Corrosion Properties of Candidate Materials in Supercritical Water Oxidation Process

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Abstract:
Corrosion of constructional materials in supercritical water oxidation (SCWO) is one of the main obstacles to commercializing the SCWO process. Statistical analysis of corrosion research results indicate nickel-based alloys are the most widely applied materials in SCWO system among the candidate metals. The present paper reviews the corrosion characteristics of iron-base alloy, nickel-based alloy, titanium-base alloy, ceramics, niobium, tantalum, thallium and gold under the condition of SCWO. The selection scheme of materials is presented in SCWO system flow diagram in detail. The effects of alloying elements on the corrosion behavior of materials in the water environment with high temperature and high pressure water are summarized in this paper. Moreover, some further researches for material corrosion in SCWO process are also proposed. All work was contributed to selecting the appropriate constructional materials and reducing the corrosion damage in SCWO system.

Keywords: Corrosion; Supercritical water oxidation; Material; Alloy element

Introduction

Supercritical water (SCW) is the water above its thermodynamic critical point (374 °C, 22.1 MPa) has attracted increasing attention in supercritical water gasification (SCWG) and supercritical water oxidation (SCWO) in the past three decades (1-5). SCWG and SCWO have been proved to be a high effective way to resource utilization and harmless for organic matter, respectively (6-8). SCWO provides a potential alternative technology for incineration technology to treat high concentration organic wastewater without secondary pollution (9). The big advantage of SCW that presents in gasification and oxidation is mainly dependent on the changed physic-chemical properties of water at the elevated temperatures and pressures, and SCW has the properties of excellent heat transfer and mass transfer. The physicochemical properties of normal state water, subcritical water, SCW and superheated steam are listed in Table 1. The main physicochemical properties of SCW are all between normal state water and superheated steam, and SCW behaves like a nonaqueous solvent (10-12).

Many nonpolar organic substances and gases are highly soluble in SCW because of the disaappearance of phase boundaries (1). The reaction condition of SCWO usually contained the temperature range of 400 °C~600 °C and pressure range of 24 MPa ~ 28 MPa (2, 4, 13). The solubility of ionic compound will decrease sharply when the wastewater is heated to supercritical condition. This property of SCW is generally used to separate salts from wastewater so that the salts will be recycled or buried. In addition, supercritical water oxidation process could realize energetically self-sufficient operation depending on the organic with high concentration in the wastewater (6, 14). However, there is serious corrosion for materials under the harsh working conditions of high temperature, high pressure and strong corrosive anions. The corrosion problem is the main obstacle to the commercialization and large-scale application of SCWO (12, 13, 15-21).

A volume of work has been carried out on solving the material corrosion problem in SCWO systems, especially in condition of hazardous waste oxidation. In order to find the appropriate material, the corrosion behaviors of candidate materials under different SCWO conditions were investigated. Ni-based alloys, iron-based alloys, ceramics, titanium based alloys, zirconium based alloys, noble metals, tantalum and niobium were selected for corrosion tests as the candidate materials for constructing SCWO system. Many corrosion control approaches were also developed to decrease the high corrosion rates for extending the service life of SCWO system (22, 23). In recent years, lots of studies about SCWO corrosion focused on the surface chemical and the migration of

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Table 1. Physicochemical properties of water as a function of temperature and pressure (1).

Parameter	Normal state water	Subcritical water	Supercritical water	Superheated steam
T (°C)	25	250	400	400
p (MPa)	0.1	5	25	0.1
ρ(g/cm^3)	0.997	0.8	0.17	0.0003
ε	78.5	27.1	5.9	1.0
p*K*w	14.0	11.2	19.4	-
*C*p(kJ/kg·K)	4.22	4.86	13	2.1
η(mPa·s)	0.89	0.11	0.03	0.02
λ(mW/mK)	608	620	160	55

elements, which were helpful to understand the corrosion mechanism (24-27). The alloy element and element content usually played an important role in the corrosion behaviors of materials under SCWO conditions. The purpose of this paper is to review and summarize the corrosion studies of candidate materials and it would also be served as a leading selecting materials for SCWO system.

The Materials for Corrosion Test

Corrosion tests of typical materials in SCWO were stainless steels, nickel based alloys, titanium, ceramic, zirconium, niobium, tantalum and gold. In consideration of economy and corrosion resistance, stainless steels and nickel-based alloys have drawn more research attention than other materials.

As Figure 1(a) shows, occurrence numbers of different test alloys were presented in the statistical analysis of 120 articles from 1994 to 2014 in SCWO corrosion research. The nickel-based alloys hold the largest proportion among all tested materials in previous corrosion researches, indicating that the nickel based alloys are excellent candidate materials for fabricating SCWO system. As shown in Figure 1(b), Inconel 625 make up the highest proportion of the occurrence numbers of nickel based alloys (accounted for 34.4%), and followed by Hastelloy C-276 (accounted for 23.6%). Both Inconel 625 and Hastelloy C-276 own good corrosion resistance and mechanical strength under the hashing operated condition, so the application of them in experimental and commercial plant is very common. In oxidizing supercritical water, due to its high chromium content, Inconel 625 showed better corrosion resistance than Hastelloy C-276. However, Hastelloy C-276 showed a lower corrosion rate than Inconel 625 in supercritical water containing chloride because of the high molybdenum content (28-30). In addition, the mechanical strength of Hastelloy C-276 is weaker than that of Inconel 625 on exposure to high temperature condition (T＞550 °C).

