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Treatment Method of Radioactive Waste Liquid Containing U and Cs

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Abstract: Radioactive waste liquid will not only cause heavy metal and radiation pollution in the environment, but also enter the human body from a variety of ways, threatening human life and health. Among them, uranium and cesium, due to their high fission yield, long half-life and high solubility of salt compounds in water, need to be treated urgently. The treatment methods of radioactive u and C waste liquid, such as ion exchange method, solvent extraction method, adsorption method, membrane separation method, chemical precipitation method, biological treatment method, adsorption method and combined process, are combed. The principle, characteristics and application of the method are reviewed, and the research direction and development trend are prospected. It has a certain reference value for the selection of radioactive waste treatment methods in different situations.

Keywords: Radioactive Waste Liquid; Uranium and Cesium; Waste Liquid Treatment.

1. Introduction

Atomic energy, also known as nuclear energy, has become an important direction in the scientific field for over 100 years due to its advantages of efficiency, cleanliness, safety, and abundant reserves[1]. While nuclear energy brings convenience to humanity, some problems also arise, such as the Chernobyl nuclear leak in 1986 and the Fukushima nuclear power plant accident in 2011, which have shown people the potential for significant harm in the process of nuclear energy utilization[2].

In addition to nuclear accidents, radioactive substances generated during the nuclear fuel cycle can accumulate over a long period of time and have significant chemical toxicity. If these radioactive substances are allowed to enter the environment, they will cause great harm to animals, plants, and humans. At the same time, this radioactivity is potentially not fully exposed and is not easily detected.

Among these radioactive substances, radioactive wastewater is particularly prominent in radioactive waste treatment due to its high volume and total proportion, as well as its high fluidity, which is difficult to control. Its sources mainly include uranium mining and metallurgy, nuclear facility operation and spent fuel reprocessing, and the application process of radioactive isotopes. For medium and low level radioactive waste liquid, various technologies are generally used to purify it to meet the emission standards before being discharged into the environment.

2. Treatment Methods for Radioactive Wastewater Containing U

2.1. Hazards of Wastewater Containing U

Uranium is currently one of the most widely used nuclear fuels and exists in the form of U(IV) in primary ore deposits. In the process of uranium mining, processing, refinement, nuclear fuel manufacturing, and spent fuel reprocessing, hexavalent uranium with high environmental mobility and high diffusivity can diffuse into the environment. In addition, uranium has a long half-life, high chemical toxicity, and strong radioactivity, making it a common pollutant in soil, surface, groundwater, and ecological environment. Due to the use of strong acids, alkalis, and other chemical substances in a series of processes such as uranium mining, hydrometallurgy, and refining, the discharge of uranium-containing wastewater into the environment through surface runoff can disrupt the acid-base balance of water and soil, thereby affecting the animals and plants that survive in the water and soil. These organisms may enrich uranium through feeding and other pathways, and then enrich it layer by layer through the food chain to reach the top of the food chain for humans, The discharged wastewater can also enter the underground drinking water layer and enter the human body through underground runoff. After uranium enters the human body through these methods, it will cause internal radiation to various organs and tissues in the body. When the dose reaches a certain limit, it can induce various diseases, and even cause cancer and pathological reactions in multiple organs such as the spleen, lungs, and kidneys. From the above, it can be seen that radioactive uranium poses great potential hazards to both the environment and the human body[3], The removal of uranium from radioactive waste liquid is of great significance. Prior to its discharge, uranium containing wastewater must undergo deep evolutionary treatment to meet national discharge standards before being discharged (≤ 0.05 mg/L)[4].

2.2. Traditional Treatment Process for Wastewater Containing U

Various methods have been developed for the treatment of uranium containing radioactive waste generated during the nuclear fuel cycle, among which ion exchange, evaporation concentration, chemical precipitation, adsorption, and other methods are the most common.

2.2.1. Ion Exchange Method

The most commonly used materials in the ion exchange method are various ion exchange resins and ion exchange fibers. As early as the 1980s, ion exchange resins were used in uranium ore mining due to their good selectivity and high

desorption coefficient. However, they are difficult to degrade, radiation resistant, mechanically stable, expensive, and have poor recycling performance. They are also prone to interference from other components in other waste liquids, resulting in a decrease in their decontamination efficiency, So its use is limited. Compared with resins, ion exchange fibers have a larger specific surface area, shorter mass transfer distance, faster adsorption and desorption speed, better cycling performance, and a wider range of sources, which can effectively improve ion exchange performance. However, pure ion exchange fibers do not have very excellent performance, and they need to be modified by certain chemical means to increase the performance of ion exchange fibers[5,6].

