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Chapter 3

Hot Corrosion of Superalloys in Boilers for Ultra-Supercritical Power Plants

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Additional information is available at the end of the chapter

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Abstract

The coal-based power plants have been plagued by twin problems—low thermal efficiency and emission of high level of pollutants into the environment. Over the last few decades, attention was paid by researchers worldwide to overcome these problems and to design, build, and operate coal-based plants with improving efficiency levels and reducing emission levels. Operating the power plants with higher levels of steam temperature and pressure was adopted as the direction toward achieving the needed improvements. The requirement to operate the plants with increasingly higher levels of temperature and pressure made it necessary to design the components with superalloys. Hot corrosion then becomes a major design consideration, particularly for superheaters and reheaters. Thus, it becomes important to study the hot corrosion behavior of candidate superalloys. The present chapter attempts to review the work done over the last two decades to understand the hot corrosion behavior of superalloys in the context of their use in advanced coal-based power plants.

Keywords: ultra-supercritical coal fired power plants, nickel-based superalloys, hot corrosion, fireside corrosion, sulfidation

1. Introduction

Ultra-supercritical (USC) power plants are expected to overcome two challenges—(i) an increase in demand for electricity and (ii) a significant reduction in the enhanced greenhouse effect due to CO_2 emission from the fossil fuel. By increasing the operating temperature and pressure of the steam systems, an increase in the power plant efficiency and a major reduction

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in the CO_2 emission from the power plants based on firing pulverized coal can be realized [1]. The goal of the latest designs in power plants is to generate 760°C steam. In the process, metal surface temperature of the key components such as superheaters and reheaters may reach as high temperature as 800°C [2]. The materials used for superheaters and reheaters in coal-fired boiler environments always suffer coal ash hot corrosion at elevated temperatures [3, 4]. Coal ash hot corrosion is the result of competition between oxidation and sulfidation. Sulfidation comes into picture as the materials get exposed to high-temperature combustion gases containing sulfate coal ash [5]. Superalloys, in particular, nickel-base superalloys, have been the candidate materials for application in boilers for ultra-supercritical power plants based on firing coal. Compared to different types of high-temperature steels, nickel-based superalloys are well known for their superior resistance to coal ash hot corrosion [6, 7]. While there have been several publications on use of high-temperature materials in advanced power plants, a systematic review of application of superalloys for critical components facing hot corrosion and high temperature creep loading conditions in advanced coal-based power plants has not been carried out in the recent years. This chapter proposes to bridge this gap.

2. What are ultra-supercritical power plants?

Because of the abundant availability of coal reserves in the world, the use of coal for electricity generation has been done extensively. Coal has thus dominated as fuel for electricity generation, even though many modern technologies have emerged for power generation. Use of coal for power production is, however, beset with serious problems. Coal-based power plants emit pollutants and CO_2 at high levels. Further, the efficiency of the conventional coal-based power plants has been very low, 35–40%. The efficiency of the power plant can be increased by increasing the operating steam temperature and pressure. Development studies were undertaken worldwide to build power plants with increased values of these two operating parameters. In addition to increased thermal efficiency, an increase in these two parameters would lead to a reduction in greenhouse gases, an issue where the public is becoming increasingly conscious of.

Several of the existing coal-based power plants belong to the category of subcritical power plants operating at a steam pressure of ~22 MPa and a steam temperature of ~550°C; the thermal efficiency of these plants is in the range of 35–37%. The plants operating in the temperature range of 540–580°C with steam pressure extending up to 25 MPa are termed supercritical power plants; their efficiency is of the order of 45%. Ultra-supercritical power plants operate

	Main steam pressure, MPa	**Main steam temperature, °C**	**Reheat steam temperature, °C**	**Average efficiency**	**CO_2 emissions (g/KWh)**
Subcritical	<22.1	Up to 565	Up to 565	36	766–789
Supercritical	22.1–25	540–580	540–580	45	722
Ultra-supercritical	>25	>580	>580	>45	<722

Table 1. Classification of power plant conditions [8].

at temperature higher than 580°C and pressure higher than 25 MPa, leading to an efficiency level of more than 45%. All over the world, several plants have been set up conforming to ultra-supercritical technology. Recent advances in the alloy making and processing capability to produce different items of hardware from these alloys have resulted in the possibility of setting up advanced ultra-supercritical power plants, where the operating temperature would be 700°C or more and operating pressure in the range of 30–35 MPa. The associated efficiency is expected to be equal to or better than 50%. These advancements in terms of temperature and pressure parallelly result in a reduction in emitted CO_2 levels. **Table 1** gives the details.

