Corrosion of structural materials and electrochemistry in high temperature water of nuclear power systems

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Abstract: The latest experiences with corrosion in the cooling systems of nuclear power plants are reviewed. High temperature cooling water causes corrosion of structural materials, which often leads to adverse effects in the plants, e.g., generating defects in materials of major components and fuel claddings, increasing shutdown radiation and increasing the volume of radwaste sources. Corrosion behaviors are much affected by water qualities and differ according to the values of water qualities and the materials themselves. In order to establish reliable operation, each plant requires its own unique optimal water chemistry control based on careful consideration of its system, materials and operational history. Electrochemistry is one of key issues that determine corrosion related problems but it is not the only issue. Most phenomena for corrosion related problems, e.g., flow-accelerated corrosion (FAC), intergranular stress corrosion cracking (IGSCC), primary water stress corrosion cracking (PWSCC) and thinning of fuel cladding materials, can be understood based on an electrochemical index, e.g., electrochemical corrosion potential (ECP), conductivities and pH. The most important electrochemical index, ECP, can be measured at elevated temperature and applied to in situ sensors of corrosion conditions to detect anomalous conditions of structural materials at their very early stages. In the paper, theoretical models based on electrochemistry to estimate wall thinning rate of carbon steel piping due to flow-accelerated corrosion and corrosive conditions determining IGSCC crack initiation and growth rate are introduced.

Keyword: nuclear power plant; corrosion; electrochemistry; electrochemical corrosion potential

1 Introduction

More than 450 nuclear reactor units are operated worldwide, producing CO_2 emission-free electricity and the total output for all units is 380GWe^[1]. Among them, 60% are pressurized water reactors (PWRs) and 20% are boiling water reactors (BWRs). In order to maintain nuclear energy as a major energy source, much higher safety and reliability are required in nuclear power plant (NPP) operation. One of the key technologies to maintain reliability against failure of the structural materials of NPPs is water chemistry control of the cooling water.

In NPPs, one of the most important roles of water is as an energy transfer medium. There, water is heated in the reactor core, where it is changes to steam which drives the turbines to produce electricity. Cooling water in fossil power plants (FPPs) plays the same role. Water chemistry control in NPPs is the same as that in FPPs and experiences with water chemistry control in the former are based on those in

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the latter. However, another important role of water is acting as a neutron moderation medium, which is not shared with FPPs. The original energy of the neutrons generated by fission of 235 U is too high to allow them to be captured by 235 U and then to maintain nuclear chain reactions. The neutron energy should be reduced to about a millionth of the original energy by elastic collisions of neutrons with hydrogen atoms in water. With light water as coolant in NPPs, the resulting high temperature water leads corrosion of structural materials, which leads to adverse effects in the plants, *e.g.*, defect generation in materials of major components and fuel claddings, increased shutdown radiation and increased volume of radwaste ^{[2]-[4]}

Continuous and collaborative efforts of plant manufacturers and plant operator utilities have been focused on satisfying these requirements for NPPs. One of the effective procedures to satisfy them is optimal water chemistry control, for which, a trio of requirements for water chemistry, *i. e.*,

a) reliability in reactor structures and fuels,

b) lower occupational exposure, and

c) fewer radwaste sources, should be simultaneously satisfied ^{[2], [5]}.

In this paper, material problems related to nuclear power plants are reviewed and interactions between materials and cooling water are discussed from the water chemistry side.

2 Optimal water chemistry

2.1 Major problems related to structural materials

Major problems related to structural materials in NPPs are listed in **Table 1**^[6]. Intergranular stress corrosion cracking (IGSCC) is one of the most

frequently reported material problems for BWR plants but no serious accidents related to it have ever been reported. Defects of steam generator tubing often interrupt PWR plant operation but they have never caused a serious accident.

