Development of Decommissioning Technology for Nuclear Power Plants in NUPEC

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1. Introduction

To reduce personnel and environmental burdens, NUPEC has been developing the technology ensuring the safe, reliable, and rational decommissioning of commercial nuclear power plants since 1982 (see Fig.1, 2). Developed technologies will be applied Tokai Power Station (GCR), and also will be applied to the light water reactors (BWR and PWR) in the next stage. To achieve these purposes, NUPEC has focused its development effort on techniques for decontamination, reactor dismantling, measurement of residual radioactivity in buildings and waste, waste recycling and decommissioning engineering.

To achieve a preliminary reduction in the work-atmosphere dose-equivalent rate during dismantling work, Techniques for Radiation Exposure Reduction before Dismantling has been developed and DF 100 or more has been proved possible, and waste decontamination liquid processing technology and decontamination effect measurement technology have been developed at the same time.

To ensure safety and minimizing dose rate of workers, mitigate impacts on the surrounding environment, the safety protection technology and remote dismantling technology have been developed.

It is necessary to verify that the concentration of radioactive substances remaining on the building's surface are below the limit in order to lift the radiation control area and dismantle the building. The wide-area contamination measuring technique, penetrated contamination measurement and final verification measurement technique have been developed. The proposed clearance level has been achieved.

The purpose of Decommissioning Waste Treatment Techniques is to reduce the amounts of radioactive waste and to reduce environmental burden. The physical and chemical decontamination techniques and the development of metal and concrete waste recycling techniques are under progress, and some techniques are quite promising for actual application, and clearance level measurement techniques have been developed, and proto-type actual plant apparatus is probed to have proper performance.

2. Techniques for radiation exposure reduction before dismantling

At nuclear power plants, activated iron rust and other substances are deposited on the inner surfaces of piping and equipment. To achieve a preliminary reduction in the work-atmosphere dose-equivalent rate during the dismantling work -- reducing the dose equivalent for workers and increasing efficiency -- technology is being developed on a "system decontamination" process for removing contaminants from piping systems, and an "equipment decontamination" process for removing contaminants from large machinery, tanks, and other equipment using a small amount of decontamination agent. A process for treating the waste liquids derived from decontamination and radiation measurement technology is developed, including rational measurement of decontamination effects. (see Fig.3)

(1) Decontamination technology

Decontamination for decommissioning purposes, unlike that during operation, is not restricted by considerations of damage to the base material of an item subject to decontamination. To respond to a variety of contamination situations, development efforts are aiming to achieve a decontamination agent with a decontamination factor of around 100 that generates less secondary waste and is easy to handle. Among the many decontamination agents available, it is difficult for dilute solution-based chelate, chelate organic acids (including CANDECON), organic acids (including CORD), and metallic ion reduction (LOMI) to achieve a level of DF 100, while concentrated solution-based organic acids (including oxalic acid) and inorganic acids (including chloric acid and nitric acid) can achieve a level of DF 100, but generate secondary wastes in amounts several times larger than the dilute solution-based agents. Efforts to improve decontamination agents are aiming to achieve better decontamination performance with dilute solutions and reduced secondary wastes with concentrated solution-based agents. These developments involve tests using decontamination skids for oxide films prepared on the surfaces of piping, valves, pumps, heat exchangers and other test subjects simulating field equipment and under field conditions, and/or with hot sample-based verification of the decontamination performance of decontamination agents that are under development.

a. System decontamination

The BWR systems, consisting of stainless steel and carbon steel, have proved capable of achieving DF 100 or more with a circulation process (95 $^{\circ}$ C, NP-treated) using a dilute chloric acid reduction agent of inhibitor-laced chloric acid and vanadium chloride mixtures. Systems in which the circulation method cannot be used have a fill & drain process, using a concentrated chloric acid reduction agent (60 $^{\circ}$ C) of chloric acid with vanadium chloride and L-ascorbic acid are added. DF 100 or more has proved possible with the PWR's primary-system stainless steel using a circulation process (95 $^{\circ}$ C, NP-treated) with oxalic acid and vanadium oxalate.

b. Large-scale equipment decontamination

Large-scale equipment has a larger capacity for its decontamination area and, if the equipment is filled with a decontamination solution, waste decontamination liquids are generated in large quantities. To avoid this, a decontamination performance (DF 45) was verified with a gel process, applying a concentrated chloric acid reduction based decontamination agent or a concentrated organic acid-based decontamination agent to the inner surfaces of tanks and other large containers.