3. Hot corrosion in coal-fired boilers

Materials that exhibited strength to withstand the higher temperatures and pressures present in the superheaters of supercritical and ultra-supercritical advanced boilers with required stress rupture strength were available, but they proved to be especially susceptible to corrosion by certain coals, most notably high sulfur-bearing varieties.

The cause of this type of corrosion, referred to as coal ash corrosion or hot corrosion or fireside corrosion in advanced power plant terminology was soon understood. It is now generally accepted to be due to the presence of liquid alkali iron trisulfates on the surface of the superheater and reheater tubes beneath an overlying ash deposit. Coal ash corrosion or hot corrosion is a widespread problem for superheater and reheater tubes, especially when high sulfur, high alkali, and high chlorine coals are used, and is a crucial problem that needs to be resolved before advanced ultra-supercritical boilers can be deployed. The loss from corrosion increased with high levels of SO_2, alkali sulfates, and temperature.

Performance of nickel superalloys in such industries as aerospace or gas or marine turbines has been well documented. However, it is important to note that the operating conditions within a coal-fired boiler are very different in terms of the service environment. Hot corrosion as it occurs in coal-fired boilers is a complex topic since it is a consequence of the combined effect of combustion gas atmosphere, fly ash deposits, and the alloy itself.

4. Components/subsystems subjected to hot corrosion in ultra-supercritical power plants

High-performance materials are required to be used for the manufacture of different sections in boilers and steam turbines of an ultra-supercritical power plant. **Table 2** summarizes the prime design requirements for the selection of materials for various critical parts in boilers and steam turbines. The superheater and reheater tubing are among the sections exposed to severe environmental conditions. The outside surface of the tubing suffers fireside coal ash corrosion, while the internal surface faces steam-side oxidation conditions. In addition, the materials used for the subject tubing have to possess adequate stress rupture strength, as they are subjected to long hours of mechanical loading at elevated temperatures.

	Design requirements
Boiler	
Headers/steam pipes	Thermal fatigue strength, weld strength
Reheater and superheater tubes	Creep rupture strength and fireside corrosion resistance
Steam turbine	
Casings/shells	Steam-side oxidation resistance
Bolting	Resistance to stress relaxation up to maximum steam temperature
Rotors/discs	Steam-side oxidation resistance, creep rupture strength, resistance to thermal fatigue cracking
Vanes/blades	Steam-side oxidation resistance, solid particle erosion resistance

Table 2. Components in ultra-supercritical power plants where superalloy grades have come into consideration.

5. When does the designer choose superalloys for these components?

Development of USC power plants requires high-performance alloys able to resist the high temperature and pressure conditions occurring in such plants. Materials for superheaters and reheaters need to possess high creep strength and fireside corrosion resistance when it comes to fulfilling the requirements of highly efficient power plants. Development of qualified alloys for boiler heat exchangers was a very important task in the last decade. At temperatures higher than about 600°C, martensitic alloys are limited by their creep strength performance [25]. Accordingly, substitution of ferritic and martensitic materials with more creep-resistant materials became necessary. Aiming at efficiencies above 40%, iron-base austenitic alloys came for consideration and design with nickel-base superalloys became essential for 700°C technology. Since the beginning of advanced ultra-supercritical power plant concept in the 1990s in Europe, different nickel-based superalloys have been evaluated for their fireside corrosion, steam-side oxidation, and resistance to creep and stress rupture. **Table 3** gives the chemical composition of nickel-based superalloys that have been the subject of studies in the context of their usage on USC power plants.

