Research Progress of Radioactive Decontamination Methods

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Abstract

This article describes the development of the nuclear industry at home and abroad, and introduces the sources and types of radioactive pollutants from nuclear facilities decommissioned. The focus is on the decontamination methods in nuclear decommissioning technology, mainly mechanical and physical decontamination methods, chemical decontamination methods, electrochemical decontamination methods and biological decontamination methods. The mechanical physical decontamination method mainly includes smelting decontamination, laser decontamination, mechanical decontamination, ultrasonic decontamination and high-pressure water cleaning decontamination technology. Chemical decontamination methods mainly include foam method, gel method, oxidation-reduction method, and complexation method. The current research status of these methods at home and abroad is also explained. A reasonable and effective combination of these decontamination methods has become the main direction for the development and application of decontamination technologies for decommissioning nuclear facilities in the future.

Keywords

Radioactive Contamination; Decommissioning Technology; Decontamination Method.

1. Introduction

Compared with traditional energy sources such as coal and petroleum, nuclear energy has a very high energy density. Nuclear fuel stores huge energy in a very small volume and can be released in a controlled manner. Therefore, it has been quickly applied since its discovery. The energy of nuclear radiation is huge, and direct contact at close range can cause great harm to the human body. Therefore, in the process of human development and utilization of nuclear energy, reducing or even avoiding the harm of nuclear radiation to humans and the pollution of the environment has become a safety that must be solved. The removal of radioactive pollution has become a new and difficult task accompanying the use of nuclear energy, especially in the process of decommissioning nuclear facilities. The removal of radioactive contamination usually uses various physical and chemical methods to remove radioactive contaminants on the surfaces of facilities and equipment contaminated by radionuclides. Based on the particularity of radioactive contamination, the development of radioactive decontamination methods is not only oriented towards decontamination, it also requires these decontamination methods to maximize purification efficiency, and minimize the amount of waste and decontamination costs (including Disposal and disposal costs of secondary waste), and will not cause damage to equipment and facilities [1]. Among them, reducing the amount of waste is one of the most important principles of radioactive decontamination and post-processing, which has played an important guiding role in the research and development of radioactive decontamination technology. The use of nuclear energy began in the 1940s and 1950s, so there are currently a large number of nuclear facilities around the world that are being decommissioned or preparing to be decommissioned due to their useful life. The decommissioning of a nuclear facility is an action taken

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after the expiration of the service period of the nuclear facility or the suspension of service for other reasons, in order to fully consider the health and safety of the staff and the public, and environmental protection, the selection and application of appropriate dismantling and purification technologies becomes The key elements of the successful decommissioning project [2]. According to the general technical system for decommissioning nuclear facilities, decommissioning technologies can be summarized into 7 types of technologies including source item investigation, decontamination, dismantling and dismantling, waste treatment, waste classification and testing, radiation protection, and site restoration and treatment. This article mainly studies decontamination technology.

Decontamination is an operation that uses different methods to completely or partially remove radioactive contaminants from the surface or interior of a nuclear facility. The main purpose is to reduce the radioactive level of the workplace and the target, and the waste level of contaminated equipment and materials. Minimize the total amount of materials to be sorted or processed as solid radioactive waste, and increase the possibility of recycling and reuse of equipment, materials, or houses. Decontamination cannot completely eliminate radioactive elements, but can only change the form or location of nuclide to facilitate radiation safety management [3].

2. Purpose of decontamination

Decontamination is divided into two categories: one type is decontamination performed during nuclear facilities in service; the other type is decontamination performed during decommissioning of nuclear facilities. Decontamination mainly has the following purposes [4]:

1) Reduce the radioactivity level of the work site and the work object, reduce the difficulty and risk in the decommissioning process, simplify the operation procedures, and reduce the radiation dose of the workers.

2) Some materials, equipment and facilities that have been contaminated by radionuclides can meet certain standards, so that the facilities can continue to be used safely or the site can be opened.

3) The materials, equipment and facilities that have been contaminated by radionuclides can be reduced in waste levels or converted into non-radioactive waste, so as to reduce the quality and volume of waste, and reduce the difficulty and cost of storage, transportation and disposal.

4) Reduce the requirements for shielding and remote operation, facilitate the maintenance and disassembly of equipment and facilities by the operators, and facilitate the handling of accidents.