(2) Waste decontamination liquid processing technology

For the smaller amounts of secondary waste generated due to decontamination, technology is being developed to process waste decontamination liquids so that their renewal/reuse rate can be increased to 70 % or more, and to dispose of chelate and other organic substance.

It has been shown that chelate decomposition in the waste decontamination liquid with peroxide hydrogen is 90 % or more, while the organic decomposition in waste liquids containing organic substance (inhibitors) is 50 % or more.

Chloric acid-based decontamination agents have proved to achieve a chloric acid recovery rate of 70 % or more using an iron exchange-electrolytic renewal method, and a recovery rate of 90 % or more for metal source ingredients in dilute chloric acid.

(3) Decontamination effect measurement technology

In planning overall decommissioning measures and evaluating work during dismantling operations, it is indispensable to know radioactivity inventories and surface dose-equivalent rates for equipment, etc. after shutdown. On the other hand, nuclear reactors and primary-system equipment, to which access is difficult due to high dose-equivalent rates, require remote measurements or fewer access for measurement.

In order to carry out fieldwork in as short a time as possible for the measurement of dose-equivalent rates on equipment surfaces, a measurement system capable of evaluation with fewer measurement points by means of an EM (expectation maximization) process is now in use, and data compensation in the CT field is under development, using small semiconductor detectors that have a light transmission function and wireless infrared transmission of measurement data between detectors.

Efforts are being directed at developing a technology for measuring radioactivity on the inner surfaces of pipes using remote detector quantification of radioactivity and identifying waste decontamination liquid radioactivity using the ratio between a γ -ray spectral scattering beam and non-scattering beam counting rates.

3. Dismantling technology

During dismantling the nuclear power plant, it is important to ensure safety and keep to minimum dose rate of workers, mitigate impacts on the surrounding environment, reduce the volume of wastes, and improve the working efficiency.

(1) Remote operating/automatic control technology

For keeping safe operation under high-radioactive environments, remote operation technique, monitoring system to keep the accuracy and efficiency on the works, and a technique to organically control element technologies and ensure reliable automatic control technique for improving the efficiency are under investigating by using full scale replica of reactor pressure vessel. (see Fig.4)

(2) Reactor core internal cutting technology

Core internal are made of stainless steel and activated heavy structural member materials. The plate thickness is mostly 50-150 mm but some parts in PWR have the thickness of 500 mm. Some tools for cutting

location and angle, etc. allow substantial plate of up to 200 mm to be cut, however our target of cutting thickness was established 300 mm.

Methods of cutting stainless steel include mechanical cutting and thermal cutting, though laser cutting method, which is surpass in ability of cutting and remote control, were implemented using 30 kW class CO laser.

To reduce the volume of secondary products, laser beam and assist gas conditions to deliver the required cutting quality with narrower cutting kerf were identified. The reliability and applicability of there conditions for field equipment of a high-quality cutting process with a special laser cutting nozzle and another cutting process where a simulated model of core internals is cut, were verified and found to generate fewer secondary products up to 300 mm thickness in air and 150mm under water.

(3) Reactor pressure vessel cutting technique

A reactor pressure vessel is a large component made of 170-420 mm-thick low-carbon alloy plate steel with stainless steel cladding up to 10 mm thickness. It is activated due to long term operation. During dismantling for decommissioning, therefore, due to the necessity of remote operating for cutting under water to reduce the dose rate during works, a combined arc gauzing and gas cutting method was verified. This process first fuses off by arc gauzing the cladding of stainless steel having high fusion temperature, allowing the low-alloy steel to be exposed, and then cuts the low-alloy steel by fusion with a propane-oxygen mixture gas using a gas cutting torch.

It was verified that a reactor pressure vessel-simulated model (maximum plate thickness of 420 mm) could be cut under water by remote operating and is thus applicable to field equipment.

(4) Biological shield wall surface layer dismantling technique

The biological shield wall is a concrete structure of up to 3 m thickness lined with 10 cm steel plate and densely packed inside with reinforced bar of 51 mm in maximum diameter and activated to its depth of about 1 m from the inner surface. To separate the surface layer, a process combining cutting with a disk cutter and separation by a wedge process was verified.

