective from April 1, 2020).

The Act on Rational Use and Proper Management of Fluorocarbons requires initiatives for controlling emissions of fluorocarbons, as shown in Table 3, by manufacturers of fluorocarbons, manufacturers of equipment which use fluorocarbons, equipment users, filling/collecting operators, and destroying/recycling operators. Furthermore, as shown in Table 4, GWP target values and target years are established for manufacturers and importers of products (designated products) such as household air conditioners, and a system requiring reduction of GWP of products which use fluorocarbons has been introduced.

Table 3 Initiatives of Emission Control of fluorocarbons by Each Responsible Party

Responsible party	Details of initiative		
Manufacturers of fluorocarbon	Substantially phase-down fluorocarbons		
Equipment/product manufacturers	Advance reduction of GWP in products which use fluorocarbons and elimination of fluorocarbons in products		
Equipment users	Prevention leakage of fluorocarbons during use of commercial refrigeration and air conditioning equipment		
Filling/Collecting operators	Filling by registered businesses and recycling by licensed businesses		
Destroying/Recycling operators	Recycling and destruction of fluorocarbons according to standards, and prohibition reckless discharge of fluorocarbons		

Table 4 GWP Target Values Required by Manufacturers and Importers for Each Product

Classification of designated product	Main refrigerant currently used, and GWP	GWP target value	Target year
Room air conditioners (excluding wall penetration type air conditioners, etc.)	R410A(2090) R32(675)	750	2018
Shop/office-use air conditioners (excluding floor type air conditioners, etc.)	R410A(2090)	750	2020
Automobile air conditioners (limited to those installed in passenger vehicles (excluding those with a seating capacity of 11 persons or higher))	R134a(1430)	150	2023
Condensing units and stationary type of freezing and refrigeration units (excluding those with compressors having a rated power of 1.5 kW or less)	R404A(3920) R410A(2090) R407C(1774) CO <sub>2</sub> (1)	1500	2025
Centralized freezing and refrigeration equipment (limited to equipment shipped to newly established freezing and refrigeration warehouses having an area of 50,000 m2 or higher)	R404A (3920) ammonia (1)	100	2019
Heat insulating materials using rigid urethane foam (limited to housing construction materials among those for in-situ foaming)	HFC-245fa(1030) HFC-365mfc(795)	100	2020
Sprayers filled exclusively with propellants (excluding those whose use requires non-flammability)	HFC-134a(1430) HFC-152a(124) CO <sub>2</sub> (1) DME(1)	10	2019

### II. NEDO R&D Projects

### **Development of HFC-23 Destruction Technology**

#### Completed NEDO Projects

**Entrustment** 

Japan Environmental Management Association for Industry (Asahi Glass Co., Ltd. (now AGC Inc.) and Daikin Industries, Ltd.)
Furnace body designer/manufacturer: Nittetsu Chemical Engineering Ltd. (now Tsukishima Kankyo Engineering Ltd.)

**R&D Period** 

FY1998-FY2001

#### Summary

An effluent/waste gas disposal facility (submerged combustion system) that destroys ozone-depleting substances such as CFCs and HCFCs as well as HFCs, PFCs and SF6, substances also known as fluorinated greenhouse gases, was developed for commercialization under this project (Figure 1). The facility also enabled the reuse of recovered fluorine.

The facility decomposes HFC-23 (trifluoromethane:CHF<sub>3</sub>) and prevents, as much as possible, the secondary emergence of harmful substances such as dioxins. HFC-23 is a major fluorinated greenhouse gas produced as a byproduct during the manufacture of HCFC-22 (chlorodifluoromethane: CHCIF<sub>2</sub>), which is a refrigerant and is also used as feedstock for resin. Following pyrolytic decomposition, the system recovers HFC-23 as harmless calcium fluoride. It is now possible to dispose of any fluorine-containing effluent or waste gas.

Figure 1 Fluorinated Gas Disposal Facilities Using Submerged

Combustion Method

(a) Submerged combustion furnace (Yodogawa Plant, Daikin Industries, Ltd.) (b) Post-treatment facility (Kashima Plant, Asahi Glass Co., Ltd.)

Through the destruction process, as illustrated in Figure

2, fluorine- and chlorine-containing effluents and waste gases can be completely decomposed of at temperatures of 1,200°C or higher (Figure 3). The system instantaneously cools high-temperature combustion gas using the submerged combustion method (Figure 4), and the hydrogen fluoride and hydrogen chloride generated are treated, respectively, in water absorption and alkali washing towers.

#### **Contribution to Addressing Global Warming**

Tsukishima Kankyo Engineering Ltd., Asahi Glass Co., Ltd. and Daikin Industries, Ltd. have promoted the development of equipment to dispose of fluorine- and chlorine-containing effluents and waste gases discharged from fluorinated gas manufacturing processes. They have also developed technology and have launched facilities for the pyrolytic treatment of organochlorine waste to recover hydrochloric acid, building a technological base to address the combustion of halogenated substances. As a result of the Development of HFC-23 Destruction Technology project, which was entrusted by NEDO and carried out from 1998 to 2001, these companies succeeded in disposing of large quantities of HFC-23 through ongoing operation of the technology.

The developed technology features high-temperature decomposition and a submerged combustion method to treat HFC-23 and restrict the secondary emergence of substances such as dioxins as much as possible, thereby enabling the disposal of any fluorine-containing waste, such as HFCs, PFCs, SF<sub>6</sub> and NF<sub>3</sub>, as well as CFCs and HCFCs. In addition, halogen resistantmaterials to prevent damage to plant equipment were identified. After the project, the two companies that built the facility successfully treated the CO<sub>2</sub> equivalent of approximately 6.9 million tons of the three fluorinated gas substitutes in 2007, thereby contributing to the mitigation of global warming.

A total of 21 (14 in Japan and eight overseas, of which three are related to the Clean Development Mechanism (CDM)) of these decomposition facilities have been constructed, establishing a safe and reliable dedicated combustion furnace to destroyfluorinated gases. The developed equipment is described as a submerged combustion facility in Article 14, of the Law Concerning the Recovery and Destruction of Fluorocarbons, under the category of fluorinated gas destruction facility. Those facilities' disposal capacity per plant is the highest in Japan.

According to the Ministry of Economy, Trade and Industry (METI), the amount of fluorinated gas destroyed based on this law reached 4,161 tons in FY2008.

A fluorinated gas destruction system utilizing the submerged combustion method has a large disposal capacity compared with other systems as exclusive combustion furnaces are employed. This type of system constitutes the majority of fluorinated gas destruction systems constructed in Japan.

In order to protect the Earth's environment, it is necessary to use centralized facilities to safely decompose of large quantities of ozone-depleting controlled substances and the three fluorinated gas substitutes that contribute to global warming. The introduction of the submerged combustion method to centralized facilities significantly contributes to the protection of the environment, and such facilities are expected to be used even more in the future due to recycling measures such as the Home Appliance Recycling Law and Automobile Recycling Law.

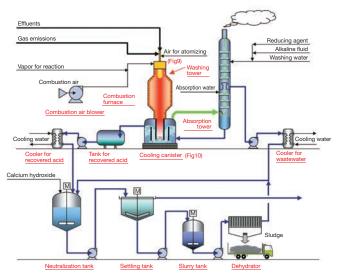


Figure 2 Process Flow of Fluorinated Gas Destruction System





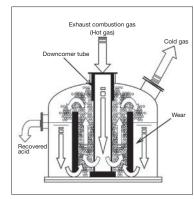


Figure 4 Structure of Cooling Canister

#### **Status of Practical Application**

The fluorocarbons destruction facility developed in this project, as a target of the Clean Development Mechanism (CDM), has expanded to outside Japan, starting with China and South Korea, approximately 30 installations of fluorocarbons destruction facility have been performed at fluorocarbons manufacturing plants, and it greatly contributes to the reduction of greenhouse gases. Also, this equipment can decompose various types of fluorine-containing gases and waste liquids of sulfur hexafluoride (SF<sub>6</sub>), which has a high greenhouse effect similar to HFC-23, as well as CFCs, HCFCs, HFCs, and PFCs. Also, the facility is useful for treatment and recovery of both fluorocarbons and greenhouse gases.