Corrosion Results of Candidate Materials

Iron Based Alloy

316 SS, 316L SS, 304 SS and 304L SS were four main iron based alloys which were selected for constructing SCWO systems by many researchers. Chromium and TiO_2 were used as coatings on 316 SS to strenghthen the resistance to corrosion under the SCWO condition so that the corrosion rate was decreased by these means (31, 32). The outer and inner layers of the tested iron based alloys would rich in Fe_2O_3/Fe_3O_4 and Cr/Ni/Mo oxide respectively in either SCWO or SCW without chloride (33-37). The depletion of Fe was also observed on outer layer when solution contained the chloride. Iron based alloys showed a weaker corrosion resistance than nickel based alloys in previous SCWO corrosion studies, especially in the chlorinated solution (38). The 316L alloys showed the highest corrosion rate about 50 mmyr^{-1} among all alloys, indicating austenitic stainless steel was not suitable for chlorinated feed in SCWO (29). When the test of decomposing organic matter was conducted, it was always found that the carbon-contaminated covered the surface of alloys due to the incomplete degradation of organic matter (26, 28). It is known that the high temperature subcritical water has higher corrosive and solubility than supercritical water. Alloys always get a higher corrosion rate at subcritical condition than supercritical condition and the higher destruction efficiency of organic matter at subcritical condition because of the more dissolved catalytic alloying elements by severe corrosion (16, 29, 39-48). The high pressure 60 MPa could cause intergranular corrosion and higher corrosion rate than 25 MPa by more than two orders of magnitude, indicating the importance of density of water on corrosion behavior of alloys (49). With improving the oxidant concentration in the supercritical water, and adding H_2SO_4 into it, the corrosion rate of 316L SS immersion coupons could reach 39 mmyr^{-1} and the fluid mechanical effects could cause high rate of 316 L tubing corrosion (50). The Ce^{3+} could form a Ce

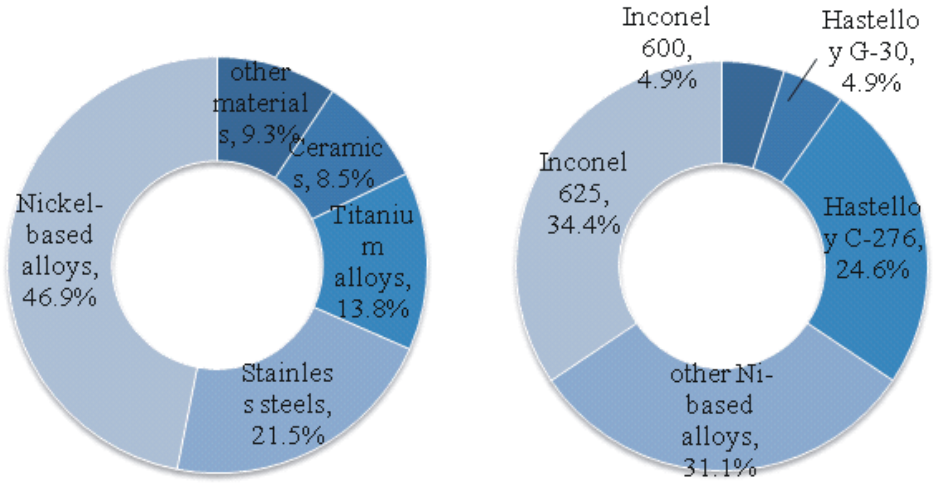

Figure 1. The percentage of alloys occurrence numbers in 120 articles from 1994 to 2014, (a) different kinds of alloys and (b) different Ni-based alloys.

oxide layer to prevent the alloy from further corrosion (51). Depending on the above results, iron based alloys can endure the corrosion conditions of supercritical water with or without oxidant (49). However, iron based alloys displayed a high corrosion rate under both subcritical and supercritical water when the feed contained aggressive feeds (Cl^-, SO_4^- and PO_4^{3-}).

Nickel Based Alloy

The nickel-based alloys are generally considered important for severe service application, however, they cannot withstand some aggressive feeds in SCW. The studies for corrosion behaviors of nickel based alloys were conducted among Inconel 625, Hastelloy C-276, Hastelloy G-30, Inconel 690 and other nickel based alloys under supercritical or subcritical conditions. The increase of chromium concentrations resulted in increased corrosion resistance of nickel based alloys under SCWO condition, indicating the protective oxide Cr_2O_3 formed on the alloys (40, 52-55). The increase of the chromium content also accelerated trans passive dissolution at 350 °C and 500 °C, but was low at temperatures of 400~450 °C (54). Stress cracking corrosion (SCC) could be observed at a low pH value, while it is not so easy to be observed in the neutral solution (16). In the corrosion test of the nickel based alloys exposed to an aqueous solution containing chloride and oxygen, pitting corrosion was observed at temperatures above 130 °C to 215 °C and the trans passive dissolution mainly occured at the critical temperature, however, trans passive dissolution corrosion rate of Inconel 625 decreased drastically in neutral, alkaline and deaerated hydrochloric acid (HCl) solution (42). Corrosion rates of the Ni-base alloys significantly increased with increasing pressure in an aqueous solution containing H_2SO_4 and O_2 at 400 °C (49). In supercritical oxidizing water, the oxides of nickel-based alloys are generally composed of duplex layer structure, and the inner layer is a chromium-richer layer containing Cr_2O_3 or $NiCr_2O_4$ and the outer layer is a nickel-rich layer containing NiO or Ni(OH), indicating the diffusivity of Ni is fast (24, 36, 37, 56-62). However, the acidic solution would lead to NiO instability in high temperature (58, 63). The most mass reduction and the most severe morphological change were both observed under subcritical condition (29, 30, 45, 64, 65). In contrast to 304L SS, 316L SS and Inconel 690, the oxide on Inconel 625 is too thin to measure in deaerated supercritical water over the temperature range 400 °C to 550 °C (36). However, the Inconel 625 also presented unacceptably high corrosion rate 18 $mmyr^{-1}$ when exposed to oxygenated ammoniacal sulphate solutions in the high-density supercritical region (50). The G30 exhibited the highest corrosion resistance and lowest corrosion rate 5 $mmyr^{-1}$ among various alloys exposed to an aggressive chlorinated SCWO feed (29). The Cr-base alloy Ducrolloy displayed a reduction of weight loss by a factor of 3 for preoxidized in air before SCWO exposure, however the alloy G30 showed no significant effect of the preoxidation on corrosion rate (66).