5) Remove loose radioactive contaminants in order to reduce waste preparation and prevent the expansion of pollution.

3. Radionuclide

3.1 Sources of radionuclide contaminants

When decontamination of nuclear facility equipment and components, you need to understand two points. The first is to clarify the parts that need to be decontaminated, and the second is to know where the radioactive contaminants come from [5]. In view of these two points, appropriate decontamination methods should be adopted. There are many sources of radioactive contaminants in nuclear facilities, which can be summarized as the following: (1) After the core of the nuclear reactor is activated, it will easily corrode, which will lead to falling off and falling into the coolant. Contact parts; (2) The coolant is easy to have impurities and these impurities will be activated by neutrons, and then deposited on the surface of the pipeline (3) Corrosion products in the loop will also be activated and deposited on the surface of the pipeline by neutrons; (4) There are elements on the surface of nuclear fuel components that are easily activated by neutrons, and then radionuclides are produced, which are then deposited on the surface of the pipeline; (5) The nuclear fuel components may also be damaged, causing the fission of transuranic elements and radioactive materials inside. If the product leaks, these substances will enter the coolant and stay in contact with the coolant; (6) During the management, storage and handling of radioactive materials, there is a possibility of leakage accidents, which will

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cause radioactive pollution; (7) When the nuclear reactor is in normal operation, it will cause radioactive pollution when the processed radioactive material is discharged.

3.2 Radioactive surface contamination layer characteristics

According to the longitudinal depth of surface contamination, the radionuclide contamination layer can be divided into loose surface contamination, corrosion product film, oxide film and contaminated base material infiltration layer [6]. There are many factors that affect the formation of surface contaminants, mainly including: surface material properties, water chemical properties of the coolant, operating pressure and temperature of the coolant, outage events, and the degree and frequency of accidents.

The main forms of binding of radioactive nuclides to the surface include: 1 non-fixed pollutants attached to the surface by intermolecular forces; 2 weakly fixed pollutants formed by chemical adsorption or ion exchange; 3 radionuclide diffusion to the substrate Deep surface contamination formed by neutron irradiation activation of trace elements inside or inside the substrate [7]. The pollution layer attached to the surface of the object can be removed by a simple physical method. However, if the pollutant interacts with the surface chemically or ion exchange, it may produce corrosion product film and oxide film, deep diffusion of pollutant nuclides or trace elements in the substrate Surface contamination caused by activation is difficult to remove. Therefore, in response to different levels of surface pollution and the requirements for decontamination effects, it is necessary to choose a reasonable decontamination technology.

3.3 The main radionuclide in the surface contamination layer

Various types of facilities, especially reactors, produce different radioactive contaminated nuclides due to the different moderators, coolants, structural materials, nuclear fuels, and auxiliary processes used. However, when considering various types of radioactive contaminated nuclides, consider various radioactive nuclides. The amount of contaminated elements should include ^{94}Nb, ^{93}Mo, ^{108}Ag, ^{131}Sm, ^{152}Eu, ^{154}En, ^{166}Ho, etc., which are generated by neutron radiation in stainless steel, and ^{14}C, ^{36}Cl, ^{151}Sm, ^{152}Eu, ^{154}En, ^{166}Ho, ^{94}Nb, etc

4. Decontamination method.

In the 1960s, a small number of countries represented by the United States took the lead in the systematic data compilation and research and development of radioactive decontamination technology. With the promotion and application of nuclear energy utilization technology on a global scale, many countries researching and using nuclear energy Also joined the research team on radioactive decontamination technology [8]. Due to the variety of objects to be decontaminated and the requirements for the degree of decontamination are not the same, so far there is no universal decontamination method suitable for all situations. Therefore, in order to cope with complex pollution conditions, the development of decontamination technologies tends to diversify, and more new decontamination technologies are gradually being developed. At the same time, traditional decontamination technologies such as mechanical physics, chemistry, electrochemistry, biology and smelting are widely used and are more mature than the new decontamination technologies that have emerged in recent years.

4.1 Mechanical physical decontamination method

The mechanical physical decontamination method mainly refers to the method of removing radioactive contamination on the metal surface through physical means such as mechanical grinding, ultrasound, laser, smelting, etc. The commonly used physical methods for the radioactive purification of metal surfaces mainly include smelting decontamination, laser decontamination [9], mechanical decontamination [10] ultrasonic decontamination [11] and high-pressure water cleaning decontamination technology [12], etc.