6. Overview of work done in the area by different researchers

A number of superalloy grades were studied by different researchers for their hot corrosion behavior in USC power plants. **Table 4** gives a summary of these studies. Different material characterization techniques were adopted to study the nature of corrosion products appearing on the exposed surface; **Table 5** gives a list of them. Based on the characterization, conclusions were drawn on the phases occurring in the scale resulting from hot corrosion. **Table 6** summarizes the findings of different authors for the two superalloy grades 617 and 740. Some of the studies carried out in the past also looked into the relative performance of different superalloy grades in USC power plant environments. Published information in this respect has been summarized in **Table 7**.

Sl. No.	Material	Ni	Cr	Co	Mo	W	Al	C	Fe	Mn	Si	S	Ti	Cu	B	Nb	La	P	Refs.
1	Inconel 740	49.5	25.0	20	0.5	—	0.9	0.03	0.7	0.3	0.50	<0.002	1.8	<0.1	0.002	2.0	—	<0.002	[9]
2	Inconel 740H	49.0	25.0	20	0.5	—	1.35	0.03	0.7	0.3	0.15	<0.002	1.35	<0.1	0.002	1.5	—	<0.002	[10]
3	Inconel 617	>44.5	20.0–24.0	10.0–15.0	8.0–10.0	—	0.8–1.5	0.05–0.15	<3.0	<1.0	<1.0	<0.015	<0.6	<0.5	<0.006	—	—	—	[11]
4	Haynes 230	57	22	<5.0	2.0	14	0.3	0.1	<3.0	0.5	0.4	—	<0.1	—	—	<0.5	—	—	[12]
5	Haynes 282	57	20	10	8.5	—	1.5	0.06	<1.5	<0.03	<0.15	—	2.1	—	0.005	—	—	—	[13]
6	Alloy 263	Bal.	19.0–21.0	19.2–21.0	5.6–6.1	—	1.9–2.4	0.04–0.08	<0.7	<0.60	<0.40	<0.007	2.4–2.8	<0.20	<0.005	—	—	—	[14]

Table 3. Chemical compositions of candidate superalloy grades for ultra-supercritical power plant applications.

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Sl. No.	Super alloy	Refs.
1	740	[5, 15–19]
2	740H	[17, 20]
3	Haynes 282	[21]
4	Alloy 263	[17, 18]
5	Alloy 617 and derivatives	[17, 18, 20, 22, 23]
6	600,601,690,602CA,214,45TM,HR160,693	[24]
7	Alloy 230	[20, 24]

Table 4. Superalloy grades studied for their hot corrosion behavior in USC power plat environments.

Sl. No.	Characterization techniques used
1	Thermogravimetric analysis (TGA)
2	Scanning electron microscopy (SEM)
3	Energy-dispersive spectroscopy (EDS)
4	X-ray diffraction (XRD)
5	Electron-probe microanalysis (EPMA)

Table 5. Techniques used by researchers to characterize the hot corrosion in simulated USC power plants.

Sl. No.	Material	Phases	Refs.
1	Alloy 740	NiO, Cr_2O_3, CrS, Ni_3S_2, (Ni, Co)Cr_2O_4, $Fe(CrAl)_2O_4$	[15]
3	Alloy 740	Co_3S_4, Ni_3S_2, (Ni, Co)Cr_2O_4, Al_2O_3, TiO_2, $NaKS_2O_7$, Cr_2O_3,	[19]
2	Alloy 740	$Cr_2(SO_4)_3$, Ni_3S_2, Co_3S_4, CrS, TiS,	[5]
4	Alloy 617	Cr_2O_3, Al_2O_3, TiO_2, CrS,$NiCr_2O_4$, $CoCr_2O_4$, $FeCr_2O_4$, Fe_2O_3, $Cr_2(SO_4)_3$, NiO	[23]

Table 6. Phases observed in the scale formed after hot corrosion of different superalloy grades in simulated USC power plant environment.