Titanium Based Alloy

Titanium and titanium alloys (Gr2, Gr5, Gr7, Gr9 and G12) are candidate alloys for conducting corrosion tests at SCWO conditions. However, the titanium cannot reach the mechanical strength requirements of

high temperature high pressure application. Instead, it is suited for using as liners in SCWO systems with high chloride concentration (21, 48, 67-74). The formation of TiO_2 oxide layer played a major role in titanium dissolution in highly chloride (14, 75). Corrosion of Gr2 became severe and Gr12 showed a low corrosion rate among candidate titanium based alloys in the presence of sulfate or phosphate (67). The titanium Gr2 showed more resistance to corrosion than other alloys (such as Inconel 600, Hastelloy C-276, Monel 400, 316 SS and Zirconium 702) in the SCWO process for decomposing halogenated compounds (26, 45, 68). The titanium displayed more resistance to corrosion than 304 SS in high temperature subcritical solution with HCl (76). The corrosion products composed of titanium oxyphosphates and titanium oxide were generated on pure titanium when treating chlorpyrifos with SCWO (48). When HCl, H_3PO_4 and oxidant were presented in SCW, titanium based alloys are an appropriate corrosion resistant material, however, when containing, titanium based alloys crept and suffered severe corrosion.H_2SO_4 (68, 70).

Ceramic Material

Zironia toughened alumina (ZTA), TAI, ALC, Sialon ceramic, HIP-BN, B_4C, Si_3N_4, TiB_2, Y_2O_3, Y-TZP, SiC, mullite, alumina, sapphire, Cr_2O_3 and titania multilayered ceramics were applied for corrosion test under SCWO condition. Corrosion resistance for the ceramics are as follows in this order: Si_3N_4 < SiC < mullite < alumina < sapphire in deionized water at 450 °C and 45 MPa (77). The SiC, alumina, Y stabilized zirconia, Si_3N_4, BN, aluminosilicate and cordierite-mullite showed poor corrosion resistance in supercritical water gasification of sugar beet slurry (78). SiC, Si_3N_4, HIP-BN, B_4C, TiB_2, Y_2O_3 and Y-TZP-based materials showed severe corrosion in oxidizing supercritical water containing HCl, however, Al_2O_3- and ZrO_2-based materials did not corrode severely (79). Due to the formation of non-protecting corrosion products, the ZTA ceramics suffered severe corrosion in high-temperature aqueous solutions containing H_2SO_4 or HCl, however, the formation of berlinite ($AlPO_4$) prevented corrosion of the coupons in H_3PO_4 solution (80). The Cr_2O_3-based ceramics, with yttrium oxide or yttrium oxide-stabilized zirconium oxide added, suffered obvious degradation with oxygen concentrations (4 vol.% hydrogen peroxide) at 650 °C (81). The titania multilayered ceramic system remained intact after 120~180 hours when exposed to a chlorinated cutting oil Trimsol in different SCWO environments, ranging from 300 °C to 650 °C (82). The α-sialon displayed better corrosion resistance and strength than β-sialon on exposure to supercritical water at 450 °C and 45 MPa for 2~50 h (83). The adding proper amounts (≤5 wt.%) of MgO and CeO_2 can enhance the stability of Cr_2O_3 based ceramics in SCW at 650 °C and 25 MPa (84).

Pure Metal and Metal Oxide

Niobium, zirconium, tantalum, gold, iron, nickel, chromium, titanium, molybdenum and chromium oxide were also selected for conducting corrosion tests in SCWO process and these tests were important to investigate the corrosion mechanism. Niobium was resistant to corrosion in oxidizing subcritical water with acid and oxidizing supercritical water without acid, however, niobium or zirconium would rapidly convert to oxides which erode into the fluid in oxidizing supercritical water with acid (50, 74, 85). Tantalum displayed that the corrosion rates increased strongly above the critical temperature in HCl and H_2SO_4, and corrosion rates in H_3PO_4 were low at all temperatures (86). The tantalum was also proved to be the most resistant metal among Hastelloy C, titanium, zirconium, niobium, tantalum and gold in high temperature oxidizing solution containing acid and NaCl (74). The gold would not be suitable to be applied in a SCWO system, due to the galvanic effects of Au (74). In supercritical pure water, chromium shows the lowest oxidation rate among all tested metals and their oxidation rates were in the following order: Fe > Ni > Ti > Mo >> Cr (87). Besides, the addition of H_2O_2 to HCl solutions accelerated the corrosion of Fe, Ni, Cr, and Mo and Ni showed the severest corrosion among five metals (87). The soluble metal cations could precipitate to form a protecting layer at supercritical conditions, indicating the addition of ionic light metal salt could reduce corrosion (88). The bare uncoated tube shows severer breakaway corrosion than TiN and $Ti_{0.35}Al_{0.65}N$ coated tube after exposure to supercritical water (89).

The research on stability of metal oxides was important to investigate the process of oxidation corrosion in high temperature water. The solubilities of three tested metal oxides were in the following order: iron oxide > nickel oxide > chromium oxide, and metal oxide solubility increased with increasing temperature at a higher pH region and at neutral conditions (90, 91).

There is not a material that can endure all SCWO conditions and different materials show different corrosion properties under the SCWO conditions. Accordingly, the most suitable material would be selected for constructing the SCWO system by taking

its corrosion resistance and cost into account. As Table 2 shows, the corrosion tests of iron based alloys, nickel based alloys, titanium alloys, ceramic, pure metals and metal oxides were conducted and the test results are useful for selecting proper material in SCWO plant. The nickel based alloy presented more resistance to corrosion than iron based alloy, however, it also showed the severe corrosion under conditions with aggressive SCWO streams. Titanium based alloy and part of partial ceramic materials presented outstanding resistance to corrosion, however they were always used as liners due to the weak mechanical strength. The corrosion tests of pure metal and metal oxide were conducted for investigating corrosion mechanism, and niobium was recommended to apply at subcritical condition.

The Effects of Alloy Elements on Corrosion Behavior

Under the same condition, different material always displayed different corrosion properties, which mainly depended on the different alloy elements. The recent studies have focused on the behaviors of different elements in candidate materials on the condition of SCWO. The content variation of alloy element, the stability of metal oxide and the element migration were the main contents of the research. The important elements included iron, chromium, nickel, molybdenum and carbon.

