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A decontamination technique for the primary cooling circuit of the research type nuclear reactor

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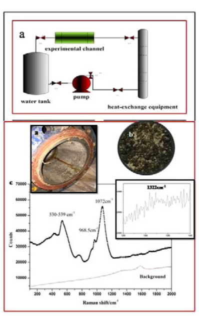
GRAPHICAL ABSTRACT

Accumulation of radioactive corrosive materials in the primary loop of reactors will lead to elevated radiation level in workplace, increased maintenance and repair costs, and greater difficulty in later decommissioning. This study analyzed the contaminants deposited on the inner walls of pipes serving a reactor by Raman microscopy. The results show that the main contaminants were $FeCO_3$ and “green rusts” and γ-FeOOH may exist. A laboratory decontamination experiment was conducted on samples from the contaminated piping using two decontaminants: decontaminant A, composed primarily of HNO_3, and decontaminant B, composed primarily of $H_2C_2O_4$. The structural material of the reactor’s cooling circuit piping restores its metallic luster after decontamination, demonstrating the effectiveness of the two decontaminants. The contaminants identified can be divided into two categories: soft materials that were easy to remove and hard materials that were firmly attached to pipe wall. The curves describing the activity of ^{137}Cs present in the solutions reveal that the decontamination process with decontaminant A or B consisted of a rapid reaction stage and a slow reaction stage. This indicates that there could be various oxides in the contaminants. An online decontamination experiment was performed on an analog primary loop. The monitoring data suggests that the decontamination method determined under laboratory conditions is applicable to online decontamination of operating cooling loops of reactors. Furthermore, the study found that the fluctuations caused by liquid motion within large devices can affect the activity of ^{137}Cs in the solutions.



1. Introduction

During operation of a reactor, the primary cooling loop is responsible for carrying heat away from the reactor core to control its temperature, so as to ensure stable nuclear energy output and reactor safety and prevent accidents resulting from overheating of the core. Therefore, the service reliability of primary loop has a direct bearing on the safety and efficiency of nuclear reactors. During operation, the water coolant in the primary loop flows through the reactor core and absorbs heat, thus becoming hot water. The hot water then flows through the primary loop piping into the steam generator, heat exchanger and other devices. After being cooled, the water is then returned to the core for another cycle. In this process, structural materials of the pipes, valves, heat exchanger, steam generator and other devices in the primary loop will inevitably undergo various types of corrosion, such as general corrosion and stress corrosion cracking (Gordon, 2013; Berry and Diegle, 1979), in a high-temperature and high-pressure environment (pressurized water reactors) (Muzeaua et al., 2011), or medium-temperature and atmospheric-pressure environment (some tank reactors). The corrosion products will be carried by the liquid coolant to the reactor core. Upon exposure to the radiation from the core, elements like Fe, Co, and Ni in the corrosion products will be

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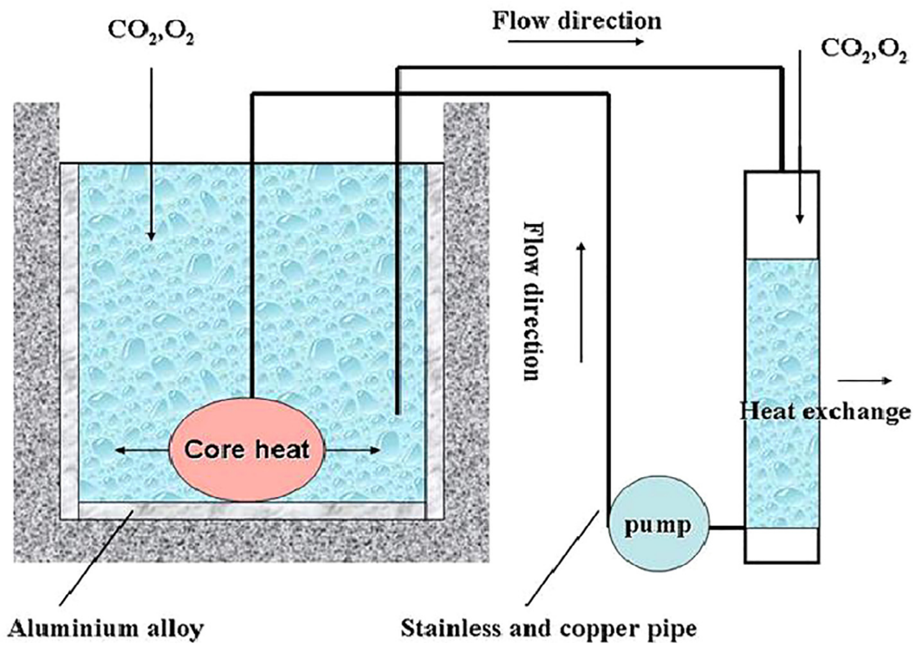