#### **Awards**

- President's Prize, Japan Society of Industrial Machinery Manufacturers
   30th Excellent Environmental Equipment Award, 2004
   CFC Destruction Equipment, Nittetsu Chemical Engineering, Ltd.
   Sponsor: Japan Society of Industrial Machinery Manufacturers
   Sponsors:Ministry of Economy, Trade and Industry, Small and Medium Enterprise Agency
- Ozone Layer Protection/Global Warming Protection Award 8th Economy Trade and Industry Minister's Award, 2005
   Asahi Glass Co., Ltd. and Daikin Industries, Ltd., Tsukishima Nittetsu Chemical Engineering Ltd. (now Tsukishima Kankyo Engineering Ltd.)
   Sponsor: Nikkan Kogyo Shimbun, Ltd.
   Sponsors: Ministry of Economy, Trade and Industry and the Ministry of the Environment



### **Development of Technology for Chemical Recycling of HCFC Refrigerants**

#### Completed NEDO Projects

**Grant awards** Asahi Glass Co., Ltd. (now AGC Inc.), Mitsubishi Electric Corporation

**R&D Period** FY2000

#### Summary

Chemical recycling technology for HCFC-22, a fluorocarbon refrigerant used in residential air-conditioners, was developed as apractical application for 3R technology, which promotes the resolution of issues related to the implementation of the Home Appliance Recycling Law as part of an effort to establish a recycling-oriented society.

Under this development project, HCFC-22 recovered from residential air-conditioners was used as feedstock for fluororesin, making it possible to reduce HCFC-22 production and the industrial waste generated when HCFC-22 is recycled. Asahi Glass Co.,Ltd. undertook the development of fractionating and resinification facilities based on technology that refines recovered HCFC-22 to a purity level of 99.95%. Mitsubishi Electric Corporation was responsible for the development of recovery technology and construction of the facilities.

#### Technical Contents

More than 800 tons of HCFC-22 (R22) used as refrigerant are recovered annually. Although production of this refrigerant is allowed until FY2020 under Japan's Ozone Layer Protection Law, 35% reductions consumption have been required since 2004.

At the time the project was started, recovered refrigerants were being destroyed and detoxified (neutralized) using pyrolytic decomposition and the resulting chemicals, such as CaF<sub>2</sub>, were being buried as industrial waste.

However, since CaF2, a feedstock for HCFC-22, is produced in limited geographic areas and could be depleted in the future, the recycling of HCFC-22 is an important technological development for the practical application of 3R technology.

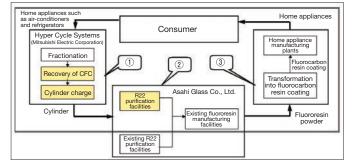


Figure 1 Scope and Concept of Development

The development project resolved the following issues through the application of manufacturing technologies developed by Asahi Glass Co., Ltd. for fluorocarbon refrigerants, including HCFC-22 and fluororesin, and home appliance recycling technology developed by Mitsubishi Electric Corporation (Figure 1).

- ①Efficient storage of recovered refrigerants
  - Development of filling equipment that specifically controls azeotropic mixtures and facilitates the recovery and transfer of sufficient volumes of refrigerants to supply purification facilities
- ②Using purified recovered refrigerants as feedstock for fluororesin
  - Design and construction of a facility to purify recovered refrigerants and implementation of a purification testing method for recovered refrigerants and fluororesin manufacturing tests incorporating an existing manufacturing facility
- 3 Using fluororesin produced from recovered refrigerants for home appliances
  - Application of recycled HCFC-22 to produce fluororesin for use in home appliances, taking advantage of its separability and antifouling properties.

Based on the above, a system with the features described below was established:

#### (1) Refrigerant recovery system

An overview of a system, from recovery to transfer/filling, that was installed at Hyper Cycle Systems' appliance recycling plant to recover refrigerants from air-conditioners is shown in Figure 2.

In the system, the purity of recovered refrigerants was measured to confirm that the purity of R22 and R12 was within standards.R22 significantly impacts purification quality and the combination of R12 and R22 forms an azeotropic mixture.

Refrigerants that met standards were then transferred into a large cylinder. This process substantially increased the acceptable amount of refrigerants recovered at the recycling plant.

### (2) Refrigerant purification facilities (Figures 3 and 4)

Based on the current results of refrigerant analysis conducted at fluorocarbon refrigerant recovery stations and taking into consideration the outlook for such refrigerants, it was determined that R410A (a mixture of R32 and R125), a new

low-boiling refrigerant for residential air-conditioners, and R134a, a new high-boiling refrigerant for refrigerators, needed to be removed. In addition, research on azeotropic mixtures identified that R115 and R12 also need be removed. In particular, since R12 has been used as a refrigerant for refrigerators, it can be recovered from recycled refrigerators and mixed at fluorocarbon refrigerant recovery stations.

Since high-boiling substances that are highly explosive in the fluororesin manufacturing process (for example, R1112 and R1113) can be generated in large quantities in the presence of a high concentration of R12, it is necessary to maintain an R12 concentration in purified refrigerants lower than the control value. Purification and fluororesin manufacturing tests were conducted using refrigerants recovered by a recovery system that was newly installed at Hyper Cycle Systems. After removing residue (mainly oil) and moisture from recovered refrigerants, R32 and R125, which have lower boiling points than R22, as well as R12 and R134a, which have higher boiling points than R22, are subsequently removed through a distillation process, thereby resulting in R22 containing 180 ppm of R12. A fluororesin manufacturing test was then conducted at an existing fluororesin manufacturing facility using the R22 obtained.

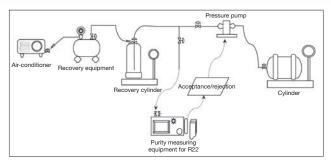


Figure 2 Basic Concept of Recovered Refrigerant Storage Facility



Figure 3 Coolant Purification Facility

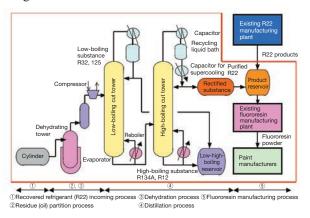


Figure 4 Process Flow of Recovered CFC Purification Facility

#### (3) Manufacturing of resin and performance evaluation

A product made with PFA fluororesin (a copolymer resin of 4-fluorinated ethylene and perfluoroalkoxyethylene), using R22 derived through the above process, was compared with currently available products. The comparison showed that the performance levels of both products were equal.

#### **Contribution to Addressing Global Warming**

After the project, full-scale recovery, transfer and filling systems for recovered refrigerants were established, and related purification facilities and application for coating materials are contributing to efforts to counter ozone depletion and global warming, as well as playing a role in boosting home appliance recycling at the same time.



# Development of Energy-saving Synthetic Technologies for Fluorocarbon Replacements

#### Completed NEDO Projects

Entrustment Asahi Glass Co. (now AGC Inc.), Ltd., Daikin Industries, Ltd., Central Glass Co., Ltd., ZEON Corporation,

Tosoh F-Tech, Inc. (now Tosoh Finechem Corporation), Japan Aluminium Association (JAA), Nagaoka University of Technology, Chiba Institute of Technology, Tosei Co., Ltd. (now STG Co., Ltd.), Ahresty Co., Ltd.

Re-entrustment National Institute of Advanced Industrial Science and Technology (AIST), Ulvac, Inc., Tohoku University

**R&D Period** FY2000-2006

#### Summary

The aim of this project was to develop energy-efficient, industrially effective synthesis technology, and thereby contribute to decreasing the burden on the environment by reducing energy consumption. The project explored and reviewed industrial processes to synthesize fluorinated gas substitutes. Fluorinated gas substitutes are widely used in the industrial sector and cause less damage to

Table 1 Industrial Applications for New Fluorinated Substitutes

Industrial Application	New Fluorinated Substitutes	
Refrigerant	HFE-143m	
Industrial detergent	HFE-347pc-f	
Foaming agent (for in-situ foaming)	HFE-254pc	
Semiconductor/LCD manufacturing	CxFy, CF3I, COF2	
Electrical equipment insulation	CF <sub>3</sub> I	
Extinguishing agent	CF3I	
Cover gas for magnesium die-casting	CF <sub>3</sub> I, HFO-1234ze(E)	

the ozone layer, do not exacerbate the greenhouse effect, and have less impact on the environment overall. Applications include refrigerants (for refrigerators, vehicle air-conditioners, etc.), industrial detergents (for electronic parts, high-precision processing parts, optical parts, etc.), foaming agents (for in-situ foaming), semiconductor and liquid crystal manufacturing (LCDs for etching, CVDchamber cleaning, etc.), electrical insulating equipment, extinguishing agents, and magnesium manufacturing (Table 1). The following shows an example of energy-saving synthetic technologies for fluorocarbon replacements.

#### **Technical Contents**

#### CF<sub>3</sub>I synthesizing technique

Tosoh-F-Tech Inc. successfully developed a synthesizing process to produce iodotrifluoromethane (CF<sub>3</sub>I, also known as trifluoromethyl iodide and trifluoroiodomethane) on a commercial scale by directly reacting trifluoromethane (CHF<sub>3</sub>) and iodine in the presence of a catalyst. Iodotrifluoromethane is a gas which has a very low GWP equivalent to that of CO<sub>2</sub> and is expected to serve a number of purposes, including use as a PFC substitute gas for manufacturing semiconductors and liquid crystals. The process developed for producing iodotrifluoromethane is expected to reduce CO<sub>2</sub> emissions by approximately 40% compared to conventional production processes.