Iron

The iron is usually used to improve economy of iron-base alloys, nickel based alloys and titanium-based alloys. In supercritical water, Fe shows the highest oxidation rates and forms stable oxide Fe_3O_4 among the other metal elements (Fe, Ni, Ti, Mo and Cr) and it also shows the major contributor to corrosion resistance at existence of oxygen in supercritical water (36, 37, 87, 92). However, the corrosion rate of Fe in the oxidizing HCl solution is three times higher than that in supercritical HCl solution, indicating the acidic chlorinated solution could also promoted dealloying Fe in high temperature aqueous solution (46, 87, 93). At low temperature 400 °C, Cr could form a protective oxide, indicating Cr-oxide is more stable than Fe-oxide and Fe-oxide solubilities were the highest among Fe, Ni and Cr (51, 91). The Fe always has a faster diffusion velocity than other elements so that the Fe is rich in the outer oxidation layer and Fe-oxides make up the majority of the outer layer at temperature higher than 400 °C (25, 36, 94, 95). The mass loss of Fe is higher than that of Mo at 300~350 °C and lower than that of Mo at 400~450 °C in supercritical water with NaCl (33). The Fe oxides could hardly exist as the temperature increased stably, especially over 500 °C. The Fe oxides could dissolve at high temperature, so the outer layer became loose and desultory. Fe would change to Fe_2O_3, Fe_3O_4 and α(γ)-FeOOH under the SCWO condition.

Chromium

The Cr is considered as the most important alloying element to improve corrosion resistance in SCW (53). It was found that 316 steel stainless would decrease the corrosion rate greatly after the chromizing treatment under SCWO condition using 5% NaCl solution. The test result indicated alloy suffered the most serious corrosion in high temperature subcritical water and chromium had excellent corrosion resistance to chloride ion from subcritical to supercritical condition. The higher Cr concentration would make lower penetration rate of alloys. The chromium oxide presented the lowest solubility among the oxides of chromium, iron and nickel (53, 58, 65, 90, 91, 93). The Cr showed the most stable state among Fe, Cr and Mo in solution with 5 mass% NaCl at temperature of 300~500 °C (33). The dissolved chromium ions can from compact chromium oxide layer to provide a protective function against corrosion at 400 °C and 30 MPa in H_2SO_4 aqueous solution (57, 96). The cracking susceptibility of nickel-based alloys also decreases as the Cr content increased in supercritical water with either HCl or NaOH (97). The higher chromium-containing alloys presented the lower oxide thickness than the higher chromium-containing alloys (60). The oxygen affinity of chromium is stronger than iron and nickel, and Cr would be selectively oxidized to $Cr(OH)_3$ and Cr_2O_3. The Cr(III) of Cr_2O_3 would be transformed into Cr(VI) through further oxidation in the high temperature oxidizing water, and the Cr(VI) would loss during the dissolution. A more continuous and stable Cr_2O_3 layer was formed at higher temperature (450 °C and 500 °C) than critical temperature (400 °C) in oxidizing supercritical water (24). As a result of slower diffusion velocity than Fe, the Cr is usually rich in the inner layer (36, 37, 95). The Cr oxides were also unstable and dissolved in supercritical strong acid solution (pH=1). The Ni-Cr-Mo alloys contain Cr at least 30 mass% show better resistance to corrosion in a supercritical water including chloride (55). The alloy also displays good corrosion resistance even at the Cr contents as high as 40% (40, 54). The NiCr25 alloy performed best resistance to corrosion among binary Ni-Cr alloys (0 to 45 mass% Cr) in an aqueous

Table 2. Summary of corrosion experiences at SCWO conditions.

Type of Material	Material	Oxidant/Acid/ Alkali/Salt	*T* (°C)	*P* (MPa)	Corrosion results
Iron Based	304L	Deionized water	550	24	304L was susceptible to stress corrosion cracking (37).
	304	0.01mol·L^{-1} HCl	380	24	Electrochemical mechanism was the dominant corrosion mechanism (106).
	304L/316L	Deionized water	400-500	24	Outer layer was porous magnetite and the inner layer was a denser iron-chromium spinel (36).
	316L	2.0% H_2O_2	500-550	24	Loose outer layer, interface and inner layer were Fe, Ni and Cr and Fe enrichment, respectively (25).
	316L	4.8wt.% H_2O_2/2wt.% H_2SO_4/2.4wt% NH_3	380-390	24	Corrosion rate was 39 $mmyr^{-1}$ at this condition (50).
	316	2.0% H_2O_2	350-500	24	Fe_2O_3/Fe_3O_4+spinel/Cr_2O_3/Ni-enrichment/316 from the outer to inner layer (107).
	316	3.0% H_2O_2	250-420	39	The maximum corrosion rate was 12.5 $mmyr^{-1}$ at 375 °C; H_2O_2 improved corrosion significantly (108).
	316	5wt.% NaCl	300-500	30	Chromizing treatment decreased the corrosion rate and corrosion mode was pitting (31).
	316	700% H_2O_2/2,4-dichlorophenol	400	25	Surface was mainly covered by carbon-contaminated and iron oxide layers (26).
	316	3% $Ce[NO_3]_3 \cdot 6H_2O$	400	30	Corrosion protection was mainly due to the formation of a Ce oxide layer (51).
	316	100% H_2O_2/200% NaOH/2-Chlorophenol	320/380	22/25	The elevation of destruction efficiency was greater at subcritical than supercritical; the various granule metal oxides covered the surface, mainly included Fe oxides (109).
	316	HCl	350-500	25	Weight losses were significantly increased with the increasing hydrogen partial pressure (103).
	316	1~5wt.% NaCl+0.01~1% H_2O_2	350-500	40	Mass loss was increasing with NaCl content; Layer riched with Cr and Mo, depleted in Ni and Fe (93).
	316	0.025mol·kg^{-1} H_2SO_4+ 0.025mol·kg^{-1} O_2	400	25-60	Good corrosion resistance at 400 °C both at 30MPa and 40MPa; Intergranular corrosion occurred and the corrosion rate increased by more than two orders of magnitude when pressure at 60MPa (49).