Sl. No.	Alloy grades studied	Rating based on corrosion resistance	Refs.
1	740, 263, 617	740 > 263 > 617	[18]
2	740, 740H	740H > 740	[17]
3	617, 617B	617B > 617	[22]
4	740H, 263, 617	740H > 263 > 617	[10]

Table 7. Relative rating of candidate superalloy grades from the point of view of hot corrosion in simulated USC power plants.

7. Mechanisms of hot corrosion of superalloys in USC power plants

7.1. Initiation and propagation of hot corrosion reaction

From the studies carried out in the past, it emerges that hot corrosion reaction occurs in two phases. In the first phase, referred to as initiation phase, there is essentially no weight change, and the chromium oxide layer formed on the surface is still protective in nature. In the second phase, referred to as propagation phase, the chromium oxide layer gets fluxed and loses its protective nature. Degradation of the material starts, manifesting itself as a loss in weight. It is often considered that extent of initiation phase is the safe regime for deploying the material in service facing such hot corrosion conditions. If the corrosive environment is more aggressive, the initiation phase may get reduced and propagation phase may take off earlier. For example, compared to simulated coal ash environment, (simulated coal ash + flue gas) environment is more aggressive; in the latter environment, initiation phase was found to be shorter, propagation phase taking off earlier [23]. **Figure 1** schematically illustrates these issues.

7.2. Effect of chemical composition

7.2.1. Effect of chromium level

Gagliano et al. [26] carried out studies on 19 alloys varying in chromium content from 16 to 44% and concluded that material's resistance to coal ash corrosion is primarily dependent on chromium content, with alloys containing greater than 22% chromium generally exhibiting satisfactory corrosion resistance. According to the rule of thumb in boiler construction industry, with increasing chromium content in the alloy matrix, lower corrosion rates are expected due to higher ability of sufficient diffusion barrier formation [27]. Based on hot corrosion studies carried out on a number of special steels and superalloys, it was observed that weight loss due to hot corrosion decreases with increasing chromium content. This is shown schematically in **Figure 2**. This tendency appears to be true for iron-based austenitic alloys, as confirmed within the COORETEC study [28, 29]. But for nickel-based austenitic alloys, this tendency cannot be confirmed when SO_2/SO_3-rich atmospheres were present. Both grades 617 and 263 have similar chromium content; however, they perform very differently in SO_2/SO_3-rich environment. Even though chromium content of alloy 617 is high lying at 22%, the oxide scale is apparently not capable of forming a diffusion barrier tight enough to protect metal against sulfur-induced corrosion [18].

7.2.2. Effect of cobalt

Cobalt is less resistant to hot corrosion than nickel. Nickel-based alloys are in general more resistant to low-temperature hot corrosion than cobalt-based alloys. For example, Tiwari [30] studied the relative hot corrosion behavior of a nickel-based superalloy and a cobalt-based superalloy in Na_2SO_4-60% V_2O_5 environment at 900°C and reported that the latter had inferior corrosion resistance.

Zhao et al. [15] reported the hot corrosion reaction in superalloy 740 propagated by the outward migration of Cu and the inward migration of SO_3. With the progressive formation of