The smelting decontamination method is to select a suitable furnace type and smelt the contaminated metal with a specific component of the co-solvent, so that most of the radionuclides in the waste are

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enriched in the slag and filter dust. Or by adopting a reasonable furnace lining formula to adsorb radionuclides. It can effectively remove radionuclides in metal wastes, achieve the purpose of purifying metal wastes, realize resource recycling and reuse, and facilitate the treatment of radioactive wastes, and realize the containment of radionuclides. Advantages: It can effectively remove radionuclides in metal wastes, and it is convenient to dispose of solid wastes enriched with radionuclides after smelting. Disadvantages: complex pre-treatment procedures, high decontamination costs, and large flue gas production.

There are many applications for the treatment of radioactively contaminated metals by smelting and decontamination methods at home and abroad. For example, foreign SiempeIkamp Foundry smelters and Saclay companies use induction furnaces to smelt and process radioactively contaminated metals, thereby purifying them [13, 14] and domestic smelting The radioactive contaminated metal produced in the uranium mining and metallurgy process is processed in a way that the pollution level of carbon steel and stainless steel with a pollution level of 4~48 Bq/cm^2 is reduced to 0.004~0.016 Bq/cm^2 after treatment, and the recovered metal is re-applied to the interior In the production system [15].

The laser decontamination method uses a high-energy laser beam to act on the metal surface, and the high temperature generated by the laser vaporizes the metal surface layer, thereby achieving the separation of pollutants from the metal matrix [16]. The research of laser decontamination technology started in the 1980s. The comprehensive use of laser radiation, heat, vibration and photochemical effects on the surface of the material to achieve effective stripping of the deposited erosion layer attached to the surface of the material is to minimize radioactive waste during the decommissioning of nuclear facilities Effective technical means. In particular, the laser decontamination device combined with laser long-distance transmission and control technology has the advantages of safety and efficiency and minimization of secondary pollutants, and has great application value in the field of radioactive contamination cleaning. There are many researches and applications of using lasers to remove radioactive contamination on metal surfaces at home and abroad. At present, the lasers studied and applied in practice mainly include Nd:YAG lasers, CO_2 lasers, etc. [17, 18] Researchers such as Aniruddha Kumar have studied the decontamination performance of CO_2 lasers to remove ^{137}Cs contamination on the surface of radioactively contaminated metals (stainless steel, copper and aluminum). Researchers in South Korea used Nd:YAG lasers to decontaminate ^{137}Cs on the surface of 304 stainless steel. After 100 times of laser irradiation with a wavelength of 1064nm and a working power of 450mJ/pulse, the decontamination rate of ^{137}Cs on the metal surface can reach more than 98%. Domestic Gao Zhixing et al [9] confirmed that the laser decontamination coefficient for metal intermediate level waste can reach more than 100 times, and the residual radioactivity of stainless steel samples contaminated with low and medium level radioactive waste after laser decontamination is close to 0.1 Bq/cm^2, reaching radioactivity. National standards for waste cleaning and control. The results showed that after 100 times of 1J/cm^2 UV laser irradiation, the decontamination rate of radioactive contamination on the surface of the contaminated object exceeded 80%.

The mechanical decontamination method uses physical principles to grind facilities or surfaces. This method is generally suitable for concrete structures, but the disadvantage of this method is that it is easy to generate a large amount of dust during use, which causes secondary pollution, so it is necessary to find a way to solve the problem of dust collection and disposal.

The principle of ultrasonic decontamination is that ultrasonic waves can produce a kind of cavitation in the solution, which can then be used for scrubbing and decontamination [11,19]. Based on the discovery of this method, some researchers have discovered a way that can use ultrasonic decontamination to remove the radioactive particles on the surface of nuclear fuel rods [20]. It takes about half an hour to remove contaminants on the surface of nuclear fuel rods, and when the temperature of the decontamination liquid is 45 ℃, the decontamination effect is the best. Using this method to decontaminate, basically no secondary pollutants will be generated, and very few pollutants can be processed by filtration. In the process of radioactive decontamination of metal surfaces, ultrasonic decontamination technology alone cannot achieve the removal of fixed pollution on the

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metal surface, and is often used in combination with chemical decontamination methods. Wu Qiang et al [21] proposed a new decontamination method that combines ultrasonic and chemical methods. The verification test results show that this method has a better decontamination effect than chemical methods, and the purification factor for radioactive contamination of stainless steel and other metal surfaces can be achieved.