Major accidents and incidents at nuclear facilities are listed in **Table 2**^[6]. Flow accelerated corrosion (FAC) has caused two serious accidents with PWRs ^{[7], [8]}. FAC is not limited to PWRs, but has also been reported in FPPs and BWRs ^{[9], [6]}. FAC in two-phase flow has often been reported in heater drain tubing and crossover piping of PWRs and BWRs^{[10], [6]}

problem	reactor type	troubled location	countermeasures
FAC	PWR	feed water piping	material exchange ①, water chemistry improvement ②
	BWR	feed water piping	2
		heater drain piping	0
SCC	BWR	primary piping	 , 2, stress improvement
PWSCC	PWR	core internals	1, 2
Fuel cladding corrosion	PWR, BWR	fuel	2
SG tubing defects	PWR	steam generator (SG)	1, 2

Table	1	Major	problems	for	structural	materials	in NPPs
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Plant	(reactor type)	Date	Causes	Environmental effects
Three Mile Island	(PWR)	Mar. 1979	LOCA	<1mSv
Chernobyl	(LGR)	Apr. 1986	RIA	31 people died, 16 k person-Sv
Surry-2	(PWR)	Dec. 1986	FAC*	4 people died
Fukushima Daini-3	(BWR)	Jan. 1989	vibration	none
Mihama-2	(PWR)	Feb. 1991	CF	none
Monju	(LMFBR)	Dec. 1995	parts defect (Na leakage)	none
JCO	(conversion facility)	Sep. 1999	critical accident	2 people died,
	10 C.	2		130 people received radiation dose
Hamaoka-1	(BWR)	Nov. 2001	H ₂ explosion	none
Mihama-3	(PWR)	Aug. 2004	FAC*	5 people died
Fukushima	(BWR)	Mar. 2011	earthquake	radioactivity release: 600 PBq
Daiichi 1-4			+ tsunami	evacuee: 160,000
*: related to material LOCA		loss of coola	nt accident RIA: reactiv	ity initiated accident
	FAC: fl	ow assisted o	corrosion CF: corrosio	n fatione





b) BWR

Fig.1 Major subjects related to materials in cooling systems of nuclear power plants. Nuclear Safety and Simulation, Vol. 5, Number 1, March 2014

2.2 Major features of water chemistry in NPPs

Distributions of materials in contact with the coolant differ between BWR and PWR systems. Material atlases are shown in Figure 1^[4]. In each system, uniformly controlled cooling water is in contact with different materials, which complicates corrosion problems. Corrosion behaviors are much affected by water qualities and differ according to the values of water qualities and the materials themselves.

BWR and PWR systems differ in their heat transporting systems. Materials in contact with the coolant also differ in both systems. Wetted surfaces of structural materials in the BWR primary cooling system are equally divided among three materials, *i.e.*, zirconium alloy for fuel cladding, stainless steel for core and peripheral structures and carbon steel for condensate, feed water and turbine structures, while about three quarters of the surfaces in the PWR primary cooling system are nickel base alloy for steam generator tubing and about a quarter are zirconium alloy for fuel cladding so that the amount of stainless steel surfaces for vessel components and primary piping is minor (Fig. 2)^[11]. Zirconium alloy, nickel base alloy and stainless steel are more expensive than carbon steel, but structural materials must have sufficient integrity against radioactive species in the fuel and in the primary cooling water which necessitate uses of the expensive materials.

Different combinations of materials and different water chemistry controls for both BWRs and PWRs cause different interactions between materials and cooling water and then, consequently different material problems. The interactions of materials and cooling water in the BWR primary cooling system, PWR primary cooling system and PWR secondary cooling system have been detailed in reference ^[12].

2.3 Optimal water chemistry

In order to control the adverse effects, *e.g.*, generation of defects in materials of major components and fuel claddings, increased shutdown radiation and increased volume of radwaste, it is essential to understand corrosion behaviors of structural materials and then to control them in both systems. In order to minimize the adverse effects, optimal water chemistry control has been proposed as

shown in Fig. 3^[6].



Fig.3 Optimal water chemistry control.

2.4 Major factors to determine material problems

Corrosion of materials is determined by the combined effects of material factors and water chemistry factors. Corrosion in aqueous systems is understood as electrochemistry phenomena. At metal surfaces, metallic ions are released from the base metal and at the same time dissolved metallic ions are oxidized, precipitating as an oxide film on the surface. Metal oxide also develops on the interface between the oxide film and base metal by direct oxidation. Development of the oxide film is one of the consequences of electrochemistry reactions, and its progression may control the reactions. The properties of the oxide film on the metal surface are much affected by material properties and the effects of complicate corrosion behaviors. In order to understand the problems related to corrosion in NPPs, understanding and analyzing the electrochemical reactions of structural materials and the contribution of oxide film to the reactions are essential. A schematic diagram for the interaction of structural

materials and cooling water are shown in Fig. 4. Oxide film on the surfaces of the materials is a consequence of the interaction and at the same time a reason why the interaction should be controlled.