Using the 110 MWe-class reactor biological shield wall-simulated model, the inner wall's steel liner and reinforced concrete were cut horizontally using a disk cutter and then vertically but slightly diagonally in two directions to verify the separation of a prism-like block. The second layer was separated into the thickdiameter reinforcement part in the vertical direction by the same process. The third layer concrete block was cut vertically and horizontally with a disk cutter and then, using a mechanical wedge, a cubic block was separated, confirming the applicability of the technique to field equipment.

4. Residual Radioactivity Measurement and Assessment Techniques for Building and Soil

After equipment has been removed, it is planned to release the radiation control area and dismantle the building. To release the control area, it is necessary to verify that the concentration of radioactive substances remaining on the building's surfaces is less than the value specified. When the clearance level now under review is fixed, it will be necessary to confirm precisely the clearance level. The Co-60 level (0.4 Bq/g) currently being proposed by the Nuclear Safety Committee is similar to the level for K-40, a typical natural radioactive nuclide contained in the building's concrete material. For this reason, the measurement technique for radioactive materials remaining in the building must be able to identify extremely low levels of contaminants contained in the concrete material precisely, at levels approaching those of K-40.

With this point in mind, technology is being developed for commercial nuclear power plants to evaluate accurately and quickly the positional distribution of residual radioactive contamination on extensive building surfaces of about 100,000 m^2 , and to assess the extent to which the contamination has penetrated into the concrete of the building. Detectors and measurement methods are also being developed to verify that no contamination exists in the building or the soil after the building has been dismantled. (see Fig.5) These tests are being carried out in Tokai Power Station.

(1) Wide-range contamination measuring technique

A collective distribution measurement system is being developed as a technology for accurately and efficiently assessing the distributed state of contamination remaining in building concrete (lower limits for measurement of the assumed clearance level: 1 kBq/m^2 , equivalent to 0.1 Bq/g; processing capacity: $100 \text{ m}^2/\text{day}$). This is based on a γ -ray camera-type 2D distribution detector of NaI, capable of one-time remote measurement at high-elevation locations. Development of a close contact-type measurement system is also underway for measuring floor surfaces and lower wall, using a plastic scintillation detector that has excellent performance in

terms of processing amounts suitable for automatic measurement.

(2) Penetrated contamination measurement

To measure the depth of penetrated contamination to effectively remove contaminated concrete, nondestructive measurement was conducted for subsurface contamination, the targets being a lower measurement limit of 0.1 Bq/g and a processing capacity of 20 m^2 /day, using a NaI and plastic scintillation detector. It was found that evaluation of surface and penetrated inner contamination is possible on the basis of scattering/linear ingredient ratios. For the penetrated type of exponentially attenuating contamination within concrete, it is possible to evaluate down to 2 cm in relation to contamination severity and distribution depth. For concrete contamination in depth, a measurement process involving boring a small hole and inserting a CsI and plastic scintillation detector is being developed, achieving a radiation source positional discrimination performance of about 1.5 cm in relation to the distance from radiation source.

(3) Final measurement for verification

Development is underway to verify that there is no contamination in the concrete and soil as a requirement for releasing the building's radiation control area, with a lower measurement limit of 1 kBq/m^2 (equivalent to 0.1 Bq/g) and a processing capacity of 100 m²/day and 200 m²/day each other as a goal.

To cover the area in which scanning measurement is difficult, a simple collective measurement system detector capable of nuclide radioactivity evaluation within the building has been developed using a Ge, confirming that the detector response can evaluate in simulation to an accuracy of about 20 %. A temperature-dependent plastic scintillation fiber detector has also been developed.

As a measurement method for verifying the absence of contamination in soil (including discrimination from fallout and natural nuclides), a scanning measurement system that combines a Ge detector and a large plastic scintillation detector is being developed, so that large area of soil can be efficiently measured. The plastic scintillation detector is used to assess contamination, and the background fluctuation due to natural radioactivity is corrected with measurements made by the Ge detector. The region likely to be contaminated is measured in detail by the Ge detector. There is also a method using the GPS (Global Positioning System) for positioning detection, and this may be able to provide accuracy levels down to a few centimeter span.

