Study on Water Chemistry Control Strategy for Ammonia-Containing Primary Coolant

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Abstract

In the primary coolant of specific reactor types, pH is regulated by adding ammonia, and its radiolysis products are utilized to suppress the concentration of oxidizing substances, thereby maintaining a reducing state of the coolant. During this process, the ammonia concentration continuously changes under the influence of radiation, exerting a significant coupled effect on pH regulation. To address this issue, this work developed a water chemistry control model for simulating the transport process of coolant radiolysis products in arbitrary reactor types based on the RETA reactor system analysis code. Using the KLT-40S reactor type as a reference, the regulatory effects, advantages, and disadvantages of initial dispersed ammonia addition, constant-rate source ammonia addition, and hydrogen-removal-optimized constant-rate source ammonia addition strategies were simulated and analyzed. The results show that the initial dispersed ammonia addition strategy is simple and straightforward, capable of maintaining system reducibility but with limited duration for coolant pH regulation; constant-rate source ammonia addition can effectively regulate coolant pH over extended periods but introduces the problem of excessively high dissolved hydrogen concentration, requiring a matching hydrogen removal scheme. Adopting the hydrogen-removal-optimized constant-rate source ammonia addition strategy can simultaneously satisfy the requirements for stable control of both pH and dissolved hydrogen concentration, with an ammonia addition rate of 1.64 $g \cdot s^{-1}$, hydrogen removal device activated 1200 s after ammonia addition begins, a hydrogen removal rate of 0.014 $g \cdot s^{-1}$, and after chemical stabilization, the coolant pHT is 6.9 with dissolved hydrogen concentration of 30–35 $mL \cdot kg^{-1}$(STP). This work is expected to provide references for new reactor type development and optimization of water chemistry control strategies.

Full Text

Preamble

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Study on Water Chemistry Control Strategy for Ammonia-Containing Coolant in the Primary Circuit

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Abstract

[**Background**] In certain reactor types, ammonia is added to the primary coolant to regulate pH, and its radiolytic products are utilized to suppress the concentration of oxidizing species, thereby maintaining the coolant in a reducing state. During this process, the ammonia concentration continuously changes under radiation, exerting a significant coupled influence on pH regulation. [**Purpose**] To address this issue, this study developed a water chemistry control model based on the RETA reactor system analysis code to simulate the transport processes of coolant radiolytic products in any reactor type. [**Methods**] Using the KLT-40S reactor as a reference, the regulatory effectiveness, advantages, and disadvantages of three strategies were simulated and analyzed: initial ammonia dispersion, constant-rate ammonia injection, and constant-rate injection optimized with hydrogen removal. [**Results**] The results show that the initial ammonia dispersion strategy is simple and direct, capable of maintaining a reducing environment but with limited duration for effective coolant pH control. Constant-rate ammonia injection can regulate coolant pH effectively over extended periods but leads to excessively high dissolved hydrogen concentrations, requiring a corresponding hydrogen removal scheme. The hydrogen-removal-optimized constant-rate injection strategy can simultaneously satisfy the requirements for stable pH control and dissolved hydrogen concentration maintenance, with an ammonia injection rate of 1.64 $g \cdot s^{-1}$, hydrogen removal initiated 1200 s after injection begins at a rate of 0.014 $g \cdot s^{-1}$, resulting in a stable coolant pHT of 6.9 and dissolved hydrogen concentration of 30–35 $mL \cdot kg^{-1}$ (STP). [**Conclusions**] This work is expected to provide valuable reference for the development of new reactor types and the optimization of water chemistry control strategies.

Keywords: Coolant, Reactor, Ammonia, Radiolysis, Water Chemistry

Introduction

Small Modular Reactors (SMRs) have demonstrated promising application prospects in specialized fields such as icebreakers and floating nuclear power plants due to their compact structure, light weight, and flexible deployment. However, the highly integrated and compact design of SMR loop systems imposes more stringent requirements on radiation protection and material service safety compared to conventional reactors. In particular, the generation and accumulation of radiolytic products, such as reactive species including oxygen (O_2) and hydrogen peroxide (H_2O_2), may adversely affect the chemical stability and operational safety of the system. Therefore, to deeply understand the generation mechanism of radiolytic products and their migration and transformation behavior in the coolant system, it is essential to obtain accurate thermal-hydraulic parameters and tightly couple them with water chemistry processes to achieve comprehensive analysis of radiochemical effects in SMR coolant systems.