	316/625/C276	0.001mol·L^{-1} HCl / 0.01mol·L^{-1} NaOH	400	25	Cracking occurred under both feed solutions (97).
	316+TiO_2	5vol% air+0.1/0.2/ 0.5mmol·L^{-1}HCl	-	-	TiO_2-coated suggested the applied coatings were permeable to sub-critical and supercritical water (32).
	UNS S31803/ Incoloy800/ Incoloy825	5500 m·L^{-1} NaCl	275	25	Dual-phase steel2205 (UNS S31803) had a lowest corrosion rate 1.8 mm/a among all test alloys (101).
	12CrMoV	water	510	9.8	The tube service life make of 12CrMoV(SA516GR70) could reach 350, 000 h (105).
Nickel-Based	Ni-based	0.01mol/kg H_2SO_4	400-650	30	The reduction in corrosion resistance was more significant for alloys with higher Cr content (53).
	Ni-based	0.1mol·L^{-1} HCl	400	25	Cracking susceptibility of Ni base alloys decreased with increasing Cr content of alloys (110).
	Ni-based	1.6g·L^{-1} NaCl/10wt% H_2O_2	400-500	22-25	Chloride leaded to crack initiation and propagation during the first few hours of constant load tests (111).
	Ni-based	pH=1, 2, 7	300/360/ 425	24.1	pH=1: chromium was selectively dissolved and only molybdenum forms stable oxide; pH=2:dealloying of Ni and Fe, and oxidation of Cr and Mo were observed; pH=7: dealloying was not observed at any temperature; SCC in acidic feed streams was observed at all temperatures (16).
	Ni-based	0.01mol/kg H_2SO_4＋ 3ppm to 800ppm O_2	400	30/60	Fe became major contributor to corrosion resistance when oxygen concentration of the environment was increased up to 800 ppm (57).
	Ni-based	0.01mol·kg^{-1} H_2SO_4+ 0.025mol·kg^{-1} O_2	400	25~60	Corrosion rates of the Ni-base alloys at 400 °C significantly increased with increasing pressure (49).
	Ni-based	HCl	300-600	-	G-30 exhibited the highest corrosion resistance; most pronounced degradation being at high subcritical temperatures (29).
	Ni-based	CH_2Cl_2/H_2O_2+2000 ppm chloride	420	40	Weight loss of all the samples varied between 1.3 mm/y for G-30 and 31.6 mm/y for alloy 686; G-30 showed good corrosion behavior due to formation of a protective oxide layer during preoxidized (112).
	625	2.0% H_2O_2	450~500	24	$Ni(OH)_2$/NiO/$NiCr_2O_4$/Cr_2O_3 from outer to inner layer (24).

625	Deionized water	500	24	Own the thinnest oxide layer and exhibited a low density of micrometer-sized pits. A two-layer structure with a chromium-rich inner and an outer layer with higher iron content (37).
625	100%H_2O_2/200%NaOH/ 2,4-dichlorophenol	340/380	25	The mass reduction at supercritical (0.074%) was higher than subcritical (0.026%) (39).
625	4.8wt.% H_2O_2/2wt.% H_2SO_4/2.4wt% NH_3	380~390	23	Corrosion rate was 18 $mmyr^{-1}$ at this condition (50).
625	HCl (pH=2)	355/350/ 299	24.1	Maximum thickness of the dealloyed oxide layer was 7.5 μm/5 μm/4.3 μm; the maximum thickness occurred at high temperature subcritical (40).
625	HCl (pH=2.0)	300-425	24.1	SCC at subcritical temperature; Ni was dissolved and Cr forms stable oxides (58).
625	0.1 $mol\cdot L^{-1}$ $C_6H_4N_2O_5$, 25 $mol\cdot L^{-1}$ $[NH_4]_2SO_4$ and 1.3 $mol\cdot L^{-1}$ NH_3	≤380	22.1	Ammonia and sulfate leaded to solution acidification and NiO instability； The alloy ultimately failed by trans passive dissolution (63).
625	100% H_2O_2/200% NaOH/ 2-chlorophenol	320/380	22/25	Subcritical contributed to the elevation of destruction efficiency was greater than supercritical (39).
625	Deionized water	400-550	24	The oxide on 625 was too thin to measure (36).
625	H_2SO_4/$NaHSO_4$/Na_2SO_4	150~500	38	Severe material loss occurred between 300 °C and 390 °C; Corrosion phenomena in oxygenated (64).
625	Cl^-+ O_2	130~500	38	In neutral or alkaline solutions and in deaerated hydrochloric acid (HCl), corrosion rates of trans passive dissolution decreased drastically (42).
625	0.2 $mol\cdot kg^{-1}$ H_2SO_4+ 1.44 $mol\cdot kg^{-1}$ O_2	250-500	38	The upper temperature limit of severe corrosion at 24 MPa and 390 °C; As temperature was increased further and the density of the solution dropped to low values, only slight corrosion was detected (113).
625 chromium	HCl+H_2O_2+NaCl+CH_3OH	500	46.5	On 625, a strong transition of Ni into the fluid phase; On pure chromium, Cr_2O_3 scales composed of several layers were formed and spallation was observed (52).
C-276	HCl	350-550	25	Containing both Cr and Mo were more resistant to corrosion than Ni-Cr binary alloys (103).

C-276	oxygen concentration＜10ppb	500-600	25	An outer NiO layer and an inner Cr_2O_3/$NiCr_2O_4$-mixed layer (56).
C-276	100% H_2O_2/200% NaOH/ 2-chlorophenol	340/400	22.29/ 25.33	The depth of oxygen penetration in metal at superitical was deeper than at subcritical and a greater loss of alloys at the subcritical than at the supercritical (45).
C-276	700% H_2O_2/2,4-dichlorophenol	400	25	Carbon-contaminated and iron oxide on layers; a signifcant depletion of chromium and nickel (26).
C-276	60wt.% 1,2-$C_2H_4Cl_2$/40wt% C_2H_6SO/35wt.% H_2O_2	538	31	Dealloying of Ni and Fe and cracking of the burst-disk (46).
C-276	PCBs/670wppm chloride	400	30	Iron was detected at 4 mg/g of effluent. Nickel (100), molybdenum (17), chromium (27), and cobalt (1.1 mg/g); The temperature range most violently corroded in the SCWO system was 372-271 °C (99).
800	25ppb dissolved oxygen	500	25	Grain boundary engineering (GBE) enhanced spallation resistance and reducing oxidation rate (114).
Alloy690	1500mg·L^{-1} H_3BO_3/ 2.3mg·L^{-1} LiOH·H_2O	325	15.6	Triple-layer oxide films; the outmost layer was big oxide particles, the inter-layer with loose oxides and the inner layer with incompact cellular oxides (61).
Alloy690	Deionized water	400-550	-	Outer layer was iron-rich and the inner layer was composed of Cr_2O_3, NiO and $Ni(Cr,Fe)_2O_4$ (36).
G-30	pH=2~6, 2000wppm of chloride/H_2O_2	100-400	25-48	At pH 6, the corrosion attack in subcritical region increased with increasing pressure between 150 and 250 °C. A reduced attacks at high pressures, whereas for 250 bars a strong increase was obvious. At pH 2, corrosion attack increased with decreasing pressure between 100 and 380 °C. Above the critical point, the corrosion attack increased with increasing pressure or density of the fluid (41).
G-30/Cr-Fe	CH_2Cl_2/H_2O_2+ 2000wppm chloride	420	40	Preoxidized would improve the corrosion resistance of Cr-Fe; 20 μm oxide film on Cr-Fe and 300 μm oxide film on G-30; G-30 lost nickel down to a depth of 200 μm (66).
Ni-Cr alloys	CH_2Cl_2 + H_2O_2	100-415	40	Kinetics for binary Ni-Cr alloys were compared with the predictions of a mathematical model (98).