5. Techniques of Decommissioning Waste Processing

The wastes arising during the decommissioning of nuclear power plant not only involve a wide variety of different types, metals and concrete, radioactive and nonradioactive materials, and irradiated or radioactive contaminated materials; but also involve large quantities. The wastes produced during dismantling amount to as much as some 500,000 ton in the case of a 1,100 MW-class light-water reactor. It is important to treat these wastes properly in order to reduce the amounts of radioactive waste produced, and also to reuse and recycle resources effectively and appropriately. In particular, radioactive waste has to be distinguished from other types by properly evaluating measurements prior to disposal for different types of treatment. It is also appropriate to separate radioactivity from decommissioning waste by decontamination.

5.1 Decontamination and radiation measurement technology

(1) Radioactivity measurement technology

The clearance level measurement process has proved effective to up to 10 t/h of processing capacity and 40 Bq/t the clearance-equivalent level, by combining Ge detectors, plastic scintillation detectors, and NaI scintillation detectors. The plastic scintillation detector has achieved a favorable measurement accuracy of \pm 50 % or less at 10 t/h, even near the lower target limit, while Ge detector has also proved fully capable of measurement over a range of up to 4 kBq/t. Evaluation of many influencing factors, including the packing density, has found such factors to be safe when applied to field equipment.

In the meantime, the contamination distribution measurement process has proved to be effective up to 2 t/h of processing capacity, 400 kBq/t of accuracy, and 10 cm of positional resolution, with complex structures (including valves) even found to be fully capable of being identified near the lower limit of the detector.

(2) Decommissioning waste decontamination technology

a. Blast decontamination technology (pre-decontamination processing)

Blast decontamination processes include a wet process, using water plus zirconia and other shots, and

a dry process, using air and dry ice, have been developed. For this development, both processes were used with aluminum, zirconia beads, dry ice, etc. as blast materials to perform tests for the removal of surface oxide films and paints off hot samples from field equipment, etc., achieving a decontamination factor (DF) of 10^2 or more. Among the blast materials, zirconium beads have proved to be effective for removing persistent surface films and paints, and for planning the surface of metal substances. Dry ice, which generates fewer secondary wastes, was proved to be effective in peeling off loose paint and other attachments, but unsuitable for shaving off hard oxide films.

b. Electrolytic decontamination technology (thorough decontamination)

The electrolytic decontamination process is used for metal surface processing in industry in general, and is very good at removing contaminants from simple-shaped objects such as pipes and plates. In this development, 5 wt% of sulfuric acid, which generates fewer secondary products, was used as the electrolyte for a direct method in which waste materials are directly connected with to decontaminate simple-shaped metals under conditions of 0.3 A/cm² in current density and 60 °C, achieving a decontamination factor (DF) of 10⁴ or more. Despite a loss of 20-50 % in electrical efficiency, an indirect method of connecting a cage in which waste materials are contained with the electrode was also found to be possible for decontamination, easily remote-controlled, and applicable to large-quantity decontamination materials on a field scale.

c. REDOX decontamination technology (thorough decontamination)

The REDOX decontamination process is a dipping method for removing contaminants from relatively complex-shaped equipment such as valves, pumps, and small-diameter pipes for which the electrolytic decontamination process is unsuitable. It was verified on a field scale that cerium nitrate (Ce⁴⁺ 0.4 mol/l, HNO₃ 2 mol/l, 50-80 °C) dissolves the surface substrate layer off complex-shaped metals evenly by some 10-1000 μ m, achieving a decontamination factor (DF) of 10⁴ or more. The metals being subjected to decontamination can be handled as in a cage; the piping length, charging rate, and other factors cause no problems for even dissolution.

d. Post-decontamination cleaning technology

The post-decontamination cleaning process based on ultrasonic cleaning technology has proved to remove contaminants attached to waste materials almost completely in a tank of about 1 m^3 using an ultrasonic wave transmitter of 1 W/cm² in output density.

e. Combined radioactivity measurement/decontamination technology

Technologies for radiation measurement and post-dismantling decontamination are assembled into a waste processing system, and this system performs successfully with a variety of wastes derived from dismantling as originally intended, on an actual scale. (see Fig.6)

f. Laser decontamination technology

Laser decontamination technology for the paint and organic material on the surface of floor, wall, tank and equipment in nuclear facility has been developed to reduce the second waste generation and to prevent the basic material from making radioactive waste. The decontamination test is been carried out by using pulse YAG laser.