The issue of irradiation in reactors has long been recognized. Scott' s research demonstrated that, in addition to direct radiation damage to materials, coolant radiolysis is also a key factor causing material corrosion. Radiolysis refers to the production of various chemical species from water in the core region under high-flux neutron and gamma radiation. Water chemistry issues primarily focus on the generation, transport, and reaction behavior of these radiolytic products. While most products have extremely short lifetimes, some relatively stable species (such as H_2O_2 and O_2) can persist in the system and potentially exert adverse effects on water quality. As these oxidizing species migrate through the primary circuit, they may cause severe oxidative corrosion, posing a potential threat to critical thin-walled structures such as stainless steel heat exchanger tubes. Recently, Sultana et al. conducted experimental studies on radiolysis effects in supercritical water (SCW) and noted that similar issues may exist in SMR systems.

Since water chemistry issues have received widespread attention, the radiolysis process of coolant has been extensively studied. Ershov et al. pioneered and validated a kinetic model applicable to pure water radiolysis, systematically describing the generation mechanisms of major radiolytic products including H_2, H_2O_2, and O_2. Subsequently, Joseph et al. further investigated radiolysis effects in pure water through extensive experiments and developed a kinetic model capable of predicting the generation behavior of multiple products. Elliot compiled and reported 60 key radiolysis reactions and their rate constants, providing an important foundation for water chemistry simulation calculations.

Subsequent research has focused on achieving better agreement with experimental results and broader simulation scope. Yakabuskie introduced mass transfer processes at two-phase interfaces to modify and improve the model based on Joseph' s work. Palfi investigated the generation processes and yields of transient products in aqueous solutions under pulse irradiation conditions using a

combined experimental and computational approach. Karditsas studied water radiolysis in ITER fusion reactors, proposing a time-dependent transport model and providing calculation examples. Yousefi extended the application scope of radiolysis models to high-temperature and high-pressure conditions, providing a theoretical basis for analyzing radiolysis behavior under more complex operating conditions.

Another research focus has been on influencing factors and control strategies for radiolytic products. Roth and Daub analyzed the effect of pH on H_2O_2 generation and carbon steel corrosion behavior. Dey reviewed the transformation processes of nitrogen-containing substances in aqueous solutions under irradiation conditions, proposing that NH_3 has the potential to regulate oxidizing species and providing a new possible means for corrosion control. Building upon this, Yakabuskie studied the radiolysis behavior of NO_3^- solutions, while Guo analyzed the radiolysis process of ammonia solutions under various influencing factors. Kumar evaluated the role of N_2H_4 in corrosion control, and Iwamatsu investigated the effectiveness of H_2 in suppressing H_2O_2 accumulation.

The aforementioned work primarily focused on homogeneous zero-dimensional systems, neglecting spatial distribution effects and lacking guidance for loop analysis. Considering that radiolytic products are generated in the core and transported through the primary circuit, their concentration evolution can be calculated by combining mass, momentum, and energy equations. In this work, integrating radiation chemistry, thermal-hydraulics, and numerical computation research, a model was developed to simulate the coupled thermal-hydraulic and water chemistry effects in SMRs. This model is based on the RETA system analysis code with an added submodule for water chemistry reaction and transport calculations. The reliability and performance of the model were subsequently verified and evaluated, and the primary circuit of the KLT-40S reactor was modeled to analyze the regulatory effectiveness and pros and cons of two ammonia addition strategies. This work is expected to provide reference for optimizing ammonia-containing water chemistry control strategies.

1. Model Development

1.1 Radiolysis Process

The radiolysis process can be summarized in three stages: (1) radiation deposits energy in the coolant, leading to the production of a series of excited species such as H_2O^*, H_2O^+, and e^-; (2) excited species react to generate intermediate products such as $\cdot H$ and $\cdot OH$ radicals; and (3) excited species and intermediate products interact to generate final radiolytic products that diffuse out of the spur, such as H_2O_2, O_2, and H_2 (as shown in R0).