2.2.2. Evaporation Concentration Method

The evaporation concentration method mainly achieves the purpose of reducing volume and concentration by heating and evaporating radioactive waste liquid, in order to prepare for the subsequent recovery and utilization of valuable nuclides or further solidification of evaporated sludge. This process is relatively convenient and has a high decontamination coefficient, but it may contain potential hazards such as steam entrainment of radioactive nuclides, corrosion, scaling, explosion of equipment caused by waste liquid, and radiation protection issues.

2.2.3. Chemical Precipitation Method

Chemical precipitation method refers to the method of purifying wastewater by adding chemical reagents (such as flocculants or coagulants) that can react with the target components in the wastewater and generate insoluble or insoluble substances. In this way, the radioactive nuclides in the wastewater will concentrate into small volumes of sediment due to precipitation, thereby reducing the activity of the radioactive wastewater. In addition, research has shown that the formed sediment will aggregate with insoluble substances present in the liquid phase to form another special type of insoluble substance. This insoluble substance not only adsorbs radioactive nuclides, but also removes other impurities in wastewater, such as suspended solids, organic matter, microorganisms, colloids, and constant salts[7]. For uranium containing wastewater, common flocculants currently include lime, ammonium salts, inorganic acid salts, aluminum salts, and iron salts, while coagulants include polyelectrolytes, clay, and active SiO_2[8]. Although the chemical precipitation method can have a certain purification effect on uranium containing wastewater, the insoluble waste residue generated after purification still needs further treatment, such as dehydration, solidification, etc. The operation is cumbersome and prone to secondary pollution, the treatment efficiency is limited, and many human and material resources are wasted in vain.

2.2.4. Adsorption Method

Adsorption is the process in which certain components in a sample are enriched and separated from the surface of a solid adsorbent through the action of molecular attraction (or molecular bonds) after contact with the sample. By adsorption, one or more radioactive nuclides in the wastewater are attached to the surface of the solid adsorbent, thereby achieving the purification of uranium containing wastewater [9], Adsorption method is an important physical and chemical separation technology that can effectively treat soluble pollutants. Commonly used adsorbents include activated carbon, kaolin, bentonite, zeolite, etc [10]. Recently, synthesized polymer materials, biomass, new carbon materials, and new materials (nano zero valent iron)[11] Adsorbents such as clay ore (montmorillonite) have also been extensively studied, and different adsorbents have their own unique advantages. The adsorption method is easy to operate and the adsorption material is cheap and easy to obtain, but there is a problem of saturated adsorption capacity during the use of the adsorbent [1], Frequent replacement is necessary, and post-treatment of the adsorbent after use is also a very important issue. If not handled properly, it can cause secondary pollution.

2.3. Emerging Treatment Process for Wastewater Containing U

With the continuous reform and innovation of China's technological level, stricter requirements have been placed on the treatment technology of uranium containing wastewater, which has led to the emergence of many emerging process technologies, including membrane treatment method, biological treatment method, molecular sieve uranium removal method, and pitchblende crystallization method [10,12].

2.3.1. Membrane Treatment Method

Membrane treatment can achieve the separation, extraction, concentration, and purification of pollutants. There are many different specifications of membranes, such as microfiltration, ultrafiltration, nanofiltration membranes, reverse osmosis, etc. Membrane separation technology has the advantages of convenient operation, simple equipment, low energy consumption, no phase change of materials, low energy consumption, high purification coefficient, and stable operation. Membrane separation technology is usually used in combination with other methods. In foreign countries, ultrafiltration technology is used as a pre-treatment method, and then combined with reverse osmosis technology or ion exchange technology to treat radioactive wastewater, playing a deeper purification role in radioactive wastewater, At the same time, it also reduces the generation of secondary waste, effectively reducing environmental pressure while bringing good social and economic benefits[13]. Ma Haitao et al.[14] are proposed to introduce membrane treatment technology into the treatment process of uranium and fluorine containing wastewater. Membrane treatment technology is added between adsorption treatment and slaked lime precipitation. After adsorption and recovery of uranium, the wastewater is first treated by ultrafiltration and nanofiltration to further improve the uranium recovery rate. Then, fluorine ions are intercepted on the concentrated water side through two-stage reverse osmosis, and slaked lime precipitation is sent to form calcium fluoride slag. Compared with traditional processes, the wastewater discharge is reduced Improved uranium recovery rate while purifying wastewater into fresh water for recycling, saving resources[15]. However, the application of membrane technology in wastewater treatment also has the following shortcomings: the compatibility of the membrane is largely influenced by factors such as pore size, pH of the waste liquid, and temperature, and the investment cost of membrane technology is higher than other methods. In addition, the lifespan of the membrane will be greatly shortened when treating certain samples that are prone to scaling.