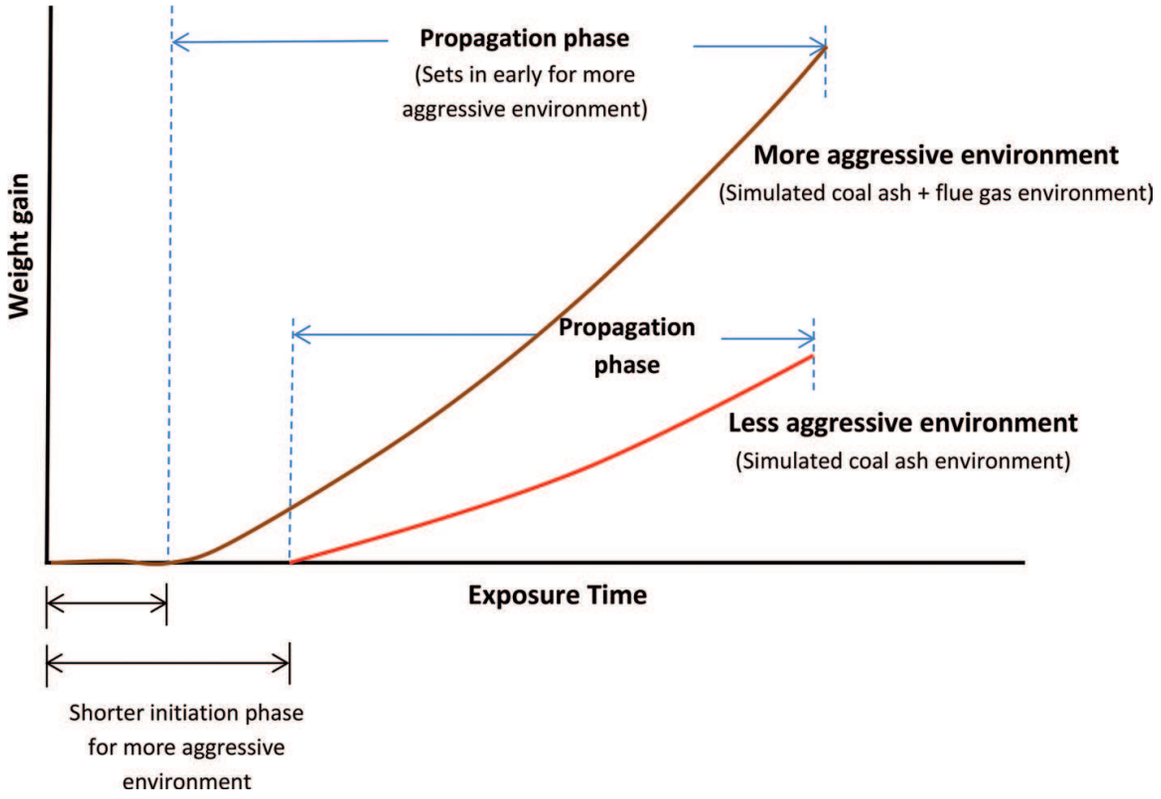


Figure 1. Schematic showing the initiation and propagation phases in the hot corrosion process.

Cr_2O_3 scale, there is an increase in Cr-depletion and enrichment of Co and Ni in the layers immediately below the scale. Due to the high diffusivity of Co in the scale, CoO forms on the exterior of the scale. CoO is surrounded by Cr_2O_3, and the solid-state reaction occurs to form $CoCr_2O_4$ spinel in the outer of the scale gradually. CoO forming on the surface layer reacts with SO_3 to form $CoSO_4$. Even though NiO may form on the surface and $NiSO_4$ formation could result through the reaction of NiO and SO_3, it is believed that $CoSO_4$ forms preferentially due to the fact that $CoSO_4$ is much more stable thermodynamically than $NiSO_4$. The dissolution of Co and CoO induces severe low-temperature hot corrosion. It may be noted in this context that some candidate superalloy grades have considerable Co level in the chemical composition. For example, superalloy 740 has as much as 19–20 wt% Co, and alloy 617 has 10–11 wt% Co. Zhao et al. [15] suggested that a decrease in Co level of the superalloy 740 will act in a beneficial way to resist coal ash/flue gas corrosion. Further studies are required to evaluate the effect of a decrease in the level of Co on the hot corrosion behavior of alloy 740.

7.2.3. Effect of molybdenum

Catastrophic or self-sustaining rapid hot corrosion can occur in superalloys, which contain molybdenum because molybdenum oxide can react with Na_2SO_4 in the salt [31]. The effect of Mo on hot corrosion of superalloys was reported [32–35]. The alloy containing Mo suffers catastrophic degradation. It is reported that MoO_2 reacts with Na_2SO_4 to produce an acid salt (SO_2-rich), leading to acidic fluxing. The MoO_3 may get incorporated into the Na_2SO_4 via the formation of complex compounds such as Na_2MoO_4, $Na_2MoO_4.MoO_3$, and $Na_2MoO_4.2MoO_4$.

All these phases are liquid and have a high solubility of Al_2O_3 and Cr_2O_3. It may be noted in this context that some of the candidate superalloys have significant levels of Mo. For example, alloy 617 has as much as ~9 wt% Mo in its chemical composition.