For zero-dimensional calculation of coolant radiolysis, the G-value is introduced to convert the radiation dose rate into a source term for radiolytic product generation, defined as:

$$R_{i,r} = \frac{\rho \dot{D} G_i}{N_A e}$$

where $R_{i,r}$ is the radiolytic production rate of species i ($\mathrm{mol \cdot L^{-1} \cdot s^{-1}}$), ρ represents the coolant density ($\mathrm{kg \cdot L^{-1}}$), e is the electron charge (1.9$×10^{-19}$ C), G_i is the radiation chemical yield of species i (/100 eV), $\dot{D}$ is the absorbed dose rate ($\mathrm{Gy \cdot s^{-1}}$), and N_A is Avogadro' s constant (6.02$×10^{23}$ $\mathrm{mol^{-1}}$).

Under continuous irradiation, various species react with each other. For species i, it is continuously generated and consumed during radiolysis, and its net generation can be expressed by E2:

$$\frac{dC_i}{dt} = R_{i,r} + \sum_{s=1}^{N} \sum_{m=1}^{M} k_{sm} C_s C_m - \sum_{s=1}^{N} k_{si} C_s C_i$$

where k_{sm} is the rate constant for reactions between species s and m (forming i) or between s and i (consuming i), C_m, C_s, and C_i are the concentrations of species m, s, and i, respectively, N is the total number of reaction equations, and M is the number of species involved in the reactions.

1.2 Thermal-Hydraulic Model

For engineering applications, zero-dimensional radiolysis calculations are insufficient and should be extended to at least the primary circuit and its main components. This means that in addition to radiolysis reactions, coupled thermal-hydraulic mass transfer calculations are required. The simplified primary circuit model and mesh division used in this work are shown in [Figure 1: see original paper].

To numerically solve the equations and integrate radiation chemistry into reactor analysis, programming was performed based on the RETA software from the University of Science and Technology of China, employing the fully implicit numerical discretization method of the Finite Volume Method (FVM). The control equations were solved using the Preconditioned Jacobian-Free Newton-Krylov (PJFNK) algorithm. Based on RETA, an additional module named R-WRC was developed, focusing on radiolysis process calculations and capable of coupling with RETA' s core functionality for system thermal-hydraulics. The implementation in R-WRC involves direct programming to include transport terms:

$$\frac{\partial \mathbf{C}}{\partial t} + \nabla \cdot (\mathbf{u}\mathbf{C}) = \nabla \cdot (\mathbf{D} \nabla \mathbf{C}) + \mathbf{R}_r$$

where $\mathbf{C}$ is the species concentration vector, $\mathbf{u}$ is the local velocity vector, $\mathbf{D}$ is the diffusion coefficient for different species, and $\mathbf{R}_r$ is the source term for radiation-induced products.

Parameters such as **u** are crucial in thermal-hydraulic calculations. When coupling radiolysis processes with thermal-hydraulics, the following assumptions were made to simplify the calculations: (1) only one-dimensional equations were considered, corresponding to the one-dimensional mesh in RETA system thermal-hydraulic simulations, similar to RELAP; and (2) diffusion terms were neglected, as the velocity in the main circuit is sufficiently high to ignore species diffusion processes.

Based on these two assumptions, the simplified equation becomes:

$$\frac{\partial \mathbf{C}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{C} = \mathbf{R}_r$$

Due to the large number of mesh nodes that need to be considered in the solution process, making it extremely challenging, the following programming structure was established: (1) each species was processed separately at all mesh nodes; (2) all species were combined into a unified vector; and (3) a large sparse matrix was used.

1.3 Model Validation

To verify the reliability of the R-WRC module in radiolysis process calculations, its results were compared with simulation data from the dedicated radiation chemistry software FACSIMILE, as shown in [Figure 2: see original paper]. Both R-WRC and FACSIMILE were used to simulate the radiolysis behavior of ammonia solutions with initial concentrations of 1–3 $\mathrm{mol \cdot L^{-1}}$ at 25 °C, comparing the variations in H^+ concentration (pH), ammonia concentration, and N_2 concentration with irradiation time. The results indicate that R-WRC can accurately describe the radiolysis process, with minimal differences from FACSIMILE, demonstrating good computational accuracy and reliability.