	Ni-Cr-Mo	Extraction from the fly ash of incinerated municipal solid waste	400	25	Exhibiting preferable corrosion resistance, the Cr content was 30 mass % at least under this environment (55).
	Fe-Cr (10-40% Cr) Ni-Cr (10-30% Cr)	1mmol·L^{-1} HCl^{-1} + 0.6mmol·L^{-1} H_2O_2	350-500	29.4	Increasing the Cr content improved oxidation resistance but accelerates trans passive dissolution at 500 °C; the addition of Mo (1-2%) to Fe-Cr alloys (> 25%Cr) was found to be effective for improvements of the corrosion resistance; transpassive dissolution rate of Cr was high at temperatures around 350 °C and above 500 °C, but was low at temperatures of 400- 450 °C (54).
Titanium-Based	Gr2	100% H_2O_2/200% NaOH/ 2-chlorophenol	340/400	24	Titanium Gr2 has the more corrosion resistance than Hastelloy C-276 and Zirconium 702 (45).
	Gr2	700% H_2O_2/2,4-dichlorophenol	400	25	Titanium Gr2 was more corrosion-resistant than 316 SS, Titanium G2 and Zirconium 702 (26).
	Gr2/Gr5/Gr7/ Gr12/P-C	O_2/HCl/H_2SO_4/H_3PO_4	600	27	Corrosion in chloride containing solution was low; In the presence of sulfate or phosphate, corrosion of Gr2 became severe and Gr12 showed a low corrosion rate (67).
	Ti-alloy	C_4H_5Cl/ion exchange resin + 2% HCl/2% H_2SO_4 + 2% H_2O_2	400	28.5	Ti alloys would be applied as reactor materials for organic waste decomposition (72).
	Ta, Ti and Ti alloys	Condition 1: 2% H_2SO_4+ 2% H_2O_2 Condition 2: 1.2% H_2SO_4+2% H_2O_2	350	30	Ti alloys was sufficient to be applied as reactor materials (73).
	TAI /ZTA/ ALC	0.1 mol·kg^{-1} H_2SO_4+0.1 mol·kg^{-1} H_3PO_4+ 0.1 mol·kg^{-1} HCl	240-500	27	In H_2SO_4, the corrosion rate was still high at higher temperatures and formed a non-protecting sale; In H_3PO_4, less corrosion due to the formation of $AlPO_4$ on the surface of the specimens; In HCl, dissolution of the alumina grains corrosion phenomenon at temperatures of 240 °C -290 °C, intergranular corrosion at higher temperatures; The order of resistant against intergranular corrosion as follows: TAI>ZTA>ALC (80).
	Four types	Deionized water	450	45	Corrosion resistance as follows: Si_3N_4 < SiC < mullite < alumina < sapphire. High purity alumina ceramics showed the highest corrosion resistance in the ceramics (77).

Ceramics	Sialon ceramics	Deionized water	450	45	α-sialon exhibited better corrosion resistance than β-sialon and α/β-sialon, strength of β-sialon decreased to 65% of its original strength and α-sialon decreased to 90% (83).
	Many types	0.44 mol·kg^{-1} O_2 and 0.05 mol·kg^{-1} HCl	465	25	HIP-BN, B4C, TiB_2, Y_2O_3 and Y-TZP disintegrated; SiC and Si_3N_4-based materials showed a large weight loss, up to above 90%; Al_2O_3- and ZrO_2-based materials did not corrode severely (79).
	Multilayered ceramics / nickel alloys	O_2/chlorinated cutting oil	300-650	-	The experimental nickel alloys performed better than the baseline nickel alloys; A titania multilayered ceramic system had withstood such an environment without significant loss (82).
	Cr_2O_3 based ceramics	Deionized water	650	25	Adding proper amounts (65 wt.%) of MgO and CeO_2 can enhance the stability of Cr_2O_3 based ceramics, because of formation of $MgCr_2O_4$ or reactive element Ce. When the addition content increased to 8 wt.%, the degradation of Cr_2O_3 based ceramics increased (84).
Pure metal and metal oxide	niobium	0.05 mol·kg^{-1} HCl + 0.48 mol·kg^{-1} O_2 0.05 mol·kg^{-1} H_2SO_4 + 2 mol·kg^{-1} O_2	350/500	24	Niobium could be an applicable material for reactors in subcritical water and not be used as reactor material for supercritical water under oxidizing conditions (85).
	niobium	4.8wt.% H_2O_2/2wt.% H_2SO_4/2.4wt% NH_3	380-390	23	The rate of corrosion of the niobium coupons was low (50).
	tantalum	0.05-0.2 mol·kg^{-1} HCl/ H_2SO_4/H_3PO_4+ O_2	360-500	24	The corrosion rates in HCl and H_2SO_4were low at 360 °C and unacceptably high at supercritical temperatures, corrosion rates in H_3PO_4 were low at all temperatures (86).
	Fe/Ni/Cr	HCl + O_2	400	25-40	The corrosion rate of chromium was the smallest; Iron oxide solubility was the highest and Chromium oxide solubility was the lowest among these metals (91).
	Fe/Mo/Cr	5wt.% NaCl	300-450	30	Pure Fe were covered with a thick oxide film; the mass changes of pure Cr were very small; the mass loss of pure Fe is higher than that of pure Mo at 300-350 °C, but is lower at 400-450 °C (33).