2.3.2. Biological Treatment Method

Biological treatment method is a new technology

developed in recent years, which has rapidly developed and achieved some significant achievements, mainly including microbial adsorption method and phytoremediation method. Since the 1960s, the use of microbial materials to treat reflective wastewater containing radioactive nuclides has been studied. This technology is mainly developed based on the outstanding advantages of microbial materials such as good selective adsorption performance, fast adsorption rate, good temperature and pH adaptability, high efficiency, wide sources, and no secondary pollution. Therefore, the use of biotechnology has great potential for development in the treatment of radioactive waste liquid contaminated with uranium. Microbial enrichment of uranium is mainly based on indirect non metabolic biological adsorption, with metabolic enrichment being secondary[16]. Data shows that microorganisms such as beer yeast[17], algae[18], radiation resistant Staphylococcus[19,20], anaerobic granular sludge, powder activated sludge, sulfate reducing bacteria granular sludge, Escherichia coli, yeast, Bacillus subtilis, and Chlorella have high adsorption capacity for uranium in water, with a removal rate of over 90%[21,22]. Microbial methods are currently mostly in the experimental research stage and have not yet been widely applied.

Plants and their coexisting microorganisms work together to adsorb, degrade, and enrich pollutants in the environment, thus achieving the goal of environmental pollution control. Phytoremediation methods have the advantages of low implementation threshold, low capital density, low energy consumption, strong environmental compatibility, and good metal recovery, playing an important role in the purification and treatment of low-level pollutants. It is precisely because of these unique advantages that phytoremediation methods are increasingly widely used in the treatment of large-scale water pollution. Scholars have pointed out that forage, Indian mustard, water sedge, pickled mustard, and American pokeweed all have strong uranium enrichment capabilities and are widely used in the study of uranium contaminated soil background restoration. Nie Xiaoqin et al.[23] found in their experiment that indigenous aquatic plants in uranium containing areas can treat uranium containing wastewater with an initial concentration of 4.5 mg/L to meet the national discharge standards. Duckweed has a rich source, mainly found in lakes and rivers of all sizes. Zhang Rujin et al.[24] studied the effects of formic acid, malic acid, and citric acid treatments on the adsorption of uranium by duckweed. Through a blank control experiment, it was found that the saturated adsorption capacity of uranium by duckweed under the three acid treatments was significantly increased, Among the best adsorption effect of duckweed is achieved under the treatment of 0.4×10^{-5} mol/mL formic acid, indicating that duckweed has a very broad prospect in the treatment of uranium containing wastewater.

2.3.3. Molecular Sieve Uranium Removal Method

Molecular sieve is a porous material that has been extensively studied. Lee S[25] chose fly ash as the substrate material and prepared NaY type nano molecular sieves using ultrasonic assisted alkali melt hydrothermal method. The application of NaY type nano molecular sieves in the adsorption of radioactive wastewater was studied. The results showed that the adsorption material had excellent effects on uranium removal at pH=4, oscillation time of 20 minutes, and uranium concentration of 100 mg/L, with an adsorption capacity of 21.23 mg/g. Molecular sieves have a wide variety of types, low cost, large adsorption capacity, and strong reusability. In recent decades, NaY type molecular sieves have rapidly developed and are widely used in various fields such as catalysis and ion exchange.