To further validate the model' s reliability, radiolysis experiments were conducted using ammonia solutions with an initial concentration of $30\,\mathrm{mg \cdot L^{-1}}$, which were continuously irradiated for 72 h. The experimentally measured concentrations of NH_3 and H_2 were compared with simulation results, as shown in [Figure 3: see original paper]. The simulated values for NH_3 and H_2 agree well with the experimental data, with mean square errors (MSE) of only 1.79$×10^{-8}$ and 5.69$×10^{-8}$, *respectively*, *indicatinghighaccuracyindescribingthemainreactionpathwaysandradiolyticproductgenerat* dissolution during the experiment affecting the system' s acidity.

In summary, the model established in this work can effectively predict the evolution of major radiolytic products during ammonia solution radiolysis and possesses high reliability.

2. Model Application and Strategy Analysis

The objective of primary circuit water chemistry control is to ensure the integrity of the primary circuit pressure boundary, fuel cladding, and fuel design performance, while minimizing the radiation field level outside the core. The core control strategy involves maintaining the coolant pH and dissolved hydrogen concentration within acceptable ranges to ensure the coolant remains in an alkaline and reducing environment. For conventional pressurized water reactors using boron-lithium coordinated control, the pHT value during operation is typically controlled between 6.9 and 7.2, corresponding to OH^- ion concentrations of approximately 10^{-5}-10^{-6} mol $\cdot$ L^{-1}, while dissolved hydrogen concentration is usually maintained within 2-4.5 mg $\cdot$ L^{-1}.

2.1 Simulation of Initial Dispersion Strategy

The initial dispersion ammonia addition strategy refers to injecting a given amount of ammonia solution into the primary circuit when the system is in its initial operating state, allowing rapid dispersion and uniform mixing throughout the coolant system. In the model, this is represented by assigning the same initial ammonia concentration to all mesh nodes to simulate uniform distribution in the system. As core irradiation continues and coolant circulates through the loop, substances in each mesh node are continuously transported and updated, with their concentrations dynamically changing under the influence of reaction and transport processes, exhibiting temporal evolution.

To investigate the effects of different initial ammonia dispersion concentrations, the model was used to calculate the time-dependent concentrations of H_2 and OH^- during radiolysis of coolant containing 0-100 mg $\cdot$ L^{-1} ammonia. The 0 mg $\cdot$ L^{-1} pure water radiolysis data served as the control group, 5 and 10 mg $\cdot$ L^{-1} as low concentration levels, 17, 34, and 51 mg $\cdot$ L^{-1} as medium concentration levels, and 60, 80, and 100 mg $\cdot$ L^{-1} as high concentration groups. To evaluate the chemical effects caused by the slow depletion of ammonia during long-term reactor operation and observe the establishment of concentration steady states, the simulation time was set to 1.6$\times$10^{4}$ s, with results shown in [Figure 4: see original paper].

[Figure 4: see original paper] shows that H_2 concentration quickly reaches chemical steady state with irradiation, and increasing the initial ammonia dispersion concentration significantly enhances H_2 generation rates. At an irradiation time of 1.6$\times$10^{4}$ s, the H_2 concentration in the 0 mg $\cdot$ L^{-1} system is approximately 0.02 mg $\cdot$ L^{-1}, low ammonia concentration groups yield 0.62-1.25 mg $\cdot$ L^{-1}, medium ammonia concentration groups yield 2.12-6.34 mg $\cdot$ L^{-1}, and the high ammonia concentration group (100 mg $\cdot$ L^{-1}) reaches 12.36 mg $\cdot$ L^{-1}. These results indicate that medium ammonia concentration dispersion strategies with initial ammonia concentrations of 17 and 34 mg $\cdot$ L^{-1} can meet the H_2 limit requirements, while low and high ammonia concentration groups cannot satisfy the requirements.