#### Application of CF<sub>3</sub>I as an etching gas for manufacturing semiconductors\*

Iodotrifluoromethane, which has a GWP that is 1/1000 that of conventional chlorofluorocarbon alternatives, has been used as a plasma dry etching gas for manufacturing semiconductors. In the process of manufacturing semiconductors compatible with 32-45nm-generation process technology, it was discovered that the use of CF<sub>3</sub>I resulted in a reduction of line edge roughness and an improvement in wiring reliability compared to products manufactured with conventional alternatives.

It was also demonstrated that the use of CF<sub>3</sub>I combined with exposure to short wavelength extreme ultraviolet (EUV) light in the etching process is effective for manufacturing semiconductors compatible with next-generation 22 nm chip technology (Figure 1).

This project aimed to accelerate the practical application of CF<sub>3</sub>I to next-generation semiconductor processing technology.

\*Although this research was concluded in FY2006, it has been continued by Semiconductor Leading Edge Technologies, Inc. in its Etching Performance Evaluation Using New CFC Substitutes project.

#### Application of CF<sub>3</sub>I and HFO-1234ze(E) as cover gases for magnesium die casting

Magnesium is an element that is widely used due to its much lighter weight and higher specific strength relative to iron, as well as the ease with which it can be recycled. The use of cover gas in die casting, the main method used to manufacture magnesium products, prevents the surface of molten magnesium in a melting furnace from being exposed to air, thereby suppressing high-temperature oxidation (combustion). SF<sub>6</sub> has traditionally been used as the primary cover gas, but due to its extremely high GWP value of 23,900,

the development of substitute gases with lower GWP values is required. In this project, two SF<sub>6</sub> substitutegases, 1,3,3,3-tetrafluoropropene (HFO-1234ze(E)) and iodotrifluoromethane (CF<sub>3</sub>I), were developed. Both have GWP values that are 1/1000 that of SF<sub>6</sub> and they are as nonflammable and effective as SF<sub>6</sub> in suppressing high-temperature combustion of magnesium.

A number of magnesium manufacturers in Japan are already using HFO-1234ze(E), which has contributed to a significant reduction in greenhouse gas emissions.

These gases are expected to serve as SF<sub>6</sub> substitutes for the manufacture of magnesium alloy die cast products and to significantly reduce greenhouse gas emissions.

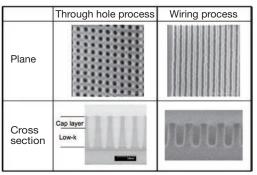


Figure 1 Configuration of Next-generation Semiconductors Produced with CF<sub>3</sub>I Gas

#### **Awards**

2009: 12th Ozone Layer Protection and Global Warming Prevention Award for Excellent Performance

### **Development of Chlorine Fluorinated Gas Substitutes**

#### Completed NEDO Projects

Entrustment

Japan Environmental Management Association for Industry, ZEON Corporation, National Institute of Materials and Chemical Research (now the National Institute of Advanced Industrial Science and Technology)

**R&D Period** 

FY1996-FY1997

#### Summary and Technical Contents

The Montreal Protocol requires that the production of ozone-depleting substances such as chlorofluorocarbons (CFCs) be phased-out in order to protect the ozone layer. In accordance with the ratification of the Kyoto Protocol, Japan is also obligated to reduce its greenhouse gas emissions to counter global warming, reinforcing the need to shift to fluorinated gas substitute compounds.

This project established industrial synthesis technology for CFC and HCFC substitutes with lower ozone depletion potential and lower GWP. Specifically, it has become possible to easily form two environmentally benign five-membered ring fluorine compounds, octafluorocyclopentene (Figure 1) and heptafluorocyclopentene (Figure 2), in large quantities through improved yields using a synthesis method with hydrogen fluoride.

Application:
Dry etching gas

Application:
Industrial detergent

Figure 1 Octafluorocyclopentene

Figure 2 Heptafluorocyclopentene

It also has been discovered that these compounds can be applied as gases for manufacturing semiconductors and LCDs as well as industrial detergents as a substitute for organochlorine compounds that have been conventionally used but which have an adverse impact on the environment. Such industrial applications were developed by studying various types of data on compound properties, including global environmental impact, physicochemical constants, stability, impact on materials and the results of safety tests conducted in accordance with the Act on the Evaluation of Chemical Substances and Regulation of Their Manufacture, Etc.

#### **Contribution to Addressing Global Warming**

ZEON Corporation is mass producing the two chemical compounds developed in this project. A survey that it conducted showed that octafluorocyclopentene accounts for more than half of the global market for dry etching gases (contact hole size:100-200 nm), and that the market for heptafluorocyclopentene as an HCFC substitute detergent has expanded. Synthesis methods and applications for these two new CFC and HCFC substitutes were developed in Japan ahead of other countries and have attracted attention overseas.

#### **Awards**

1998: 1998 Environmental Protection Agency (EPA) Stratospheric Ozone Protection Award

2000: 8thChemical and Biotechnology Tsukuba Prize, Tsukuba Foundation for Chemical and Biotechnology

2000: 32nd JCIA Award for Technological Excellence from the Japan Chemical Industry Association

2003: 2ndGreen and Sustainable Chemistry Award, Minister of the Environment

Green & Sustainable Chemistry Network, Japan (GSCN)

2008: 11th Ozone Layer Protection and Global Warming Prevention Award for Excellent Performance



# R&D of SF<sub>6</sub> Substitute Gas Cleaning System for Electronic Device Manufacturing

#### Completed NEDO Projects

#### **Entrustment**

Research Institute of Innovative Technology for the Earth (RITE), Asahi Glass Co., Ltd. (now AGC Inc.), Kanto Denka Kogyo Co., Ltd., Showa Denko K.K., Daikin Industries, Ltd., Canon Anelva Corporation, Hitachi Kokusai Electric Inc., Ulvac, Inc., Tokyo Electron Limited, Fujitsu Limited, Hitachi, Ltd., Matsushita Electric Industrial Co., Ltd. (now Panasonic Corporation), Toshiba Corporation, Mitsubishi Electric Corporation, Oki Electric Industry Co., Ltd., Sony Corporation, NEC Corporation, Sanyo Electric Co., Ltd. (now Panasonic Corporation), Sharp Corporation, Semiconductor Leading Edge Technologies, Inc. (SELETE), Japan Electronics and Information Technology Industries Association (JEITA)

Joint Research / Re-entrustment

National Institute of Advanced Industrial Science and Technology (AIST), Ibaraki University, Anelva Corporation, Central Glass Co., Ltd.

**R&D Period** 

FY1998-2002

#### Summary

The following studies and research and development were carried out with the goal of developing gas for chemical vapor decomposition (CVD) cleaning with a lower environmental burden, including a lower GWP:

- 1) Study on the basic performance of reaction gas for cleaning
- 2) R&D on new substitute gases for CVD
- 3) R&D on CVD equipment using new substitute gases
- 4) Comprehensive evaluation study

Corrosion and the durability of CVD chambers and materials for exhaust equipment when SF<sub>6</sub> substitute gases are used were evaluated, and research was conducted on how to improve CVD cleaning efficiency and reduce greenhouse gas emissions. Research was also conducted on new substitute cleaning gases

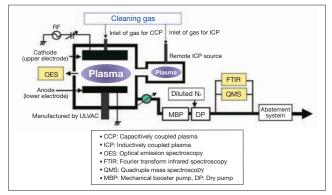


Figure 1 Schematic Diagram of Experiment Facility

in an effort to reduce greenhouse gas emissions. In addition, a prototype plasma CVD apparatus was developed for the semiconductor manufacturing process (Figure 1).

#### **Technical Contents**

Since gases with high GWP values are currently being used for semiconductor manufacturing, the resulting emissions need to

be reduced as soon as possible to protect the global environment. A BAU estimate of greenhouse gas emissions from semiconductor manufacturing, lower actual emissions because of this project, and a 2010 target are shown in Figure 2. It was estimated that emissions would have increased 10% a year and would have amounted to about four times their current level if no measures had been taken. As shown in Figure 2, emissions in 2001 were approximately five million tons, about the same as the level in the base year of 1995.

Japan's voluntary target was to reduce its emissions by 10% or more by 2010 through such measures as increasing installations of abatement equipment, utilizing substitute gases, optimizing processes and adopting new processes. (Figure 2)

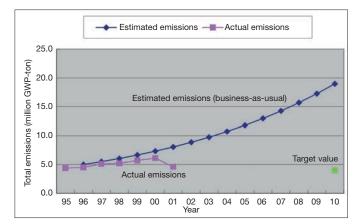


Figure 2 Estimated Emissions, Actual Emissions and Targeted Value for 2010

New substitute gases that can be used in the processes for manufacturing electronic devices such as semiconductor ICs and LCDs have been developed in order to reduce the use of fluorinated substitute gases and the emission of greenhouse gases with high GWP values, such as SF<sub>6</sub>. More specifically, the gases are designed to be used in the plasma CVD cleaning process for insulation film.