5.2 Decommissioning waste recycling technology

(1) Metal recycling technology

a. Pyro-metallurgical separation technology

To expand the recycling range of metal decommissioning wastes, a process of separating Ni and Co from metals, capable of achieving a decontamination factor (DF) of 10 or more and a recyclable metal recovery rate of 60 % or more, is being developed for a target reduction of processing loads by some 3,000 ton.

An oxygen sparging method (oxygen gas oxidation method), a method of separation using oxidation energy (oxidation speed) difference-based selective oxidation, has proved to achieve DF 100 or more for Ni in relation to stainless steel, 10 or more for Co in relation to both carbon steel and stainless steel, and a recyclable metal recovery rate of 60 % or more. (see Fig.7)

b. Molten Metal casting technology

Low-level metal wastes can be used by melting and filling in place of mortar, etc., so that the amounts disposed of in waste form can be reduced. The metal filling rate has reached 95% or more in a simulated waste

form of 1/2 scale, and the thickness of the container can be reduced to only 6 min.

(2) Concrete recycling technology

a. High-quality aggregate recovery technology

The objective of this development is to recover coarse and fine aggregates from decommissioning concrete waste, and to meet the Japan Architecture Society standards JASS5N (standards for nuclear facility). It has been confirmed that the aggregate recovery rate can be raised to 70 % or more to provide a method of recycling, making it possible to recycle concrete wastes by some 500,000 ton.

The mechanical grinding method for the recycling of aggregate (a process of removing hydrated concrete adhering to raw aggregate by grinding it with a crusher) allows recycled coarse aggregate to meet the JASS5N standard. In the case of the selective heating method (a process for renewing aggregate by selectively heating cement paste with microwaves for crusher-based separation and removal), both coarse and fine aggregates meet JASS5N. The whole-heating method (a process of grinding with a crusher after whole-heating treatment to recover the aggregate) has also enabled both coarse and fine aggregates to satisfy JASS5N. (see Fig.8)

b. By-product powder recycling technology

A process for recovering the powder derived from the high-quality aggregate being recovered into aggregate, which meets JIS cement standards and JASS5N standards, is being developed in order to recycle concrete wastes by some 500,000 ton.

It was possible to manufacture Portland cement to JIS standards using a burnt cement manufacturing process.

Recycling Technique for Radioactive Concrete

Recycling technology of radioactive concrete into solidification material mortar of waste form is carried out to establish the production technique for solidifying materials and for sludge filler waste form.

Graphite waste treatment technology

The underground geological disposal of graphite removed from the reactor core was investigated from the viewpoint of reduction of underground disposal cost. By cutting a portion of the graphite blocks using a mechanical cutting machine such as a band saw and packaging them in waste disposal containers, fundamental data was accumulated for the high density storage concept and it was certified that this concept could exist rationally.

The incineration disposal of graphite removed from the reactor core was investigated from the viewpoint of reduction of the radioactive material inventory emitted into the atmosphere. A system concept was established in which C-14 in the off gas is first separated with isotope in the form of either CO_2 or CO, and then recovered as carbon by deoxidization.

Technical problems that should be cleared in the future for both of these disposal options were identified and verification test plans were established. Through execution of various tests based upon established verification test plans, we hope to realize safe and rational treatment and disposal of the graphite waste.

6. Conclusion

In NUPEC the decommissioning technology started in the beginning of 1980's and the technical development on decommissioning waste processing, radiation exposure reduction before dismantling, residual radioactive measurement and assessment for building and soil and dismantling has been carried out. We consider that the technical development according to the decommissioning state of the research nuclear facilities, nuclear power plants and so on is necessary for further rational decommissioning.

These technical developments are carried out as MITI's trust.