From the perspective of coolant pH, all ammonia dispersion strategies fail to maintain alkaline conditions over extended operation times, as shown in [Figure 4: see original paper]. After initial ammonia addition and irradiation commencement, coolant pH rapidly decreases, falling back from weakly alkaline to neutral state (at 280 °C) and departing from the alkaline control limit range within 1–5 h, eventually approaching neutral levels within 24 h. It should be noted that while short-term pH changes do not directly intensify material corrosion, the loss of alkaline environment may induce more unfavorable corrosion tendencies for metallic materials, leading to potential risks in actual reactor operation.

In summary, the initial dispersion ammonia addition strategy can maintain dissolved hydrogen concentration within the target limit range by controlling ammonia concentration between 17–34 $mg \cdot L^{-1}$, thereby achieving reductive control of the coolant. However, due to the difficulty in maintaining alkaline conditions long-term and the rapid pH decline to neutral after irradiation begins, this strategy cannot simultaneously achieve the two major objectives of coolant chemistry control: meeting dissolved hydrogen concentration standards and maintaining a weakly alkaline environment.

2.2 Simulation of Constant-Rate Injection Strategy

Since the initial dispersion strategy cannot meet primary circuit water chemistry control requirements, this study employs a constant-rate ammonia injection strategy to achieve long-term stable pH control in the coolant system. This involves applying a stable ammonia source term before coolant enters the core to simulate the continuous ammonia injection effect of the charging pump in the primary circuit chemical volume control system. Compared to initial dispersion, this strategy can maintain relatively stable ammonia concentration in the core over longer periods. This section calculates the evolution of dissolved hydrogen and OH^- concentrations under continuous injection rates of 0–16.4 $g \cdot s^{-1}$ and square-wave intermittent injection at 1.64 $g \cdot s^{-1}$, with results shown in [Figure 5: see original paper].

[Figure 5: see original paper] demonstrates that excessive ammonia injection rates can easily cause dissolved hydrogen concentration to rapidly exceed limit values. For example, at an injection rate of 1.64 $g \cdot s^{-1}$, H_2 concentration exceeds 20 $mg \cdot L^{-1}$ at 8000 s, far above the upper limit. Even at lower injection rates (0.164 $g \cdot s^{-1}$), H_2 concentration may exceed limits if irradiation time is sufficiently long. Although stopping ammonia injection can suppress further H_2 accumulation, the concentration does not quickly return to normal range and may continue increasing due to residual ammonia radiolysis in the system. These results indicate that the constant-rate injection strategy carries potential risks of exceeding H_2 concentration limits.

An improved approach based on constant-rate injection was attempted using square-wave intermittent injection: injecting ammonia at 1.64 $g \cdot s^{-1}$ for 1 h, stopping for 1 h, and repeating this cycle. Simulation results show this improved

strategy still carries the risk of dissolved hydrogen concentration exceeding limits within a short time (approximately 2000 s). Although pH can be maintained relatively stable initially, as shown in [Figure 5: see original paper], the cumulative effect of ammonia in the system with increasing cycles may cause pH to continuously rise, bringing potential overrun risks.

Additionally, as shown in [Figure 5: see original paper], square-wave intermittent injection demonstrates stronger pH controllability compared to continuous injection. The latter only shows short-term pH controllability at injection rates of 0.164 and 1.64 $g \cdot s^{-1}$, while both too low (0.0164 $g \cdot s^{-1}$) and too high (16.4 $g \cdot s^{-1}$) rates cannot effectively regulate coolant pH.

In summary, square-wave intermittent injection has certain effectiveness in pH control, allowing system pH to fluctuate slightly within the target range by adjusting injection frequency and concentration. However, regardless of the control method, H_2 concentration risks exceeding limits. Therefore, this ammonia addition strategy still cannot fully meet established water chemistry control objectives. To address the continuous H_2 concentration rise under this strategy, hydrogen removal measures can be further introduced to simulate their effectiveness in controlling H_2 concentration within limits, thereby achieving comprehensive optimization and regulation of water chemistry status.