Various gases were evaluated as substitute cleaning gases for CVD equipment. It became clear that COF<sub>2</sub> (carbonyl fluoride) can reduce greenhouse gas emissions by 99% or more (Table 1) while retaining a cleaning speed (etching rate) equivalent to that of C<sub>2</sub>F<sub>6</sub> (Figure 3).

This is because the GWP100 of COF<sub>2</sub> is low and its by-products contain only trace amounts of high GWP gases. Moreover, COF<sub>2</sub> has an advantage in that it does not require special abatement equipment due to its high reactivity with water.

A reduction of greenhouse gas emissions and the total cost of CVD cleaning systems is possible by using gases, with comprehensive safety measures, that are reactive and that have superior cleaning ability, such as COF<sub>2</sub>, rather than gases that are stable in the atmosphere, such as conventional PFC or SF<sub>6</sub>. In addition, an ongoing evaluation of the cleaning properties of COF<sub>2</sub> showed no increased particles and stable deposition. This suggests potential for application to the mass production of semiconductors.

With regard to F<sub>2</sub>, it was discovered through an evaluation of its basic properties that it has superior cleaning properties and results in almost no greenhouse gas emissions and is therefore more environmentally friendly. However, there remain some issues regarding how to supply and handle F<sub>2</sub>, making it difficult to apply F<sub>2</sub> to large-scale facilities.

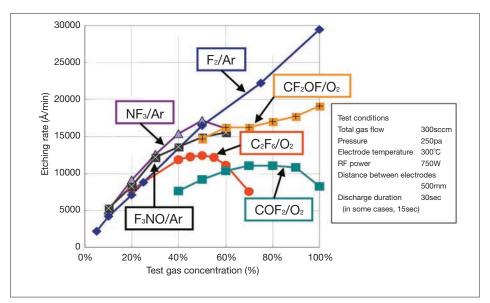


Figure 3 Relationship between Etching Rate and Gas Concentration

Table 1 Comparison	of GHG Emissions	Relative to C <sub>2</sub> F <sub>6</sub>
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Technologies	Emissions	
	C <sub>2</sub> F <sub>6</sub>	100%
Existing technologies	C₂F <sub>6</sub> +abatement	23%
3.55.11.51.59.55	NF₃+abatement	0.80%
Innovative technology	COF <sub>2</sub> +abatement	0.30%

<sup>\*</sup>Roughly calculated values when manufacturing gases (gas leakage) and/or in cleaning processes (plasma energy, energy during abatement, gas leakage after abatement)

#### Contribution to Addressing Global Warming

Daikin Industries, Ltd. first produced COF<sub>2</sub> on a commercial basis as a CVD cleaning gas in 2003. Kochi Casio Co., Ltd., an affiliate of Casio Computer Co., Ltd. and a producer of TFT-LCDs, adopted COF<sub>2</sub> as a cleaning gas for its manufacturing-processes in 2005. Kochi Casio received a special award at the 9thOzone Layer Protection and Global Warming Prevention Grand Prix in 2006 for its introduction of COF<sub>2</sub>. In the future, use of COF<sub>2</sub> is expected to expand to the semiconductor and LCD industries.

In addition, in 2008, COF2 has been commercialized by Kanto Denka Kogyo Co., Ltd..



# Development of Non-SF6 Melting Process and Microstructural Control for High- performance Magnesium Alloy

#### Completed NEDO Projects

**Grant Awards** 

Sankyo Tateyama Aluminium, Inc. (now Sankyo Tateyama,Inc.), Sumitomo Electric Industries, Ltd., The Japan Steel Works, Ltd., Daido Steel Co.,Ltd.

R&D Period F

FY2004-FY2006

#### **Summary**

One aim of this project was to develop magnesium processing technology without the use of SF<sub>6</sub> gas, a gas that has an extremely high GWP value of 23,900, by adding calcium to molten magnesium in order to make the alloy, and products containing the alloy, nonflammable. Another aim was to produce magnesium parts that are lighter than conventional aluminum alloys but which have comparable or superior mechanical properties. In order to accomplish these objectives, melting and refining process technology as well as solidification technology that gives a very fine grain microstructure were developed.

#### **Technical Contents**

Melting and refining process technology for magnesium and technology to solidify magnesium alloys without the use of SF<sub>6</sub> gas as well as molding process technology that improves the mechanical properties of magnesium alloy (Figure 1) were developed in this project.

Through the process of developing melting and refining process technology for magnesium, the optimum amount of calcium to be added to molten magnesium was identified, eliminating the need to use SF<sub>6</sub> gas. In addition, industrial melting process technology for magnesium alloys containing calcium, impurity/inclusion removal and analysis technologies, and crystal grain refinement technology for manufacturing billet were established. The conditions necessary for producing actual components were also clarified.

The development of molding process technology established production methods for specific components and products by developing high-toughness expansion process technology, including extruding, cupping and rolling of magnesium alloys containing calcium, high creep resistance injection-molding process technology using particle composites of magnesium and reinforced materials, and high rigidity combined processing technology.

This project contributed to the practical use of magnesium alloy as a structural material for motorcycles and expanded structural materials for railcars and health-care products, as well as the production of welding rods and screws that connect structural components (Figure 2). Application of the developed materials to the production of motorcycles, railcars and automobiles will result in lighter weight

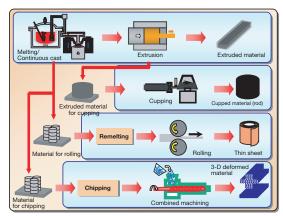


Fig. 1 Production of Materials with Non-SF Melting Process



Fig. 2 Applications for Materials Developed in this Project

transport vehicles, lower energy consumption and, therefore, reduced CO2 emissions.

#### **Awards**

2009: Toyama Alloy, an affiliate company of Sankyo Tateyama Aluminum, Inc., won the 12th Ozone Layer Protection and Global Warming Prevention Award for Excellent Performance (sponsored by Nikkan Kogyo Shimbun Ltd. and supported by the Ministry of Economy, Trade and Industry and the Ministry of the Environment).

### Project to Support the Practical Implementation and Application of Emission Control Equipment to Control Three Fluorinated Gas Substitutes

#### Completed NEDO Projects

**Grant Awards** 

Asada Co. Ltd., Iceman Co. Ltd., Kohji Corporation, NKK Co. Ltd., Kanto Denka Kogyo Co., Ltd., Consumers Cooperative Co-op Sapporo, Nikkin MgCast Co. Ltd., and the others (82 Companies in all)

**R&D Period** 

FY2006-2010

#### **Summary**

Ozone-depleting substances such as controlled substances (CFCs and HCFCs) are required to be phased out in order to protect the ozone layer under the terms of the Montreal Protocol. For this reason, three fluorinated gas substitutes (HFCs, PFCs and SF6) that do not deplete the ozone layer were developed as alternatives to controlled substances. They have been used as refrigerants (for freezers/refrigerators, air-conditioning equipment and vehicle air-conditioners), foaming agents, detergents, insulation, etc., due to their useful properties. The use and emissions of these substitute gases are expected to increase as ozone-depleting substances are

phased out. However, since these three substitute gases can stably exist in the atmosphere for a long period of time and because they have an extremely high GWP value, emissions resulting from their use had to be reduced in accordance with the terms of the Kyoto Protocol.

In this project, advanced and broadly applicable equipment and technology development proposals related to emission reduction in all fields and industry sectors that use the three fluorinated gas substitutes were solicited. Outstanding proposals were then subsidized as leading model projects (applied research at an advanced stage) in order to promote practical application (see Figure 1).

Until FY2007, this project was known as the Project to Support the Practical Implementation and Application of Emission Control Equipment and Facilities to Control Three Fluorinated Gas Substitutes.

The following shows some of the results of the project (Figures 2 to 4).

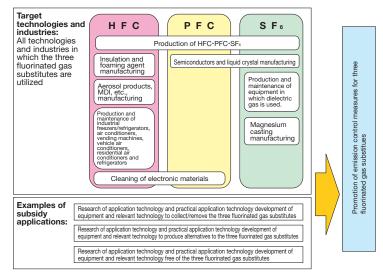


Figure 1 Project Overview



Figure 2 Project result 1:Fluorocarbon-free air duster production facility (NKK Co., Ltd.)