High-pressure water cleaning and decontamination technology [22] has been widely promoted and has been applied in practice. This technology can not only be used in the various stages of reactor operation and maintenance that have been decommissioned, but also can be used for general industrial equipment decontamination. The main purpose of this technology is to remove the oxide attached to the metal surface. This is because the oxide generally has a large number of cracks and is loosely attached to the surface of the device. Using this technology to decontaminate surface contaminants has certain advantages [23]. Because the water itself is clean and environmentally friendly, and the cost is low, and there will be no secondary pollution during the cleaning process. In the process of decontamination of radioactively contaminated components, special care should be taken to collect the waste liquid and waste generated to avoid secondary pollution. my country has successfully used high-pressure water jet cleaning technology to decontaminate my country's 801 pairs, and established a "three tanks, three tanks and three pumps" chemical decontamination process, which has realized the engineering decontamination of stainless steel pipe fittings [24].

4.2 Chemical decontamination

Chemical decontamination mainly uses chemical decontaminants to dissolve the greasy substances, paint coatings or peeling oxide film layers on the surface of equipment, parts and materials contaminated by radionuclides, so as to achieve the removal of radionuclides adhering to the oil and oxide films. purpose. Chemical decontamination of fixed pollutants on the surfaces of radioactively contaminated pipelines, components, facilities and equipment is often used. Generally speaking, the most effective and ideal substrate for chemical decontamination is a non-porous surface [25]. The use of chemical decontamination technology generally requires consideration of the matrix condition, the characteristics of the contaminants, the financial resources, the allowable corrosion depth of the material, the safety plan and the subsequent waste treatment and disposal technology [26], and the decontamination method or multiple types can be reasonably selected mix.

Chemical decontamination is currently highly versatile and has many researches. Its advantages are mainly that there are many industrial application examples of detergents, high decontamination efficiency, and little or no airborne pollutants are generated. Chemical detergents are easy to prepare and decontaminate. The agent is mainly composed of inorganic acids, organic acids, redox, chelating agents, alkalis, surfactants, solvents, corrosion inhibitors, accelerators, etc. The main disadvantage is that it is difficult to deal with porous and corrosive surfaces. Generally speaking, there are many kinds of chemical detergents, and the cost of decontamination is generally not low.

Commonly used chemical decontamination methods include foam method, gel method, redox method, complex method, organic acid method, inorganic acid method, gas phase method, etc. Usually the decontamination effect is related to many factors such as the type, concentration, action time, temperature, stirring condition of the detergent, and should also be based on the nature of the pollutant, the matrix condition, the budget, the allowable degree of corrosion of the material, the safety design, the cost and the subsequent waste For specific conditions such as disposal methods, chemical decontamination methods should be reasonably selected or used in combination with several other methods to improve the decontamination effect.

Acid treatment is divided into organic acid treatment and inorganic acid treatment [27]. For example: when decontamination in an aqueous phosphoric acid solution, the decontamination effect is related to the corrosion time of the phosphoric acid solution [28]. HP/CORD-UA: In the acid treatment of mixing potassium permanganate and oxalic acid, hydrogen peroxide is added after the decontamination is completed, and then the detergent is catalytically decomposed under ultraviolet light. The presence of oxalic acid can easily induce intergranular stress corrosion (IGSCC) and intergranular corrosion (IGA) on the decontaminated stainless steel. In addition, the storage and transportation of potassium permanganate also poses a certain degree of safety hazards. CAN-DEREM [29]: It is a decontamination method used in CANDU heavy water reactor, which was developed by the National Atomic Energy Company of Canada; this technology is to directly combine the compound pickling detergent (EDTA and citric acid) It is added to the equipment for decontamination, and then the decontaminated waste liquid is recycled through the ion exchange resin, which takes a relatively long time to decontaminate the equipment.