2.3.4. Pitchblende Crystallization Method

The main principle of the pitchblende crystallization method is to produce relatively stable pitchblende or other insoluble substances in the uranium containing wastewater through chemical treatment, thereby achieving the treatment of uranium containing wastewater. This method draws inspiration from the mineralization conditions of naturally occurring pitchblende, reflecting the waste treatment concept of nuclear waste returning to nature. It is a new uranium purification method generated on the basis of traditional technology. Cui Chunlong et al. [26] investigated the treatment of uranium in the liquid phase by the crystallization method of pitchblende, and set a series of different influencing factors. The results showed that increasing temperature and decreasing Eh accelerated the uranium removal rate and improved efficiency. Among them, under physical and chemical conditions of T=85℃, pH=3, and Eh= +61.1 mV, uranium element precipitated from the solution in the form of pitchblende, with a uranium removal rate of up to 99.6% and a good effect.

3. Treatment Methods for Radioactive Wastewater Containing Cs

3.1. Hazards of Wastewater Containing U

Radioactive wastewater containing Cs comes from a wide range of sources, such as the treatment process of nuclear fuel in industrial applications such as nuclear power plants, nuclear fission products from atomic power plants, and research institutions that use radioactive isotopes. Its water content can reach hundreds of kilograms to tens of thousands of tons. Among, ^{137}Cs are β and γ main source of radiation, the proportion of its radioactivity in the total radioactivity of fission products increases with the prolongation of decay time. Liquid waste treatment is a gradient process, which first reduces high radioactive wastewater to medium to low radioactive wastewater. After further treatment, standard wastewater can be discharged or reused. The concentrated product is solidified and then deeply buried for disposal[27].

3.2. Research on the Treatment of Wastewater Containing Cs

3.2.1. Chemical Precipitation Method

The chemical precipitation method is based on the solubility product theory. The smaller the solubility product of a compound, the easier it is to generate precipitation. Therefore, selecting a suitable precipitant is crucial. In order to avoid organic pollution, inorganic substances are the preferred precipitant. Table 1 lists the solubility products(K_{sp}) of inorganic insoluble compounds of cesium at 25 ℃.

Table 1. Solubility product of insoluble compounds in Cs

chemical compound	K_{sp}	chemical compound	K_{sp}
$Cs_3[Co(NO_2)_6]$	5.7×10^{-16}	$CsClO_4$	4.0×10^{-3}
$Cs_2[PtCl_6]$	3.2×10^{-8}	$CsBrO_3$	5.0×10^{-2}
$Cs_2[PtF_6]$	2.4×10^{-6}	$CsIO_4$	4.3×10^{-3}
$Cs_2[SiF_6]$	1.3×10^{-5}	$CsMnO_4$	8.2×10^{-5}
$Cs[BF_4]$	5.0×10^{-5}	$CsReO_4$	4.0×10^{-4}

From Table 1, can be seen that $[Co(NO_2)_6]^{3-}$ can react with

Cs^+ to generate precipitation to remove Cs^+. According to $Ksp=[c(Cs^+)]^3\times c([Co(NO_2)_6]^{3-})$ calculation, the required concentration of $[Co(NO_2)_6]^{3-}$ is very high, and there is no feasibility of application, nor is there any literature report on this method. Platinum compounds are expensive and not suitable as precipitants. Therefore, the possibility of finding a precipitant for cesium in inorganic materials is extremely low.

D. J. McCabe's research has shown that NaTPB can react with Cs^+, and the K_{sp} of CsTPB at 25℃ is 1.0×10^{-10}. According to the K_{sp} calculation, it can be concluded that TPB as a precipitant is completely feasible [28]. R. A. Peterson et al. calculated the radioactivity to be 1.85×10^{10} Bq/L of high-level cesium containing wastewater from the Savannah River in the United States and 0.31 mol/L of NaTPB solution were added to the container at different flow rates, and stirred at 400 r/min for 30 minutes. Considering the high radioactive activity of the initial wastewater, this method DF>105. M. F. Debreuille et al. sent flammable gases such as benzene generated during the reaction to the incinerator for disposal. This technology has been industrialized and applied in the United States, with a temperature of 20-30℃, a residence time of 0.5-2 h, initial concentration of Cs^+ is 1×10^{-4} mol/L, and 50% excess of NaTPB substance, the DF exceeds 1000. E. H. Lee et al. used NaTPB to treat wastewater containing simulated Cs^+. When the pH is 6.3-13.2 and the initial concentration ratio of NaTPB to Cs^+ is greater than 1, when mixed and stirred for about 10 minutes, 99% of Cs will be generated as precipitates from the liquid phase, and the temperature (25-50℃) and stirring speed (400-1000 r/min) have no effect on the precipitation amount[29]. When NaTPB is used to treat high level radioactive waste water containing cesium, the reaction time is short and the sedimentation effect is good, but foam will be generated when running in the reactor, because the organic parts such as TPB will generate benzene, phenyl-boron, phenol and other flammable volatile products after being irradiated. These decomposition products pose potential safety risks to this method.