Figure 3 Scope and Concept of Development



Figure 4 Scope and Concept of Development

#### **Awards**

- 2008: NKK Co., Ltd. was awarded the Economy, Trade and Industry Minister's Award at the 11th Ozone Layer Protection and Global Warming Prevention Award for Excellent Performance ceremony.
- 2011: COOP Sapporo was awarded the Economy, Trade and Industry Minister's Award at the 14th Ozone Layer Protection and Global Warming Prevention Award ceremony for the introduction of non-fluorinated showcases.
- 2012: Lawson, Inc.was awarded the Economy, Trade and Industry Minister's Award at the 14th Ozone Layer Protection and Global Warming Prevention Award ceremony for its installation of refrigeration systems at convenience stores.

<sup>\*</sup>Sponsored by the Nikkan Kogyo Shimbun Ltd. and supported by the Ministry of Economy, Trade and Industry and the Ministry of the Environment.



# Development of Non-fluorinated Energy-saving Refrigeration and Air-conditioning Systems

#### Completed NEDO Projects

**Entrustment** 

Shin Nippon Air Technologies Co., Ltd., Chubu Electric Power Co., Inc., Mitsubishi Heavy Industries, Ltd. (now Mitsubishi Heavy Industries Thermal Systems, Ltd.), Honda R&D Co., Ltd., The Japan Refrigeration and Air Conditioning Industry Association (JRAIA), National Institute of Advanced Industrial Science and Technology (AIST), The University of Tokyo, Kyushu University, and the others

**Grant Awards** 

Daikin Industries, Ltd., Sinko Industries, Ltd., Mitsubishi Electric Corporation, Panasonic Corporation, Sanden Corporation (now Sanden Holdings Corporation), MAC Co., Ltd., Mitsubishi Heavy Industries Air-Conditioning & Thermal Systems Corporation (now Mitsubishi Heavy Industries Thermal Systems, Ltd.), Maekawa Mfg. Co., Ltd., General Heat Pump Industry Co., Ltd., Sanyo Electric Co., Ltd., (now Panasonic Corporation), and the others

**R&D Period** 

FY2005-FY2010

#### Summary

The production and use of refrigerants such as CFCs and HCFCs are to be phased out in accordance with the Montreal Protocol's control measures to protect the ozone layer. Because of this, refrigerants that use fluorinated gas substitutes were developed. The air-conditioning and refrigeration industry promptly responded, and their most common models now employfluorinated gas substitutes. Some of these substitute compounds, however, have extremely high GWP values and the emission of these gases must be reduced in accordance with the Kyoto Protocol.

Although some air-conditioners that use fluorocarbon-free refrigerants that have low GWP have been commercialized, they are not widely used yet due to low energy efficiency and because of safety concerns, including worries about the potential for

refrigerant leakage. Moreover, it is a technical challenge to utilize fluorocarbon-free refrigerants in air-conditioners and research is still, therefore, ongoing. In order to realize commercialization, it will be necessary not only to develop elemental equipment but also a safe, highly energy-efficient system.

NEDO carried out a project to promote the improvement and development of safe and comfortable refrigeration and air-conditioning systems using fluorocarbon-free substances that do not damage the ozone layer, have a low impact on the environment and a low GWP, while aiming at further reducing the overall environmental impact from the viewpoint of energy saving (see Figure 1).

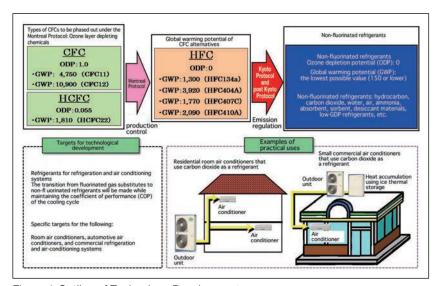


Figure 1 Outline of Technology Development

#### **Technical Contents**

Sanden Corporation, a participant in this project, successfully developed a refrigeration and air-conditioning system for convenience stores. This system employs ammonia (NH<sub>3</sub>), which does not deplete the ozone layer and has no global warming effect, as a refrigerant. The greatest challenge in using ammonia is its odor and toxicity. Sanden enhanced the safety of the system by minimizing the use of ammonia, completely sealing the ammonia in an outdoor unit, and eliminating the use of ammonia inside convenience stores.

Based on a final verification using a laboratory with a built-in full-scale small store, the new system will improve the energy efficiency of convenience stores by approximately 21%. Verification tests were also conducted at a number of convenience

stores in Japan. The new system is expected to be introduced to approximately 42,000 stores nationwide, which will reduce CO<sub>2</sub> emissions by as much as 640,000 tons per year, thereby contributing to the mitigation of global warming. In addition, the world's first non-fluorinated refrigeration and air-conditioning system for convenience stores was put on the market in FY2009.

SANYO Electric Co., Ltd., another participant in this project, developed Japan's first refrigeration system using natural refrigerants (CO<sub>2</sub>) for use in refrigerated showcases in supermarkets (Figure 2). Though it is neither toxic nor flammable, CO<sub>2</sub> as a refrigerant has some drawbacks. It is less efficient and requires a higher working pressure compared to HFCs. In particular, in summer, when ambient temperatures exceed 30°C, it is not possible to attain a highly-efficient refrigeration cycle with CO<sub>2</sub> refrigerant. To overcome this, SANYO developed a circuit cycle that employs CO<sub>2</sub> refrigerant to achieve a high-efficiency refrigeration cycle. SANYO succeeded in developing the system without having to make special modifications by employing, in the newly developed cycle, a two-stage rotary CO<sub>2</sub> compressor. In addition, because CO<sub>2</sub> refrigerant has a high heat transfer capacity, it is possible to use smaller diameter pipes when building systems. As a result, the weight of copper piping used in the system can be reduced by up to 37%, thereby saving resources.

Demonstration experiments of the system, which can reduce power consumption by approximately 10% compared to conventional systems, were confirmed at supermarkets. In addition, considering the direct impact on power consumption reduction and the indirect impact on refrigerant leakage, the system can reduce CO<sub>2</sub> emissions by a maximum of approximately 60% (Figure 3). SANYO Electric introduced the system to the market in FY2010.

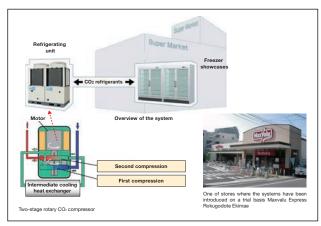


Figure 2 Development of Refrigeration Systems for Supermarkets Equipped with CO<sub>2</sub> Refrigerants

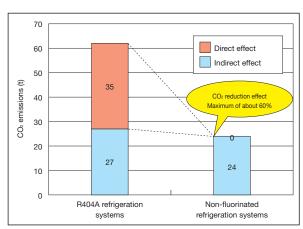


Figure 3 Forecast of CO<sub>2</sub> Emissions Reduction Effect Resulting from CO<sub>2</sub> Refrigeration Systems

#### Status of Practical Application

This project confirmed the energy saving and the CO<sub>2</sub> emission reduction performance through the development of an energy-saving freezing and refrigeration showcase in which the conventional HFC refrigerant was replaced with CO<sub>2</sub> refrigerant and the establishment of basic technologies for solving the technological problems present when CO<sub>2</sub> is used as the refrigerant. Furthermore, NEDO has deployed CO<sub>2</sub> refrigerant freezing and refrigeration showcase using this system to actual shops and has provided support for technological demonstration of solutions and the like to the technological problems for assuring reliability, improving performance, and performing expansion in accordance to the operating conditions of field circumstances. This system, as a result of support projects conducted thereafter by the Ministry of Economy, Trade and Industry and the Ministry of the Environment, exhibits a massive expansion, primarily at convenience stores and supermarkets.

#### **Awards**

- 2009: Sanden Corporation was awarded the Economy, Trade and Industry Minister's Award at the 12th Ozone Layer Protection and Global Warming Prevention Award ceremony\* for its development and practical application of energy-efficient refrigeration and air-conditioning systems equipped with non-fluorinated refrigerants for use at small stores.
- 2010: SANYO Corporation was awarded the Economy, Trade and Industry Minister's Award at the 12th Ozone Layer Protection and Global Warming Prevention Award ceremony\* for refrigeration and air-conditioning systems equipped with non-fluorinated refrigerants for use at small stores.

<sup>\*</sup>Sponsored by the Nikkan Kogyo Shimbun Ltd. and supported by the Ministry of Economy, Trade and Industry and the Ministry of the Environment.



# Project to Develop Innovative Non-Fluorocarbon Heat Insulation Technology

#### Completed NEDO Projects

**Entrustment** 

Kyoto University, National Institute of Advanced Industrial Science and Technology, Nisshinbo Chemical Inc., C. I. Kasei Co., Ltd., Tokyo University of Science, Asahi Fiber Glass Co., Ltd., Toray Industries, Inc., Kaneka Corporation, Tokyo Institute of Technology, Japan Testing Center for Construction Materials, Achilles Corporation

**Grant awards** 

Asahi Glass Co., Ltd.(now AGC Inc.), BASF INOAC Polyurethanes Ltd., Achilles Corporation

Joint Research / Re-entrustment

Yamagata University, Hokkaido Northern Regional Building Research Institute

**R&D Period** 

FY2007-FY2011

#### **Summary**

Rigid urethane foam is widely utilized in building structures. However, because insulation and foaming agents made from fluorinated gas substitutes have a high global GWP, there is a need to develop insulation and foaming agents from chemicals with lower GWP values.