3 Theoretical approaches to quantify materials and water interactions

3.1 Electrochemistry

Electrochemistry has been developed as a branch of chemistry studying chemical reactions which take place in a solution at the interface of an electron conductor (the electrode) and an ionic conductor (the electrolyte). These reactions involve electron transfer between the electrode and the electrolyte or species in solution. The electrochemistry has been often applied to explain corrosion phenomena but it has seldom been applied for quantitative prediction of corrosion problems. In this paper, two examples for application of electrochemistry to prediction of corrosion rate due to FAC and electrochemical corrosion potential under radiation irradiation are introduced.

3.2 Prediction of ECP and corrosion rate

Under the equilibrium conditions, anodic and cathodic current densities are balanced without any net current between the electrode and the electrolyte (Fig. 5) ^{[3], [13]}. From the balance diagram electrochemical corrosion potential (ECP) and then the corrosion rate can be obtained. The anodic current density is much affected by the protective oxide film developed on the metal surface, which contributes ECP increase and corrosion rate decrease.



Fig.5 Anodic and cathodic current densities.

3.3 Prediction of oxide film thickness

Thickness of oxide film developed on the surface can be calculated according to the mass balance on dissolution of metallic ion, its oxidation and film forming (Fig. 6)^{[3], [14]}.



Fig.6 Modified double oxide layer model.

3.4 Coupled electrochemistry and oxide layer growth model

ECP and corrosion rate can be calculated by coupling both models, *e.g.*, the electrochemistry model to determine corrosion rate and ECP based on oxide film thickness and their properties, and the oxide film growth model to determine oxide film thickness and properties based on corrosion current (**Table** 3). The details of the numerical expression the model are shown in the references ^{[3], [13]}.

As an example of the coupled model, the hysteresis of the anodic polarization is shown in Figure 7^[3].

Corrosion behaviors of carbon steel and stainless are compared in **Table 4**^[3].

Sub-	electrochemistry	oxide layer growth
model	(static model)	(dynamic model)
Input	temperature, [O ₂] , pH,	temperature
	mass transfer coefficient,	i _a , i _c
	oxide film thickness,	ECP
	oxide properties	Coupling calculation
Output	anodic/cathodic	oxide film thickness
	current densities, i _n , i _e	properties
	ECP	(Fe ₂ O ₃ /Fe ₃ O ₄ ratio)

Table 3 Coupled electrochemistry/oxide layer growth model





b) Mass transfer coefficient dependence

Fig.7 Calculated anodic polarization responses.

Table 4 Major corrosion properties

Materials	Carbon steel	Stainless steel		
Corrosion rate	high	medium		
Oxide film	magnetite/hematite	Cr rich nickel ferrite		
Application	piping of secondary	piping of primary		
	system	system		
Problems	FAC	IGSCC		
		radioactivity accumulation		
Effects of	strong	medium		
electrochemistry				

In the next chapter, corrosion of carbon steel (FAC corrosion rate) and corrosion of steel (ECP in BWR primary cooling system) are discussed related to the coupled model are demonstrated.

4 Carbon steel: FAC

4.1 Phenomena of FAC

FAC is divided into two processes: the corrosion (chemical) process and the flow dynamics (physical) process ^{[9], [15]-[18]}. The former is the essential process to cause FAC and the latter is the accelerating process to enhance FAC occurrence. Metallic ions, mainly ferrous ions (Fe^{2+}), are released into the water at the boundary layer, where their concentration becomes supersaturated; some of the Fe²⁺ will form oxide particles and they deposit on the metal surface to become a magnetite oxide layer. This oxide layer plays an important role in preventing more release of Fe^{2+} (corrosion reaction). The thickness of the boundary layer is much affected by fluid dynamics. For these processes, oxygen concentration $([O_2])$ in the boundary layer also plays an important role for oxidizing the magnetite to the hematite which contributes to much higher corrosion resistance.