Foam decontamination method: In order to improve the problem of a large amount of secondary waste liquid generated by chemical cleaning and purification technology, Ayres [30] first used corrosive foam to remove radioactive contamination on metal surfaces in 1960. This technology is to prepare a chemical cleaning agent by mixing a foaming agent with an acid, alkali, oxidizing/reducing agent and a complexing agent. After foaming, the corrosive components in it will corrode and decompose the metal surface, so as to achieve the radionuclide Remove. In the decontamination of nuclear facilities, the core advantage of foam decontamination technology is that the amount of secondary waste generated is particularly small. The reason is that the volume of air in the foam occupies about 90%, and the process water consumption and detergent usage are very low, So the output of secondary waste is very low; the foam has strong permeability characteristics, and has strong cleaning ability on crevices and blind holes. The foam decontamination method is not limited to the shape of the decontamination object, so it can be applied to various complex decontamination environments, especially large tanks and various components with complex configurations. There are many research results on foam decontamination technology at home and abroad, and a large number of applications have been carried out in engineering. At present, it is quite mature. The most widely used foam cleaning technology abroad is the two-step foam decontamination device jointly developed by Britain and France [31]. The research on foam cleaning technology in China started relatively late, and the research on the foam cleaning system has been preliminarily completed so far, including the preparation of foam cleaning agent, the design and manufacture of construction facilities, and post-treatment research. Guan Haiyang [32] applied foam decontamination technology to the refueling tank in the pressurized water reactor nuclear power plant. Compared with the traditional decontamination technology in the past, it was found that the foam decontamination technology had better decontamination effect in the refueling tank. Its comprehensive advantages are as follows: the amount of secondary waste generated is reduced by more than 50%; the construction period is shortened from the original 5h to 3h, which saves costs; reduces the exposure dose of the operator; the decontamination efficiency is higher than that of the high-pressure water gun The flushing is high, from the original decontamination coefficient of 30 to 83. At present, there are two main development directions for foam decontamination technology: the use of biomass foaming instead of chemical foaming can effectively remove radioactive pollution and reduce environmental pollution; reduce the use of solvents to reduce the workload of the post-decontamination process. Research on solvent-free decontamination foam [33].

In gel decontamination, gel is a fluid whose viscosity decreases with time under the constant shear stress. When the shear stress is removed, the viscosity gradually recovers. This property is thixotropic, so that the gel is not only easy to spray, but also firmly adheres to the facade or top surface after spraying, that is, the gel has good construction performance. The gel has good construction performance and decontamination effect, so it can be widely used in the removal of radioactive contamination on different metal surfaces in nuclear facilities. For example, the metal cladding of the chemical equipment room, the metal cladding of the hot cell, the metal glove box, the wall of the reactor pool, and the large tank, etc. The gel decontamination technology was developed by three French companies and used in the radioactive decontamination of nuclear facilities, making the gel decontamination technology gradually mature [34]. France and the United States [35] were the first to apply gel decontamination technology to the removal of radioactive contamination on metal surfaces in nuclear facilities. Many practical results show that the gel decontamination method has excellent performance in radioactive decontamination of metal surfaces. On this basis, the Chinese

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Academy of Radiation Protection introduced and continued to develop this technology, made new progress in technology and equipment, and verified it in practice, and achieved good results. The advantages of gel decontamination technology are very significant. It can not only meet the decontamination requirements of different decontamination objects, but also generate a smaller amount of secondary waste. However, gel decontamination technology is more complicated and not easy to use on a large scale.

Liu Yuchen and others of the Chinese Academy of Radiation Protection [36] improved chemical gel decontamination. Based on their original work, plasticizers and polymer film-forming agents were added when preparing the gel, and then modified by blending. The principle of sex produces a peelable gel. This kind of peelable gel is more convenient to collect and process than the original gel, and it may be applied to the surface of lightly polluted materials. This method can remove contaminants on the surface of PCB boards, asbestos, etc., and is applicable to a variety of objects (concrete, plastic, wood, steel, etc.). The generated waste is recovered by vacuum cleaning technology, and the entire decontamination treatment system is relatively complicated [37]. Wang Zhentao [38] et al. used polyvinyl alcohol aqueous solutions to prepare film-forming agents, and used complexing agents, thickeners, plasticizers, surfactants and other preparation aids to study the removal of 90Sr radioactive pollutants on the surface of stainless steel. Stain effect.