In response, non-fluorocarbon insulation and foaming agents utilizing gases with lower GWP values, such as CO<sub>2</sub> or cyclopentane, are being developed.

However, several issues need to be addressed regarding new non-fluorinated foaming agents, such as insulating efficiency, combustibility during manufacturing, and workability, before they can commercialized.

NEDO undertook this project to develop innovative technology for non-fluorocarbon insulation materials used as building insulation materials.

The features of non-fluorocarbon insulation materials are an insulation efficiency equal or superior to conventional rigid urethane foam materials and a high insulation efficiency for a long period of time.

#### **Technical Contents**

The following four major technologies were developed in the project: (Figure1 and Figure2)

- Technology to control high porosity foam structure by mixing micrometer-sized pores and nanometer-sized pores in order to improve insulation efficiency of CO<sub>2</sub> foam insulation materials
- Technology to control the diffusion process of foaming agents contained in pores in order to maintain long-term insulation efficiency
- Synthesis technology to produce HFO\* foaming agents with a low GWP as a substitute for conventionally used foaming agents with a high GWP
- Technology to produce heat insulation materials by hybridizing aerogels which have an extremely high insulation efficiency with polymer

Based on the above-mentioned technology development, there are future prospects for commercialization of non-fluorocarbon heat insulation having the same insulation efficiency as fluorinated foaming agents.

With the aim of evaluating the developed heat insulation materials, two other technologies were developed: technology to measure insulation efficiency and thermal conductivity in order to evaluate developed heat insulation materials and technology to evaluate the practicality of developed heat insulation materials. (Figure2-(4))

\*HFO stands for hydrofluoroolefin. It is fluorine compounds characterized by a double bond with a much lower GWP than that of HFC.

#### **Contribution to Addressing Global Warming**

It is expected that the development of new non-fluorocarbon insulation materials having a high insulating efficiency will reduce CO<sub>2</sub> emissions. In addition, since the development of this technology is expected to benefit not only the construction industry but also other industries that use insulation materials, such as for refrigeration and transportation, the ripple effect is expected to significantly boost climate change prevention efforts.

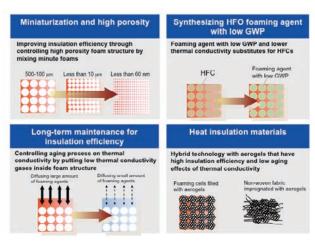


Figure 1 Overview of Technology Development

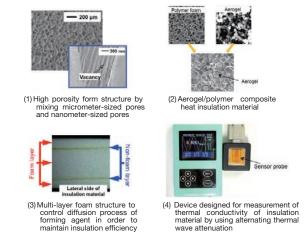


Figure 2 Development Results

# **Technology Development of High-efficiency Non-fluorinated Air-conditioning Systems**

#### Completed NEDO Projects

**Entrustment**Tokyo University of Science, Suwa (now Suwa University of Science), The University of Tokyo, Kyushu
University

Grant awards

Asahi Glass Co., Ltd. (now AGC Inc.), Daikin Industries, Ltd., Panasonic Corporation, Sanden

Corporation (now Sanden Holdings Corporation), Mitsubishi Heavy Industries, Ltd. (now Mitsubishi

Heavy Industries Thermal Systems, Ltd.), Mitsubishi Electric Corporation

Joint Research / Re-entrustment

National Institute of Advanced Industrial Science and Technology (AIST), Tokyo University of Science, Suwa (now suwa University of Science), Saga University, Iwaki Meisei University, Kyushu Sangyo University

**R&D Period** FY 2011-FY2015

#### Summary

In order for the field of commercial air conditioners, in which there is a large potential for reducing greenhouse gases, to realize energy saving and high efficiency using low-GWP gases, development has been conducted for basic technologies which realize performance equal to or greater than commercially available HFC products while using low-GWP gases, by developing new refrigerants, developing component devices such as compressors and heat exchangers, and developing systems.

Furthermore, the study group for the risk evaluation of mildly flammable refrigerants was started as part of the implementation of this project, and it evaluated the safety and risks with regards to various types of mildly flammable refrigerants, and has conducted initiatives for establishing regulations concerning safety and risks according to requirements.

#### **Technical Contents**

This project consists of the following three research and development items, and the primary research results of each item are as follows.

1. Development of primary devices for achieving high performance with low GWP refrigerants (Daikin Industries, Ltd.)

With the aim of achieving performance at the same level of conventional products using R410A refrigerants with cooling rated COP\*1 by using CO2 refrigerants in air conditioners, performance tests were ultimately conducted after installing a novel multi-stage compressor, aluminum micro-channel heat exchanger, composite selector, novel liquid suction heat interchanger, and expander in 5 HP test equipment. The results of calculating system performance of the 10 HP final product based on observations of this test equipment confirmed the anticipation of the achievement of all objectives. Furthermore, cooling rated COP achieved improvements up to 92% compared to R410A products.

However, as increases in efficiency require the installation of numerous internal heat exchangers, oil separators, and expanders, a volume ratio of 146% against the casing of R410A products having the same performance was achieved.

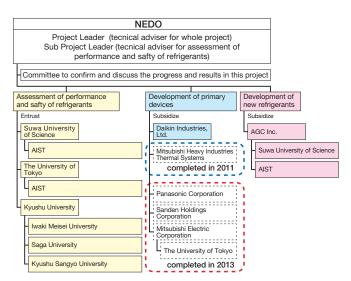


Figure 1 Implementation system of R&D in this project

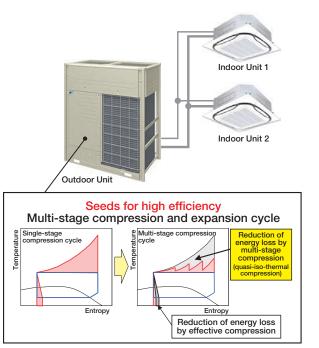


Figure 2 High-efficiency cycle and unit in test operation

<sup>\*1:</sup> COP stands for Coefficient Of Performance. It is indicates the efficiency when an air conditioner is operated at specific temperature conditions.

2. Development of new refrigerants with high efficiency and low GWP (Asahi Glass Co., Ltd.)

NEDO has developed (1) a mixed refrigerant that contains HFO-1123 as an alternative to R410A, and (2) HCFO\*2-1224yd(Z) as an alternative to R245fa. These new refrigerants feature a much lower greenhouse gas effect than conventional refrigerants and the refrigerating performance is equivalent to conventional refrigerants.

\*2 HCFO stands for hydrochlorofluoroolefin. It is fluorine compounds characterized by a double bond with a much lower GWP than that of HFC.

R410A = 100%		HFO-1123/HFC-32/HFO-1234yf (40/44/16%)	HFO-1123/HFC-32 (40/60%)		
COP	In cooling mode	110%	116%		
COP	In heating mode	96%	99%		
APF		96%	97%		
GWP		14%	20%		

Table 1 Basic performance of refrigerants developed

3. Assessment of performance and safety of refrigerants

(The University of Tokyo, Kyushu University, Tokyo University of Science, Suwa)

Low-GWP refrigerants are slightly flammable, which may impede their commercialization. A study group for the risk evaluation of mildly flammable refrigerants was established to serve as a structure for risk evaluation and deliberation in collaboration with industrial, governmental and academic sectors and to collect the findings of safety and risk assessment conducted in NEDO's projects and knowledge of risk assessment by the Japan Refrigeration and Air Conditioning Industry Association. A report from this study group helped to revise the High Pressure Gas Safety Act on November 1, 2016, which led to new provisions for the use of low-GWP refrigerants. In addition, a large capacity centrifugal chiller\*3 using a low-GWP refrigerant was commercialized.

NEDO also developed a method for testing the burning velocity of mildly flammable refrigerants and established a quantitative measurement method to identify the quenching diameter\*4 under conditions of practical use. Therefore, it was discovered that in the event of a spark inside the electromagnetic switch in room air conditioners with a mildly flammable refrigerant, the resulting flame will be extinguished at the opening of the relay cover and will not spread from the cover to the outside if the opening is smaller than the quenching diameter. Based on this result, a revision to the safety requirements of relays in IEC-60335-2-40 (Household and similar electrical appliances - Safety - Particular requirements for electrical heat pumps, air conditioners and dehumidifiers) was proposed and the revised edition was published on January 26, 2018.

The results of the safety assessment are expected to help spread the use of mildly flammable refrigerants and related equipment.