4.2 FAC evaluation procedures

Six calculation steps were prepared for predicting FAC occurrence and evaluating wall thinning rate (Fig. 8) ^{[19]-[22]}. Flow pattern and temperature in each elemental volume along the flow path are obtained with a (one dimensional 1D) computational fluid dynamics (CFD) code and then corrosive conditions, e.g., $[O_2]$ and electrochemical corrosion potential (ECP), along the flow path are calculated with the O₂-N₂H₄ reaction code ^[19]. Precise flow patterns around the structure surface are calculated with a 3D CFD code and then distributions of mass transfer coefficients at the surface are obtained ^[20]. The high FAC risk zone is evaluated by coupling major FAC parameters obtained by Steps 1 through 3^[21]. At the indicated high FAC risk zone, wall thinning rates are calculated with the coupled model of static electrochemical analysis and dynamic double oxide layer analysis (Table 5).



Fig.8 Evaluation inspection steps for wall thinning due to FAC.

Table 5 Code system for FAC evaluation

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Calculation targets	Flow pattern	[O ₂], [Fe ²⁺]	Wall thinning rate and ECP (anodic/cathodic current density) (oxide film formation)
Input	Reactor parameters: geometries, heat flux (Q), temperature (T)	Reactor parameters: T, flow velocity (v), surface/volume rate, mixing rate	[O ₂], T, pH, k _m , oxide film thickness, oxide properties	i _{corr} , ECP
Computer programs	1D CFD 2-3D k-ε CFD 3D LES	N ₂ H ₄ -O ₂ reaction code	Static model (Electrochemistry)	Dynamic model (Oxide layer growth)
Output	T, v distributions along the flow path	[O ₂] and [Fe ²⁺] distributions	i _{corr} , ECP wall thinning rate	oxide film thickness oxide properties (Fe ₂ O ₂ /Fe ₂ O ₄ ratio)

As a final evaluation, residual lifetime of the pipes and applicability of countermeasures against FAC are evaluated in Step 6. The details of calculation procedures for Steps 3 and 5 were shown previously. In order to narrow down the number of inspection zones, evaluated prediction results are applied. Precise measurements at the restricted area are expected to obtain reliable data sets. And evaluated measured wall thickness data can be fed back for improvement of the prediction code. Such coupling of the estimation and inspection procedures should allow effective and reliable preparation to be made against FAC occurrence and propagation.

4.3 Wall thinning evaluation

Wall thinning rate can be calculated as a function of temperature, pH and mass transfer coefficient at the surface. As an example of calculated results, wall thinning rates are shown as a function of mass transfer coefficient for neutral and high pH conditions in Fig. 9^{[21], [22]}.



Fig.9 Calculated wall thinning rate as a function of mass transfer coefficient.

From comparison of the calculated wall thinning rates due to FAC with hundreds of measured results for secondary piping of an actual PWR plant, it was confirmed that the calculated wall thinning rates agreed with the measured ones within a factor of 2. Most of the calculated/measured ratios were in the factor of two regions, except those for the very low thinning rate were still outside the region and had values larger than a factor of two. One of the causes of the discrepancies between the measured and calculated results was the very low thinning rate with large errors by ultrasonic testing (UT). Certainly, calculation procedures should be improved, and measurement accuracies should also be improved. Mitigating the discrepancy between the calculated and measured values at low measured thinning rate regions is a future subject for consideration, and the root reasons for underestimation should be carefully investigated to establish the proposed six-step process as a standard code for FAC evaluation. The reliability of piping is determined by residual thickness. So it is important to evaluate residual thickness with high accuracy. The accuracy of the evaluation model for pipe wall thickness both for bend piping and T junctions was confirmed to be less than 20 %, as shown in Fig. 10^[22].



Fig.10 Calculated results based on 3D FAC code: residual thickness.

5 Stainless steel: IGSCC

One of the most serious problems with structural materials in BWRs is IGSCC of stainless steel ^[30]. Under severe neutron and gamma ray irradiation in

the reactor core region, corrosive conditions in the BWR primary cooling system are determined by corrosive radiolytic species. At the same time, structural materials themselves are irradiated by neutrons and their physical and chemical properties are changed during plant operation.