2.3 Simulation of Hydrogen-Removal-Optimized Constant-Rate Injection Strategy

Since both initial dispersion and constant-rate injection strategies have limitations, this section introduces a hydrogen removal mechanism based on the constant-rate injection strategy. Corresponding hydrogen removal rates were matched for injection rates of 0.164 and 1.64 $g \cdot s^{-1}$ (for 0.164 $g \cdot s^{-1}$ NH_3 injection: -0.00148 $g \cdot s^{-1}$; for 1.64 $g \cdot s^{-1}$ NH_3 injection: -0.0140, -0.0150, -0.0160 $g \cdot s^{-1}$). The hydrogen removal process was simulated by setting a negative H_2 source term at the top of the meshed core region. Since the initial H_2 concentration in the coolant system is zero, to avoid calculation errors, this negative source term was set to activate after a specific time (1200 s, 10800 s). Simulation results are shown in [Figure 6: see original paper].

[Figure 6: see original paper] shows that excessively high hydrogen removal rates can cause dissolved hydrogen concentration to drop rapidly, potentially leading to undesirable consequences such as oxygen removal failure. For example, under 1.64 $g \cdot s^{-1}$ ammonia injection, when removal rates are 0.016 and 0.015 $g \cdot s^{-1}$, H_2 concentration reaches the lower limit at approximately 2000 and 7200 s, respectively. In contrast, appropriately reducing the removal rate (e.g., to 0.014 $g \cdot s^{-1}$) can maintain H_2 concentration within a reasonable range. Similarly, under lower ammonia injection rates (0.164 $g \cdot s^{-1}$), further reducing the removal rate (-0.00148 $g \cdot s^{-1}$) also helps achieve H_2 concentration compliance.

pH variations are shown in [Figure 6: see original paper]. In constant-rate injection strategies without hydrogen removal, pH continuously increases with

irradiation time, eventually exceeding the upper pH limit. When ammonia injection rate is 1.64 $g \cdot s^{-1}$ with hydrogen removal rates of 0.016 and 0.015 $g \cdot s^{-1}$, pH can only be maintained for 2 and 4 h, respectively. Further analysis of two ammonia addition strategies meeting H_2 concentration requirements reveals that both can satisfy pH limit ranges, with the general pattern that lower ammonia injection rates result in lower H_2 pressure and lower stable pH and dissolved hydrogen levels.

In summary, through hydrogen removal optimization studies of the constant-rate injection strategy, two reasonable control schemes were obtained: (1) constant-rate ammonia injection at 1.64 $g \cdot s^{-1}$, with hydrogen removal device activated 1200 s after injection begins at a controlled removal rate of 0.014 $g \cdot s^{-1}$, corresponding to pHT = 6.9; and (2) constant-rate ammonia injection at 0.164 $g \cdot s^{-1}$, with hydrogen removal device activated 10800 s after injection begins at a removal rate of 0.0014 $g \cdot s^{-1}$, corresponding to pHT = 6.5. The dissolved hydrogen concentration for both schemes is approximately 2.7–3.1 $mg \cdot L^{-1}$, equivalent to 30–35 $mL \cdot kg^{-1}$ (STP). However, considering that higher coolant pH can effectively reduce corrosion of metallic materials in the primary circuit coolant system, the first scheme is superior.

Conclusion

This study established a water chemistry control model for ammonia-containing coolant in the primary circuit and investigated the radiolysis behavior of ammonia-containing coolant using the KLT-40S reactor as an example. The effects of initial dispersion ammonia addition, constant-rate injection, and hydrogen-removal-optimized constant-rate injection strategies on dissolved hydrogen and pH were compared. The results demonstrate that: (1) the initial dispersion strategy can achieve dissolved hydrogen concentration compliance and reductive control of coolant but cannot effectively regulate coolant pH; (2) the constant-rate injection strategy has shortcomings in dissolved hydrogen concentration control, and while square-wave intermittent injection can achieve pH regulation, it cannot maintain dissolved hydrogen concentration within limits for extended periods; (3) the hydrogen-removal-optimized constant-rate injection strategy can simultaneously achieve coolant pH regulation and dissolved hydrogen concentration control by matching appropriate hydrogen removal rates; and (4) this work provides a reasonable water chemistry control scheme with an ammonia injection rate of 1.64 $g \cdot s^{-1}$, hydrogen removal initiated 1200 s after injection begins at a rate of 0.014 $g \cdot s^{-1}$.

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Note: Figure translations are in progress. See original paper for figures.