Fig. 1. A schematic diagram of the structure of the experimental nuclear reactor in China.

activated and result in activation products (Rafique et al., 2005; Varga et al., 2001). Moreover, the structural materials around the core will also be activated and the activation products will also enter the liquid coolant. In the case of fuel element failure during reactor operation, fusion products can leak into the liquid coolant (El-Jaby et al., 2010). These radioactive materials are transported by the liquid coolant into the pipes and other components of the primary loop outside the reactor core, where they will be deposited along with the corrosion products and scales. The resulting dose accumulation can increase the radiation level in the workplace, posing a threat to the health of the personnel working there and the safety of the reactor system. Therefore, regular decontamination must be done for the pipes and components of a reactor primary loop in order to reduce the level of radioactive contamination and guarantee the system's operational safety. The present paper systematically describes the research and development process for a primary loop decontamination technique applicable to the research reactor developed by China. This is China's first effort to study and apply online decontamination for research reactor cooling system. This reactor is a typical experimental tank reactor and uses deionized light water as the coolant. Its structure is shown in Fig. 1. The main coolant circulation devices are all positioned outside the nuclear island and the primary cooling loop was designed to operate in medium-temperature, atmospheric-pressure conditions. As can be seen in Fig. 1, the main corrosive medium to which the cooling system is exposed is medium-temperature water at atmospheric pressure. The concentrations of oxygen, nitrogen and carbon dioxide dissolved in the water are in equilibrium with their partial pressures in the atmosphere.

The main structural material of the cooling loop is 1Cr18Ni9Ti, which corresponds to the ASSI 321 stainless steel specified in a United States standard. Its nominal chemical composition is provided in Table 1. The major sources of radiation in the cooling loop include activation products and the fission products that leak from failed fuel elements in the reactor core. Experimental research on online decontamination technology was conducted, in order find a solution to reducing the level of radioactivity in the workplace and the amount of radioactive waste produced during decommissioning and dismantling.

2. Experiments

2.1. Sampling from operating reactor components

All samples included in this study were collected from operating pipes, elbows and valves that served the reactor. The metal components taken from the reactor for sampling were divided into two groups: one group was for use in the laboratory experiment while the other group was preserved for later use after being cleaned. The components for use in the experimental were split into small pieces using plasma cutting or a dedicated reciprocating saw. Then the pieces of materials were shaped by precision lathes into circular (with 5 cm length and the same diameter as the original component) or rectangular (4 cm × 5 cm) samples. After that, the samples went through a cleaning process using deionized water, acetone, absolute ethyl alcohol, and deionized water in sequence. The levels of radiation (α, β, and γ rays) from all the components and samples collected were carefully measured in every step of the preparation process.

2.2. Structural characterization of deposits on sample surfaces

After cutting, the samples were put into sealed containers for storing quartz samples to prevent radioactive leaks. Then Raman microscopy, a non-destructive and non-contact testing method, was performed on these samples to analyze the chemical composition of the deposits on surfaces of the metal samples.

Table 1
The chemical composition of the cooling loop structure material.

Elements	C	Si	Mn	S	P	Cr	Ni	Ti	Fe
Contents (wt%)	0.12	0.80	2.00	0.025	0.035	17.0–19.0	8.0–11.0	0.50–0.80	balance