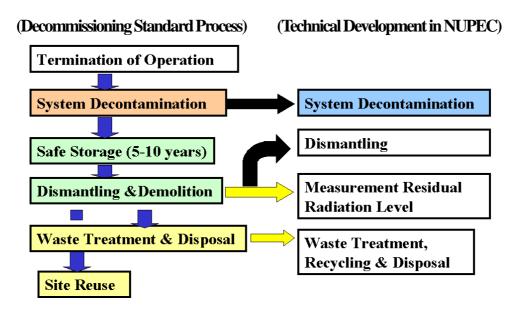


Fig.1 Decommissioning Technology Development in NUPEC

FY	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	0	1	2
Cutting Technique for RPV																					
Taking off Technique for BSW																					
Cutting Technique for RCI																					
Waste Treatment Technique		Decontamination & Clearance Lebel Measurement														Recy Tech					
Decontamination Technique before Dismantling																					
Residual Radiation Measurement Technique																					
Dismantling Technique																					

Fig.2 Decommissioning Technology Development Schedule in NUPEC

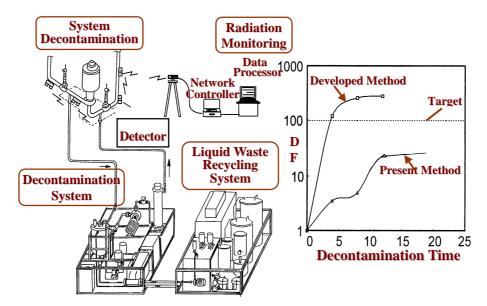


Fig.3 Decontamination Technology before Dismantling

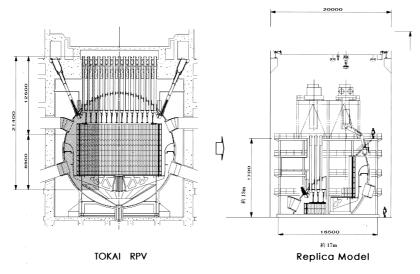


Fig.4 Reactor full Scale Replica in Katsuta Engineering Laboratory of NUPEC

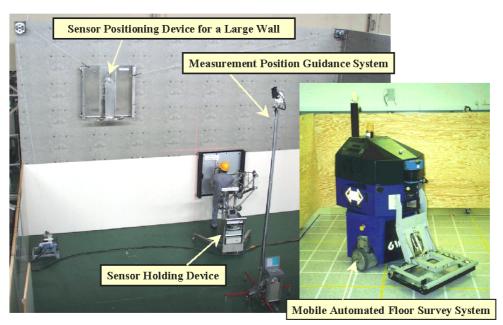


Fig.5 Residual Radiation Measurement

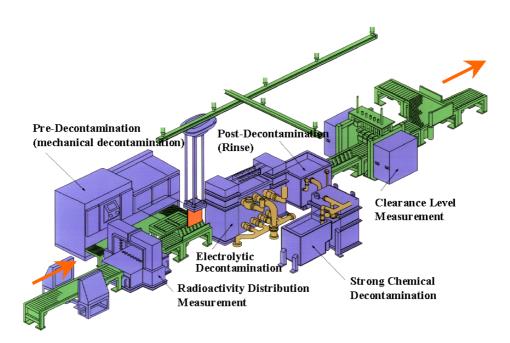
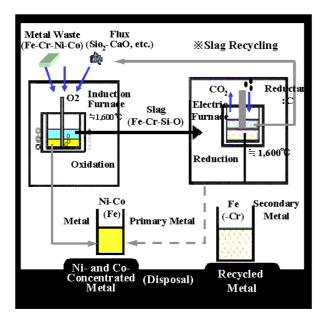


Fig.6 Decontamination and Measurement System for Decommissioning Waste





Removal of Recycled Metal (after secondary reduction)

Fig.7 Dismantled Metal Recycling Technology

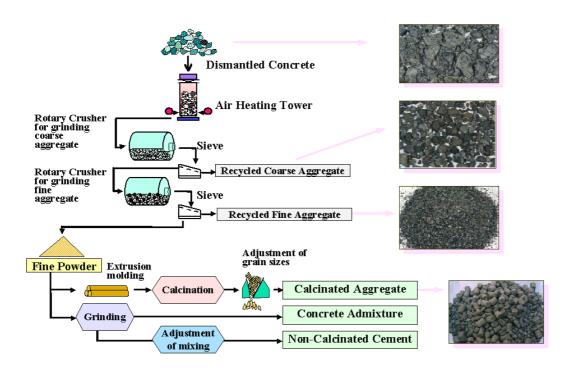


Fig.8 Dismantled Concrete Recycling Technology