- \*3: The fundamental research and development for this product is based on the results of the Development of the Non-Fluorinated Energy-Saving Refrigeration and Air-Conditioning Systems project for FY2005–FY2010.
- \*4: The quenching diameter is the maximum size of the opening that extinguishes flames that have already started to spread. It is an inverse function of the burning velocity. An opening with a size equivalent to or smaller than this diameter will quench fire and block flame from passing through the opening.

Safety and risk assessment of mildly flammable refrigerants

Revision to the High Pressure Gas Safety Act

Paving the way for handling designated mildly flammable refrigerants in the same manner as non-flammable refrigerants Providing for safe utilization of low GWP mildly flammable refrigerants

Commercialization of a product with low GWP refrigerant



Commercializing a large capacity centrifugal chiller\*1 with HFO-1234ze(E) low GWP refrigerant

Proposal for revision to IEC 60335-2-40

Suggesting a revision concerning mildly flammable refrigerants for the safety requirements of electromagnetic switches (or relays) in IEC 60335-2-40



Revision to the IEC standard

#### [Key points]

- ●The revision stipulates the maximum size of opening for relays and other devices that prevents proliferation of flame from mildly flammable refrigerants
- With respect to burning velocity required to determine the quenching diameter, the impact of humidity must be taken into consideration.
- Electric contacts of relays and other devices with a predetermined capacity will not be regarded as ignition sources for mildly flammable refrigerants.

#### Awards

Ozone Layer Protection/Global Warming Prevention Award

Sponsor: Nikkan Kogyo Shimbun Ltd.

Supporters: Ministry of Economy, Trade and Industry/Ministry of the Environment

2016 19th Economy, Trade and Industry Minister's Award

Project awarded: Risk Evaluation for Proper Use of Mildly Flammable Refrigerants

Recipient: Japan Society of Refrigerating and Air Conditioning Engineers, The Japan Refrigeration and Air Conditioning Industry Association

# Technology Development of Air-Conditioning Systems Using High-Efficiency, Low-GWP Refrigerants

#### Completed NEDO Projects

**Entrustment** 

Tokyo University of Science, Suwa (now Suwa University of Science), Kyushu University, The University of Tokyo, National Institute of Advanced Industrial Science and Technology (AIST)

**Grant Awards** 

Asahi Glass Co., Ltd. (now AGC Inc.), Panasonic Corporation, Mitsubishi Electric Corporation, DENSO Corporation

Joint Research / Re-entrustment

Toyama Prefectural University, Nihon University, National Institute of Advanced Industrial Science and Technology (AIST), Nagasaki University, Kyushu Sangyo University, Saga University, Tokyo University of Marine Science and Technology, Waseda University, The University of Tokyo, Toyohashi University of Technology

**R&D Period** 

FY2016-2017

#### Summary

Converting refrigerants from specified chlorofluorocarbons (CFCs and HCFCs), which are ozone-depleting substances, to fluorinated gas substitutes (HFCs), which do not deplete the ozone layer, has increased the amount of market stocks of fluorinated gas substitutes used in refrigerants of refrigeration and air conditioning equipment such as air conditioners and refrigeration/freezing showcases since the year 2000.

Among refrigeration and air conditioning equipment, the amount of market refrigerant stock of room air conditioners is extremely high, and they have a significant impact as a source of leakage to the atmosphere. Therefore, this project performed research and development of component technologies which enable transfer to low-GWP refrigerants to medium and small air conditioning systems (mainly room air conditioners). It conducted research into high-efficiency and low-GWP refrigerants and research into primary equipment (compressors, heat exchangers, etc.) for achieving higher efficiency. Safety evaluations and performance evaluations of low-GWP refrigerants were also conducted.

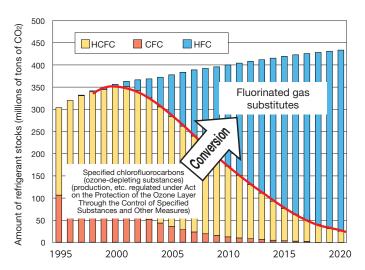


Figure 1 Market Stocks of Refrigerants in refrigeration and air conditioning equipment (BAU (Business As Usual) Estimate)

\*The results are values published by the government. The 2020 forecasts are estimates from the Ministry of Economy, Trade and Industry based on factors such as the number of refrigeration and air conditioning equipment shipped (The Japan Refrigeration and Air Conditioning Industry Association), leakage during use coefficient, disposal coefficient, collection results, etc. (2015)

#### Results

Through this project, core and component technologies necessary for practical application of medium and small air conditioning systems for achieving high efficiency (energy saving) equal to or greater than current products by using low-GWP refrigerants were established. Also, initiatives were conducted for evaluating the performance and safety of new low-GWP refrigerants.

In order to evaluate the performance of and to optimally design refrigeration and air conditioning equipment using new refrigerants, a mathematical model (equation of state) representing the thermodynamic properties of the refrigerant is essential, and a highly reliable equation of state was developed based on thermodynamic properties (critical constant, density, saturated steam pressure, specific heat capacity, acoustic velocity, etc.) measured with a high precision. Kyushu University conducted precise measurements of the thermodynamic properties necessary for performance evaluations and optimal design of refrigeration and air conditioning equipment for practical application of new low-GWP refrigerants (HFO-1123 and HCFO-1224yd (Z)), which are substitutes for HFCs, and developed a high-precision equation of state based on them. Furthermore, this new equation of state was registered in the REFPROP (Version 10) of the National Institute of Standards and Technology (NIST), a database of international standards for thermodynamic properties.

This has enabled simple calculation of the thermodynamic properties of HFO-1123 and HCFO-1224yd (Z) and of the thermodynamic properties of mixed refrigerants using existing refrigerants with these refrigerants. In the future, performance evaluations and optimal design of refrigeration and air conditioning equipment using new low-GWP refrigerants will be possible, and they are expected to have large contributions to the practical application of new refrigerants.



# Development of Technology and Assessment Techniques for Next-Generation Refrigerants with a Low GWP Value

#### Ongoing NEDO Projects

**Entrustment** 

Kyushu University, Waseda University, The University of Tokyo, Suwa University of Science, National Institute of Advanced Industrial Science and Technology (AIST)

**Grant Awards** 

Mitsubishi Electric Corporation, Toshiba Carrier Corporation, Panasonic Corporation, DAIKIN INDUSTRIES, LTD.

Joint Research / Re-entrustment

Toyama Prefectural University, Nihon University, National Institute of Advanced Industrial Science and Technology (AIST), Nagasaki University, Kyushu Sangyo University, Saga University, Tokyo University of Marine Science and Technology, The University of Electro-Communications, Shizuoka University, Hiroshima University, University of Fukui

R&D Period

FY2018-2022

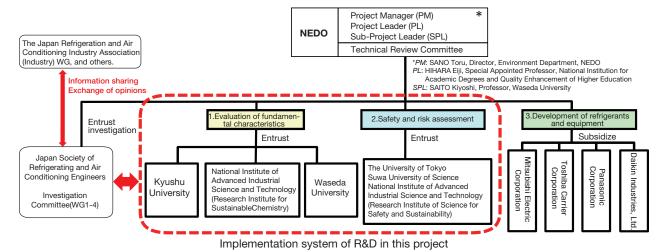
#### Summary

Meanwhile, many nextgeneration refrigerants with a low GWP value are said to have high technological hurdles in terms of maintaining or exceeding the equipment performance of conventional refrigeration and air conditioning equipment that use HFC refrigerants. They also present challenges in terms of safety, including flammability and chemical instability.

In response, NEDO has undertaken the development of assessment techniques for performance, safety, and risk of next-generation refrigerants used in refrigeration and air conditioning equipment since FY2018. Furthermore, the interim evaluation was conducted in an open, publicly-accessible format in October 2020. This project has been highly evaluated as all interim targets have been achieved and the final targets are also expected to be achieved.