3.2.2. Ion Exchange Method

The existing form of Cs in the aqueous phase is a monovalent cation, which belongs to the same main group element as K^+. Therefore, the cation exchange method can be used to treat Cs elements. Organic ion exchangers are prone to damage under high temperatures and ionizing radiation, which limits their application; Inorganic exchange materials are easy to operate, and the ion exchange positions of inorganic crystals are more uniform. These properties make the material have different specificity for specific elements. In recent years, inorganic ion exchange agents that have been extensively studied include artificial zeolites and clay minerals, heteropolyacids and composite ion exchange materials, metal ferrocyanide, titanium silicon compounds.

3.2.3. Zeolite and Clay Minerals

Qiu L et al.[30] shows that compared with naturally occurring clinoptilolite, natural mordenite, and synthetic mordenite, natural rhombite has a better adsorption effect on cesium with a distribution coefficient K_d (K_d can reflect the migration ability and separation efficiency of the adsorbate in the solid and liquid phases). For a radioactive activity of ^{134}Cs solution is 2.28×10^4 Bq/L, with a natural zeolite dosage of 0.01 g/L, has a K_d for ^{134}Cs as 4.97×10^3 mL/g. JiaojiaoWu et al.[31] treated cesium nitrate solution with montmorillonite at a concentration of 30 μg/L When the dosage of montmorillonite is 20 g/L, the adsorption rate of cesium at room temperature exceeds 98%, and dynamic equilibrium can be achieved within 5 minutes. The adsorption is better than the composite Langmuir isotherm model. The pH value and salinity of the solution have a certain effect on the exchange capacity of the material. Under high salinity and strong acidity, the exchange capacity of cesium is low. Therefore, this type of ion exchange agent is more suitable for treating radioactive wastewater with low acidity and low salt content.

3.2.4. Polyvalent Metal Phosphates and Composite Ion Exchange Materials

S. A. Shady synthesized organic composite ion exchanger resorcinol formaldehyde (R-F) and inorganic composite ion exchanger zirconia ammonium pyromolybdate (ZMPP), and investigated their exchange ability for Cs, Co, Zn, and Eu. The results show that the selective exchange order of ions between R-F and ZMPP is $Cs^+>Co^{2+}>Eu^{3+}>Zn^{2+}$[32]. At the same pH, R-F has a higher K_d for cesium. When the pH is 7.21, the K_d of R-F is 6.4×10^3 mL/g, while the K_d of ZMPP is 158 mL/g. Y. J. Park et al. studied ammonium molybdate polyacrylonitrile as a removal agent, investigated the removal efficiency of different nuclides, and investigated the effects of coexisting ions and surfactants on the removal efficiency. According to the results, AMP-PAN has an adsorption capacity of 0.61 mmol/g for Cs under optimal conditions, and Na^+ and anionic and cationic surfactants can reduce the adsorption capacity. Polyvalent metal phosphates are susceptible to interference from coexisting Na^+, which affects the treatment effect. The removal effect of organic composite ion exchange materials on cesium is relatively good, but their radiation resistance is low, and the subsequent treatment of concentrated products is difficult.