5.1 Corrosive conditions in BWR primary coolant

The primary scheme for water radiolysis in BWR primary coolant is shown in Figure 11 ^[23]. Water radiolysis in the reactor core and its peripheral regions generates radiolytic species, *e.g.*, O_2 and H_2O_2 (oxidants), which cause highly corrosive conditions in BWR primary cooling water ^[24].



Fig.11 Theoretical determination of corrosive conditions of BWRs based on water radiolysis model.

Unfortunately, in the BWR cooling system, hydrogen injection for fully oxidant suppression was not applicable due to hydrogen loss through the main steam line, hydrazine injection was also not applicable due to nitrogen compound effects on fuel cladding materials and the need for highly purified neutral water to be used to minimize corrosive conditions ^[24]. As a result of release of injected hydrogen through the main steam line, suppression of water radiolysis in the core was limited in only the non-boiling region and then oxidant concentrations in the main steam line were not sufficiently reduced. However, oxygen concentration ([O₂]) in the recirculation water was sufficiently reduced (Fig. 12).



Fig.12 Effects of H₂ injection in BWR plants.





The difference was from oxidant and hydrogen recombination in the downcomer region under weak irradiation conditions ^[25]. The experience with reduced oxidant concentration in the recirculation water has led to development of hydrogen water chemistry (HWC, with hydrogen injection) to moderate corrosive conditions in the recirculation line and the lower reactor plenum.

With increasing concentration of hydrogen injected in the feed water, oxygen concentration measured at the sampling point decreased rapidly and, at the same time, the main steam line dose rate (MSDR) increased gradually (Fig. 12 a)) ^{[25], [26]}. The optimal hydrogen injection amount was determined to suppress [O₂] without serious increase of MSDR. Several indexes for corrosive conditions have been proposed to evaluate the effects of HWC in BWR plants, one of the most common is electrochemical corrosion potential (ECP) ^{[27], [28]}. ECP is determined by a combination of factors; these are concentrations of oxidants, *e.g.*, O₂, H₂O₂ and other corrosive radiolytic species, and specimen surface conditions. Measured ECP for HWC is also shown in Fig. 12 a). From the viewpoint of IGSCC mitigation, the threshold ECP has been proposed as -230 mV-SHE. However, in the optimal hydrogen concentration determined by $[O_2]$, the ECP was still too high to mitigate IGSCC.

5.2 Theoretical ECP evaluation

Non-linear rate equations (**Table 6**) were calculated to obtain the concentration distributions of radiolytic species throughout the primary coolant ^[27]. Major constants, *i.e.*, G-vales and rate constants, for water radiolysis calculation are shown in the reference ^[26].

ECP calculation based on mixed potential model follows water radiolysis calculation as shown in Figure 13^{[13], [14]}.

Concentrations of oxidants ($[O_2]$ and $[H_2O_2]$) change along the recirculation flow path so that the corrosive conditions differ between locations in BWR primary cooling systems ^[26]. The only point to determine oxidant concentrations along the recirculation flow is at the end of the sampling line installed in the recirculation line, where water is cooled down and depressurized for measurement by the probe type oxygen detector. In order to determine the oxidant distribution in the primary system, theoretical models have been prepared to calculate the concentration distribution.



Fig.13 ECP evaluation procedures.

The calculated results are shown in Figure 14 for normal water chemistry (NWC: without hydrogen injection) and HWC.



Fig.14 Maps of distribution of $[O_2]_{eff}$ in RPV (Effects of H_2 injection on suppression of $[O_2]$).

NWC: normal water chemistry (without hydrogen injection) HWC: hydrogen water chemistry (with hydrogen injection) The gap between the measured ECP and measured oxygen concentration shown in Figure 12 b) ^[26] is explained by the contribution of H_2O_2 to ECP especially at high H_2 injection rate ^[28]. The concentrations of H_2O_2 and O_2 were calculated based on the coupled model of electrochemistry and oxide film growth model and for both NWC and HWC conditions, H_2O_2 concentration was high enough to determine ECP by only H_2O_2 ^[29].



Fig.15 Crack growth rate as a function of ECP.