Research and development item	Theme	Name of business operator	Objective
1 Evaluation of fundamental characteristics	Evaluation of fundamental heat transfer characteristics, and basic cycle performance of next-generation refrigerants Ky		Acquisition of property data of HFO- type refrigerant mixtures
	Safety evaluation of low GWP, low flammability refrigerant mixtures	National Institute of Advanced Industrial Science and Technology (Research Institute for Sustainable Chemistry)	Flammability evaluation of HFO-type refrigerant mixtures
	Research and development of the evaluation of the practical use of next-generation refrigeration and air-conditioning technologies using low GWP refrigerants	Waseda University	Development of performance evaluation simulation
2 Safety and risk assessment	Development of safety and risk assessment methods for next-generation refrigerants	The University of Tokyo Suwa University of Science National Institute of Advanced Industrial Science and Technology (Research Institute of Science for Safety and Sustainability)	Safety assessment of HC-type and HFO-type refrigerants
3 Development of refrigerants and	Research of large cooling units using natural refrigerants and ultra-low GWP refrigerants	Mitsubishi Electric Corporation	(Development of equipment) Cooling units for large refrigerated warehouses
equipment	Development of technologies for applying next- generation low GWP refrigerants to condensing units	Toshiba Carrier Corporation	(Development of equipment) Condensing units
	Development of energy-saving refrigerator systems using CO <sub>2</sub> refrigerants in low temperature equipment and their evaluation in commercial buildings	Panasonic Corporation	(Development of equipment) CO <sub>2</sub> refrigerator systems at convenience stores, supermarkets, distribution warehouses, and food processing plants
	Development of mildly flammable refrigerants (under GWP10) for direct expansion air- conditioning systems	Daikin Industries, Ltd.	(Development of refrigerants) Refrigerants, direct expansion air- conditioning systems (part of residential-use and commercial-use air conditioners)

List of project themes



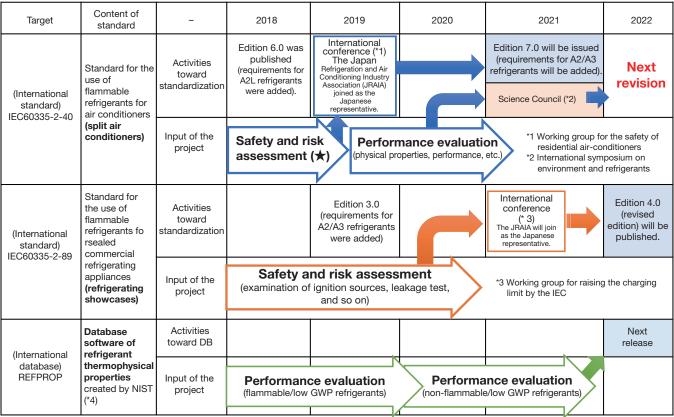


External view of refrigerant charge evaluation system

#### **Target Results**

NEDO is striving to contribute to the practical application and dissemination of next-generation refrigerants and equipment in a variety of ways. These include initiatives to link the results of this research and development activity to the registration of results in an international database, the creation of a practical safety standard for the industry, and the formulation of international regulations and standards. In addition, from FY2019, NEDO has been facilitating the dissemination of next-generation refrigerants by developing technologies to address technological roadblocks in areas where next-generation refrigerants have been adopted somewhat but have not seen widespread use, as well as improving their efficiency and expanding their scope of application.

Furthermore, through these initiatives, the aim is to achieve a reduction of approximately 10.2 million tons of CO<sub>2</sub>, a reduction of 70% compared to the reference value by 2029 and a reduction of 85% by 2036, which are the targets for reduction of the production and consumption of HFCs in Japan in the Kigali Amendment to the Montreal Protocol, and a reduction of 32% by 2013, the target for the reduction of HFC emissions in the Paris Agreement.



<sup>★</sup>Examination of ignition sources, countermeasures against chemical instability, and so on

Activities towardthe formulation of international regulations/standards and registration in international databases



<sup>\*4</sup> National Institute of Standards and Technology, which develops the international standard database for thermophysical properties.

### III. Project List

Starting Year	Year of completion	Executing agency				
R&D of energy use rationalization new refrigerant						
1994	2001	Research Institute of Innovative Technology for the Earth (RITE				
1996	1997	Japan Environmental Management Association for Industry (JEMAI)				
1996	1998	Kobe Steel, Ltd.				
1998	2002	Research Institute of Innovative Technology for the Earth (RITE) AGC Inc., Kanto Denka Kogyo Co., Ltd, Showa Denko K.K. Daikin Industries, Ltd., Canon Anelva Corporation, Hitach Kokusai Electric Inc., ULVAC, Inc., Tokyo Electron Limited Fujitsu Limited, and the others				
1998	2001	Japan Environmental Management Association for Industry (JEMAI)				
in the man	ufacturing c	of electronic devices				
1999	2003	Association of Super-Advanced Electronics Technologies (ASET)				
t of sustain	able society	y				
2000	2000	Mitsubishi Electric Corporation, AGC Inc.				
2000	2000	Achilles Corporation				
2000	2000	Mitsubishi Electric Corporation				
2000	2000	Mitsubishi Electric Corporation				
2002	2006	AGC Inc., Daikin Industries, Ltd., Central Glass Co., Ltd., Zeor Corporation, Tosoh Finechem Corporation, Chiba Institute of Technology, Japan Aluminium Association(JAA), Nagaoka University of Technology, Ahresty Corporation, STG Co., Ltd.				
2004	2006	Sankyo Tateyama, Inc., Sumitomo Electric Industries, Ltd. Daido Steel Co., Ltd., The Japan Steel Works, Ltd.				
Air-conditio	ning System	ns				
2005	2010	Chubu Electric Power Co., Inc., Mitsubishi Heavy Industries Thermal Systems, Ltd., and the others				
2005	2010	Earthship Co., Ltd, Honda R&D Co., Ltd				
2005	2010	The Japan Refrigeration and Air Conditioning Industry Associa- tion (JRAIA), National Institute of Advanced Industrial Science and Technology (AIST), and the others				
2005	2010	Shin Nippon Air Technologies Co., Ltd., Daikin Industries, Ltd. Panasonic Corporation, Mitsubishi Electric Corporation, and the others				
2006	2010	Asada Co. Ltd., Iceman Co. Ltd., Kohji Corporation, NKK Co Ltd., Kanto Denka Kogyo Co., Ltd., Consumers Cooperative Co-op Sapporo, Nikkin MgCast Co. Ltd., and the others(82 Companies in all)				
2007	2012	Kaneka Corporation, National Institute of Advanced Industria Science and Technology (AIST), Tokyo University of Science Nisshinbo Chemical Inc., Achilles Corporation, Kyoto University, Toray Industries, Inc., Asahi Fiber Glass Co., Ltd., Tokyo Institute of Technology, Japan Testing Center for Construction Materials(JTCCM), and the others				
2011	2015	AGC Inc., Daikin Industries, Ltd., Mitsubishi Heavy Industries Thermal Systems, Ltd, Panasonic Corporation, Mitsubish Electric Corporation, Sanden Holdings Corporation, The University of Tokyo, Kyushu University, Suwa University of Science, National Institute of Advanced Industrial Science and Technology (AIST)				
2016	2017	AGC Inc., Panasonic Corporation, Mitsubishi Electric Corporation, DENSO Corporation, Suwa University of Science, Kyushu University, The University of Tokyo, National Institute of Advanced Industrial Science and Technology (AIST)				
2018	2022	Kyushu University, Waseda University, The University of Tokyo Suwa University of Science, National Institute of Advanced Industrial Science and Technology (AIST), Mitsubishi Electric Corporation, Toshiba Carrier Corporation, Panasonic Corporation, DAIKIN INDUSTRIES, LTD.				
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#### Domestic Offices

#### Head Office

MUZA Kawasaki Central Tower, 16F-20F 1310 Omiya-cho, Saiwai-ku Kawasaki City, Kanagawa 212-8554 Japan

Tel: +81-44-520-5100 Fax: +81-44-520-5103

#### ●Kansai Branch Office

9th Floor, Knowledge Capital Tower C Grand Front Osaka 3-1 Ofuka-cho, Kita-ku, Osaka 530-0011 Japan Tel: +81-6-4965-2130

Fax: +81-6-4965-2131

#### Overseas Offices

#### •Washington

1717 H Street, NW, Suite 815 Washington, D.C. 20006, U.S.A. Tel: +1-202-822-9298

Fax: +1-202-733-3533

#### Silicon Valley

3945 Freedom Circle, Suite 790, Santa Clara, CA 95054 U.S.A. Tel: +1-408-567-8033 Fax: +1-408-567-9831

#### Europe

10, rue de la Paix 75002 Paris, France

Tel: +33-1-4450-1828 Fax: +33-1-4450-1829

#### New Delhi

15th Floor, Hindustan Times House, 18-20 Kasturba Gandhi Marg, Connaught Place, New Delhi 110 001, India Tel: +91-11-4351-0101

Tel: +91-11-4351-0101 Fax: +91-11-4351-0102

#### Beijing

2001 Chang Fu Gong Office Building Jia-26, Jian Guo Men Wai Street Beijing 100022, P.R.China Tel: +86-10-6526-3510

Tel: +86-10-6526-3510 Fax: +86-10-6526-3513

#### Bangkok

8th Floor, Sindhorn Building Tower 2 130-132 Wittayu Road, Lumphini Pathumwan Bangkok 10330, Thailand

Tel: +66-2-256-6725 Fax: +66-2-256-6727

MUZA Kawasaki Central Tower, 1310 Omiya-cho, Saiwai-ku Kawasaki City, Kanagawa 212-8554 Japan Tel: +81-44-520-5248 Fax: +81-44-520-5253 URL: https://www.nedo.go.jp/english/index.html