3.2.5. Metal Ferrocyanide

Zhang Qi et al.[33] conducted experiments on the treatment of Cs isotope ^{134}Cs in low-level radioactive waste liquid and explored the effects of different conditions on removal. According to the results, when the pH is 1-10 and the initial radioactive activity of ^{134}Cs is 3.2-160.0 kBq/L, the adsorption equilibrium is reached within 60 minutes, with an adsorption amount of 9.6-463.0 kBq/g. When the temperature is 0-50℃ and Ca^{2+}, Fe^{3+}, Mg^{2+}, HCO_3^-, CO_3^{2-}, Cl^-, SO_4^{2-} are present, the removal effect of zinc ferrocyanide on ^{134}Cs is not affected. However, when the solution contains K^+ or Na^+, the removal rate of ^{134}Cs decreases. Zhang Xiaoxia et al.[34] used potassium zinc ferrocyanide to remove cesium from simulated wastewater. When the mass concentration of Cs^+ is 100 μg/L, temperature is 15℃, and contact time is set to 60 minutes, the adsorption effect of cesium can reach about 98%, with a DF of 76; V. V. Milyutin et al. studied the co precipitation of trace amounts of cesium (10^5 Bq/L) with ferrous cyanide of different metals. When the molar ratio of Me^+ to $Fe(CN)_6^{4-}$ is 1.33, the distribution coefficient of ferrous cyanide on ^{137}Cs is shown in Table 2.

Table 2. Distribution Coefficients of Metal Ferrocyanide on ^{137}Cs

10^5 mL/g					
metal ion	K_d	metal ion	K_d	metal ion	K_d
Mg^{2+}	1.7	Ni^{2+}	200	ZrO^{2+}	0.5
Ca^{2+}	0.6	Cu^{2+}	10	MnO_2^{2+}	1.0
Sr^{2+}	0.7	Zn^{2+}	1.5	Al^{3+}	1.5
Ba^{2+}	0.6	Cd^{2+}	14	Cr^{3+}	5.2
Mn^{2+}	2.5	Hg^{2+}	1.2	Fe^{3+}	1.1
Fe^{2+}	42	Pb^{2+}	0.5	Ce^{3+}	0.7
Co^{2+}	24	UO_2^{2+}	54	Bi^{3+}	0.5

Table 2 shows that the divalent transition metals (Ni^{2+}, Fe^{2+}, Co^{2+}, Cd^{2+}, Cu^{2+}) and uranyl ferrocyanide have higher distribution coefficients. Abdel Al M et al. [35] fixed $Me_2Fe(CN)_6$ and $MeK_2Fe(CN)_6$ (Me = Cu, Co, Ni) onto carboxylic latex to form a stable colloidal adsorbent to remove cesium from the solution. This adsorbent can penetrate porous media, so this substance can be applied in the treatment of solid waste contaminated with Cs. The synthesis process of this compound is simple, the raw materials are easy to obtain, and the cost is low. However, the ability of ferrocyanide to remove cesium varies among different metals, with transition metal ferrocyanide having the best effect. In application, suitable metal ferrocyanide can be selected based on the characteristics and treatment requirements of wastewater.

3.2.6. Titanium Silicon Compound

Kubota et al.[36] first studied and prepared crystalline titanium silicon compounds (CST or TAM-5), and the results showed that CST can selectively remove Cs^+ from 5.7 mol/L Na^+ solution; When treating a mixed solution containing 5.7 mol/L $NaNO_3$, 100 mg/LCs^+, and 20 mg/LSr^{2+}, the distribution coefficient of CST for Cs^+ was $>10^5$ mL/g. S. Solbra et al. prepared hydrated sodium titanium silicate STS and investigated its removal effect on cesium. From the results, it can be seen that STS has good ion exchange performance for cesium under acidic, neutral, and alkaline conditions, with a K_d of up to 10^6 mL/g. When using STS to treat Cs^+ Ringer solution with an initial mass concentration of 6.0 mg/L, the DF>1000 is obtained at a dosage of 0.01~0.001 g/mL. The research on titanium silicates in China started relatively late. Wang Xuqian et al.[37] synthesized a new type of crystalline hydrated titanium silicate (CST) by hydrothermal method. The exchange capacity of CST for Cs^+ in 1 mol/L HNO_3 solution was 0.63 mmol/g. Although there is still a gap in the effectiveness of cesium removal in domestic synthesized ion exchange agents, titanium silicon compounds are still a promising new type of ion exchange agent.

4. Conclusion

1) Further optimize the traditional radioactive wastewater treatment process, selecting different treatment and disposal methods based on different objects to achieve the goal of reduction, resource utilization, and harmless treatment.

2) Invest more energy and attention in developing safe and efficient membrane separation combination processes, achieving a high degree of automation control during operation.

With the continuous maturity and improvement of existing technologies and the rational utilization of some new technologies and methods, the future use of nuclear energy will be safer and more reliable.

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