Fe/Ni/Cr/ Mo/Ti	1 mmol·L^{-1} HCl	500	29.4	Oxidation rates: Fe>Ni>Ti>Mo>> Cr; The addition of H_2O_2 to HC1 solutions accelerated the corrosion of Fe, Ni, Cr, and Mo; Ni showed the largest weight gain among the five metals examined (87).
Cr_2O_3/Fe_2O_3/ NiO	Deionized water	450	26	Chromium oxide solubility in high temperature region was the lowest among the three oxides; Metal oxide solubility increased with increasing temperature (90).
Ti/Zr/Nb/Ta/ Au/SS/ Hastelloy C	0-0.02 mol·kg^{-1} Acid 0.1-1 mol·kg^{-1} O_2 0-0.2 mol·kg^{-1} NaCl	200-400	26	Nb and Zr rapidly converted to oxides; Ti proved resistant to dissolution, but was subject to pitting corrosion; Au was subject to galvanic effects; Ni and Mn of alloys preferentially dissolving out of the metal and Cr and Mo remaining begin; Ta proved to be the most resistant metal tested (74).
Ni/Mo/Cr	HCl	350	24	Ni and Mo showed strong material loss; Corrosion products formed at the surface consisted of oxygen and chromium (65).

solution resulting from the oxidation of CH_2Cl_2 at 40 MPa and temperatures between 100 °C and 415 °C (98).

Nickel

The toughness, strength and corrosion resistance of the steel would be enhanced when it contains Ni element. The oxygen affinity order for metal element is Cr ＞ Fe ＞ Ni, indicating the Cr and Fe could easily form the oxides under the SCWO conditions. The nickel based alloys often show a severe depletion of nickel and the migration processes of elements presented in Figure 1. In this simplified model, nickel ions migrated to solution and ultimately changed to dissolved form. Oxygen ions migrated to the matrix and combined with the chromium to form chromium oxide. In contrast to other metal elements like Fe, Cr, Mo, the Ni usually presents a worst serious depletion and the highest concentration of nickel is detected in the corrosion effluent, especially in acidic chlorinated solution (26, 40, 46, 52, 58, 66, 93, 99, 100). The pure chromium presents greater corrosion rate than Inconel 625 in supercritical water containing HCl, NaCl and H_2O_2 (52).

Molybdenum

The Mo can improve high temperature strength, resistance to pitting corrosion and resistance to crevice corrosion in the high temperature environment. The Mo could from more stable Mo-oxides (MoO_2, MoO_3) than other metals in strong acidity and chlorinated solution at subcritical or supercritical temperature (16, 93). The Mo improved the resistance to pitting corrosion in subcritical salt solution obviously[101]. In nickel based alloys, the synergistic effect of Mo and Cr showed more resistant to corrosion than Ni-Cr binary alloys (102, 103).

Carbon

The hardness of steel stainless increased and the malleability decreased as the Carbon content increases. Carbon contamination was significant when treating organic wastes with SCWO, implying the destruction reaction occurs also on the material surface. Therefore, the high content of carbon could be observed in the outer layer oxides (26).

Other elements like niobium, titanium, zirconium and tantalum were also investigated under the SCWO conditions. Titanium, niobium and tantalum are stable as oxides in subcritical condition and these elements could enhance the corrosion resistance in subcritical condition (50). Ta_2O_5 and Nb_2O_5 are both the stable states in subcritical condition. However, the Zr oxides were more easily dissolved at the supercritical condition than at the subcritical condition.

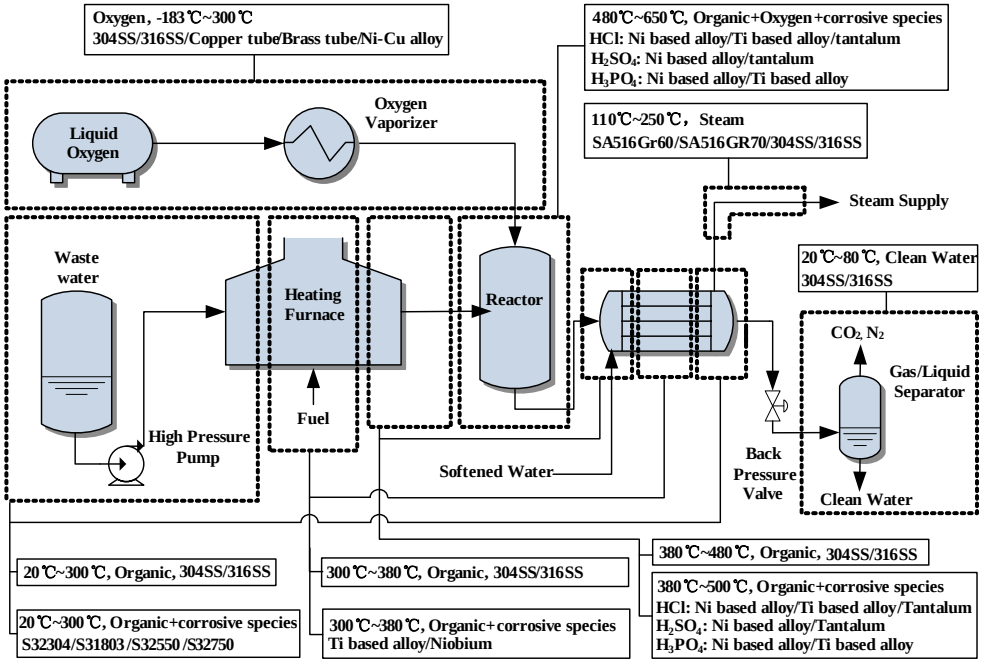

Figure 2. Material selection for SCWO system in flow diagram.