4.3 Electrochemical decontamination method

The electrochemical decontamination method is to use electrolysis technology to immerse the contaminated surface in a certain electrolyte as the anode. The electrolysis corrodes the anode metal surface, so that the nuclide pollutants in the metal surface layer are dissolved into the electrolyte solution to achieve the purpose of removing nuclides. It is suitable for deep decontamination of carbon steel, stainless steel, aluminum and other metal surfaces produced during the decommissioning of nuclear facilities to achieve clean, decontamination and recycling. In principle, electrochemical decontamination technology can be understood as chemical decontamination under the combined action of an electric field. The harmful and toxic radioactive contaminants on the metal surface can be removed by electrochemical decontamination. It uses the principle of electrolysis, which can make the contaminants on the metal surface and the substrate fall off. In other words, it is to pass an electric current and then produce anode dissolution. If the conditions are right, the metal surface will become smoother and smoother. If the conditions are not appropriate, when the current density is too small, the metal surface may be etched; when the voltage and current density are too large, pitting corrosion may occur. In electrochemical decontamination technology, current density and electrolyte solution are particularly important. Existing studies have found that both are essential to decontamination benefits and decontamination effects. When this technology was first used in the United States, nitric acid was used as the electrolyte. However, due to the strong corrosiveness of nitric acid, the surface of the decontamination object is extremely uneven [39]. Later, phosphoric acid electrolyte was introduced. Its main advantages are high safety, good stability, and a larger scope of application, and the complexation of phosphate ions to metal cations is better. In addition, due to the non-volatility of phosphoric acid, airborne pollution can also be reduced. Yusuke Ohashi et al [40] used an electrochemical method to recover uranium from solid waste contaminated with uranium using 1-butyl-3 methylimidazole chloride as an electrolyte, which can convert the hexavalent uranium in the contaminated steel waste into five Valence uranium, and electrolytic recovery of dissolved uranium. The coating film electrolysis technology jointly developed by Sichuan University and the Institute of Engineering Physics can limit the removal of radioactive pollutants on the surface of metal materials. This technology can prevent pollutants from entering the electrolyte and entering the peelable membrane during the electrolytic decontamination process. The generation of radioactive waste liquid [41]. The mobile electrolytic decontamination engineering test device independently designed and processed by the Chinese Academy of Engineering Physics uses electrolytic decontamination technology to achieve deep decontamination of metal equipment contaminated by uranium and plutonium. The average decontamination efficiency for plutonium samples is 99.8%. These results

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showed that: using the specific electrolyte and process parameters obtained in the previous study, electrochemical decontamination and common decontamination Compared with the pollution method, it is faster and more efficient. For metal equipment with different degrees of pollution, the radioactivity can be lower than the detection limit of the instrument within five minutes. Lu Chunhai of the Chinese Academy of Engineering Physics [42] and others developed a set of 200L tank electrochemical decontamination device that can decontaminate surface radioactive contaminants based on the electrochemical decontamination method, which realizes the automatic control of the electrochemical decontamination tank.

4.4 Biological decontamination method

The biological decontamination method mainly uses the microbial degradation method. Generally, a brush, spray gun or roller is used to coat the microbial solution on the contaminated surface, allowing the microbes to pass through the surface and contact the contaminated surface. When the microorganisms completely consume the contaminants, detergents or solvents are used to wash away the reactants and most of the microorganisms. Drying will destroy the remaining microorganisms. If it is not damaged, heating or chemical treatment will deactivate the microorganisms. Finally, use fresh solvent to wash off the residual dirt or its derivatives on the decontaminated surface. It is necessary to maintain proper humidity on the surface during decontamination. At present, this technology has been successfully applied to mineral oil, alcohol, phenol, etc. in sludge tanks and cabins. Biological decontamination method is highly selective. The disadvantage is that the decontamination time is long and the decontamination effect is poor.

5. Conclusion

Decontamination methods mainly include physical decontamination, mechanical decontamination, chemical decontamination and biological decontamination, but they are diverse in terms of technology. As far as the current single technologies are concerned, there are always constraints of one kind or another, related to the substrate material, the decontamination object, the type of contamination, and so on. Combining various decontamination methods will often have better results. Therefore, a reasonable and effective combination of these decontamination methods has become the main direction for the development and application of decontamination technologies for decommissioning nuclear facilities in the future.

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