5.3 IGSCC crack growth rate

A schematic diagram for IGSCC crack growth rate is shown in Figure 15^[30]. As a result of high oxidant concentration, ECP at the outer surface of structures is high, while it is low enough in the crack tip due to the decreased concentration of oxidants, which are spent as oxidation reactions along the crack inner surface. A low local ECP at the crack tip leads to the potential difference between the tip and the surface, which results in transfer of metallic ions from the tip to bulk water and transfer of anions from bulk to the tip. Accumulation of anions, *e.g.*, $SO_4^{2^-}$, results in lower pH around the tip, which accelerates dissolution of the crack tip.

It is important to reduce ECP for mitigating IGSCC occurrence and crack growth due to decreasing $[O_2]$.

6 Future Subjects

6.1 Standardization of theoretical approaches

Theoretical tools for evaluating FAC and IGSCC are quite useful to predict future trouble and early preparation for suitable countermeasures to prevent future trouble. It is important for the tools to be recognized as standard and authorized procedures. For this, verification and validation (V&V) evaluation has been applied ^[31].

The verification and validation (V&V) processes for the FAC simulation code and the corrosive condition calculation code were done in conformity with the ASME "Guide for Verification and Validation in Computational Solid Mechanics."

The definitions of V&V are as follows:

- code verification: addressing errors in the software
 calculation verification: estimating numerical errors due to under resolved discrete representations of the mathematical model
- 3)validation: assessing the degree to which the computational model is an accurate representation of the physics being modeled, based on comparison between numerical simulations and relevant experimental data (predictive capability of the model).

V&V evaluation for FAC code has been done and the results have been published ^[22]. But its standardization has not been established yet. V&V evaluation for coupled water radiolysis and ECP codes are on progress.

As a consequence of the coupled analysis, the final corrosive condition by applying the ECP as the parameter of corrosive conditions can be compared with the code results. An example of the relationship between the measured ECP at the plant and ECP calculated for the same plant is shown in Fig. 16 with the discrepancy of ± 50 mV.

The approaches towards standardization have been prepared ^[32].



Fig.16 Relationship between ECP measured at the plant and the calculated ECP.

7 Conclusions

The conclusions are summarized as follows.

- 1) Optimal water chemistry control has been established to satisfy multi-problems related to interaction of materials and water is introduced.
- 2) Theoretical approaches as well as empirical ones have been established to quantify the interaction of materials and water and to establish suitable countermeasures for those problems.
- 3) Electrochemistry procedures have been successfully applied to determine corrosion related problems.
- 4) As examples of application of theoretical electrochemistry procedures, a prediction model for FAC and prediction models for water radiolysis are introduced.
- 5) As future subjects of the theoretical models related to corrosion problems, standardization of the codes should be established based on V&V evaluation procedures

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Nomenclatures

- C_1 : concentration of radiolytic species (mol/m³)
- G_i^{γ}, G_i^{n} : g-value (mol/J) [superscripts: γ, γ rays; n, neutrons]
- k_{mn}^{i} : rate constant between species m and n $(m^{3}/mol/s)$
- k_e: electrochemical reaction constant (m/s) (superscripts: a, anodic; c, cathodic; H, H₂O₂ oxidation)

 P^{γ}, P^{n} : energy deposition rate $(J/m^{3}/s)$

- q_l : generation and appearance term by chemical reactions in an elemental volume $(mol/m^3/s)$
- t: time (s)
- U: flow rate (m^3/s)
- V_f: void fraction rate (-)
- $\epsilon *_{1,} \epsilon_{1}$: mass transfer coefficient from gas to liquid and vice versa (1/s)

Abbreviations

- 1D: one dimensional
- AGR: advanced gas cooled reactor
- BWR: boiling water reactor
- CF: corrosion fatigue
- CFD: computational fluid dynamics
- ECP: electrochemical corrosion potential
- FAC: flow accelerated corrosion
- FPP: fossil power plant
- IGSCC: inter-granular stress corrosion cracking
- LOCA: loss of coolant accident
- MSDR: main steam line dose rate
- NPP: nuclear power plants
- PWR: pressurized water reactor
- PWSCC: primary water stress corrosion cracking
- RIA: reactivity initiated accident
- VVER: Novo Voronezh-type PWR

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