Suggestions for Material Selection in SCWO System

As shown in Figure 2, the material selection of a conventional SCWO process was divided into seven parts according to the different reaction conditions and mediums. In this flow diagram, oxygen and waste water both entered reactor after they were preheated by different preheating pipes, which could avoid synergistic effect of oxidant and aggressive feeds effectively.

The Heating Pipe for Oxygen

The high carbon alloys are not recommended to build transport pipe for oxygen, due to the fact that a high rate of oxygen could react with the carbon, resulting in explosion. The copper tube, brass tube and Ni-Cu alloy can be used for transporting the high speed oxygen, and the stainless steel can be used for transporting the low speed oxygen among -183 °C~300 °C at supercritical pressure (104).

The Low Temperature Heating Pipe for Feed

The low temperature preheating process is used for preheating feed from normal temperature to 300 °C. There are two situations that should be taken into consideration for materials selection in this low temperature process. In the first case, 304 SS and 316 SS are recommended at the weak corrosive condition that the feeds did not contain halide. In the second case, dual phase steel can endure the severe corrosion due to the feeds containing halide. The dual phase steels like S32304, S31803, S32550 and S32750, own an excellent corrosion resistance to halide, however they cannot be used at temperature over 300 °C due to the decline of mechanical strength (101). The materials applied to the low temperature preheating pipe for wastewater could be also used in the low temperature zone of vapor generator.

The High Temperature Subcritical Heating Pipe for Feed

The high temperature heating process is used for heating feed from 300 °C to 380 °C and this high temperature subcritical process is considered as the severest corrosion region in the SCWO system. When the feed does not contain corrosive species like halogenated hydrocarbons or halide ion, 304 SS or 316 SS would be selected as candidate materials. When the feed contain corrosive species in this condition, the titanium based alloys, niobium and tantalum are the suitable materials for constructing the heating pipe. The medium temperature zone of vapor generator could use the same materials which are applied to the high temperature heating pipe for feed.

The High Temperature Supercritical Heating Pipe for Feed

This is high temperature supercritical area from 380 °C to 480 °C in heating furnace. The 304 SS and 316 SS would also be applied for constructing the supercritical heating pipe in absence of halide. When the feed contained the aggressive species like HCl, H_3PO_4 and H_2SO_4, the nickel based alloys are usable materials and the corrosion rates should be measured

through corrosion tests under different conditions. However, the titanium based alloys showed a good resistance to corrosion in the oxidizing supercritical water containing aggressive species. The materials selection of this process was as same as that of the high temperature zone of vapor generator.

The Reactor

The temperature of reactor is usually among 480 °C to 650 °C and the hazardous waste decomposes immediately in this process. The reactor would suffer a severer corrosion from the synergistic effect between oxygen and aggressive species than that from the individual effect. The materials used in the high temperature supercritical heating area could also be used for constructing reactor. Moreover, the wall thickness of reactor should be thick enough to reach the strength requirement under this high temperature condition.

The Pipe for Transporting Steam

The work temperature of pipe for transporting steam is usually among 150 °C to 250 °C and the softened water was heated into create clean steam which was a weak corrosive medium (105). The low alloy steel like SA516Gr60 and SA516GR70 could be used to construct the weak corrosion part in the system. The high alloy steel like 304 SS and 316 SS are also recommended as the constructing materials in this process.

The Separation Unit for the Gas and Liquid

The heat exchanger cooled the treated wastewater from supercritical temperature to room temperature in this unit. The work temperature of this unit among 20 °C to 60 °C, and the common stainless steel (304 SS, 316 SS) are the suitable materials in this low temperature area.

The selection of materials depended on corrosion resistance and mechanical strength of materials for constructing SCWO plant, especially at supercritical condition. In this section, a guidance of candidate materials selection is supplied for SCWO system. According to the above suggestion, the kinds of candidate materials would be determined. Then, the corrosion behavior and rate of selected material should be investigated through the corrosion test in specific condition, which is the foundation work for building SCWO system.

Conclusions

The aggressive and corrosive feed causing the severe corrosion prevents the large-scale application of SCWO, even though SCWO is a high efficient process to treat organic waste. Among all the candidate materials, the nickel based alloy is the most widely studied and used constructing material for SCWO. As a candidate material, Inconel 625 is used more frequently than the other nickel based alloys. The corrosion results of all candidate materials were summarized to investigate the corrosion properties of different kinds of materials. The material selection of a whole SCWO process includes seven parts and the guideline of material selection is provided in detail. The effects of the alloy elements on the corrosion properties had contributed to investigate the mechanism of corrosion at SCWO condition. The above work is helpful for selecting candidate materials and developing effective corrosion control methods.

In conclusion, no single material can withstand all kinds of SCWO conditions, however the suitable material could be selected for a certain condition, corrosion resistance, strength and economy taken into consideration. The appropriate corrosion control approaches could also reduce the corrosion effectively, especially in the highly corrosive feed with high pressure and high temperature requirements. In order to accomplish the commercialization of SCWO, much deficiency of work in controlling corrosion and the future work are summarized as follows.

1) On-line corrosion monitoring methods. The corrosion monitoring technologies such as electric resistivity technology, pulsed magnetic flux leakage technology and ultrasonic technology can be applied in SCWO system. The effective monitoring methods can predict the service life of the equipment and the appropriate corrosion control technology can be used to reduce corrosion timely. The corrosion monitoring and corrosion control methods can prevent dangerous accidents from happening under condition of high temperature and high pressure.
2) Corrosion control approaches. The effective control approaches should be applied to reduce the corrosion in the whole SCWO system. Based on the analyzing of various reasons for the corrosion, the control approaches consists of three aspects: the equipment structure optimization, reducing the corrosion of reaction medium and optimizing operation parameters. The effective corrosion control approaches could reduce the the manufacturing cost and the service life of equipment would be prolonged.
3) Corrosion test under the real condition. The high pressure and flow must be considered as the important factors in the further corrosion test. In

the previous corrosion researches, these two important factors were rarely taken into account. The high pressure and flow which can accelerate corrosion significantly are consistent with the real situation.

4) Exact calculation model. Based on corrosion test results and theoretical analysis, the exact calculation model should be established for predicting the tendency of corrosion. The good model would make up for the shortage of monitoring methods under high temperature and high pressure condition. The effective factors and causes can be analyzed according to the accurate simulation calcution results.

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