7.3. Effect of temperature

Operating temperature is one of the important factors that influence sulfidation tendency in coal ash hot corrosion of nickel-based superalloys [5]. A high sulfidation tendency can be observed at low temperatures due to the formation of low-melting alkali sulfate eutectic mixtures [5]. This is the reason why Ni-based superalloys suffer rapid degradation in the temperature range of the so-called Type II hot corrosion. But, the sulfidation tendency decreases at the high-temperature form of hot corrosion, in the temperature of the so-called Type I hot corrosion. Extent of hot corrosion of Inconel 740 was found to be an important function of the temperature in the temperature range of 700–800°C studied by the authors. The corrosion rate was well in the acceptable range at 700°C. There was a 10-fold increase in the rate on increasing the temperature to 750°C, and this was associated with large-scale internal sulfidation and TiS formation. The corrosion rate at 800°C was lower than at 750°C, and the extent of TiS formation was also lower than at 750°C.

The course of corrosion reaction can change with operating temperature. For Ni-XCr-10Al alloy, corrosion follows a multistage parabolic rate law at 700°C, but it changes to a quasiparabolic rate at 800°C [36].

Zhao et al. [15] studied the hot corrosion of alloy 740 at 550 and 700°C. Molten sulfate-induced hot corrosion did not occur at 550°C since the sulfates on the coal ash do not melt at this

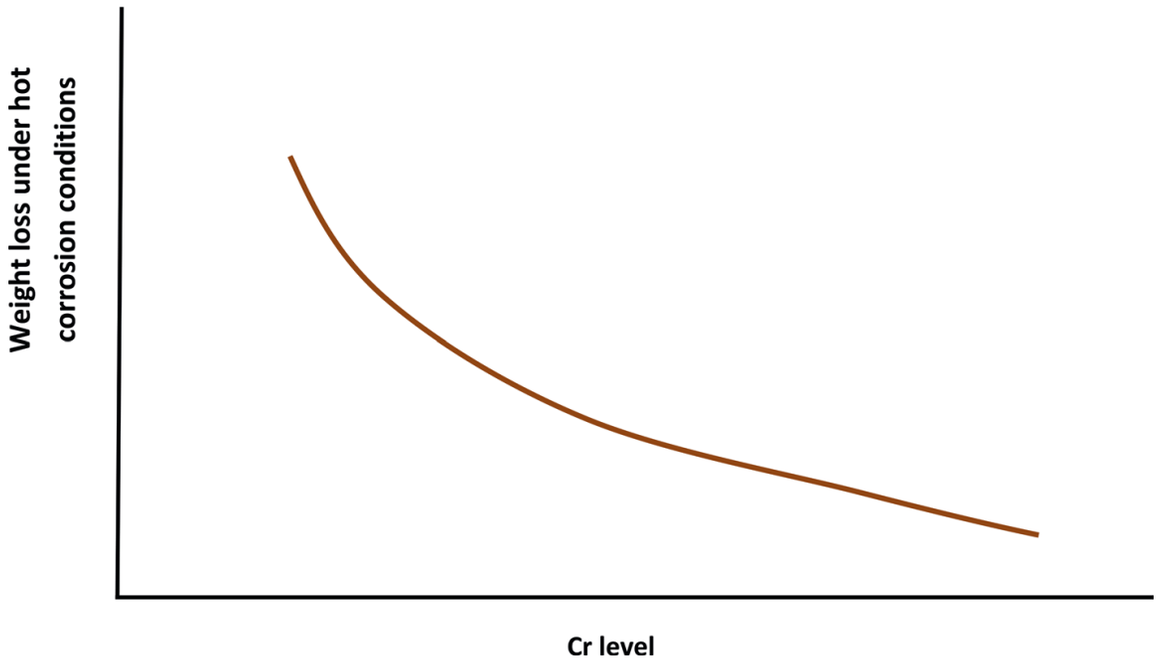


Figure 2. Schematic of the dependence of weight loss due to hot corrosion at 650°C on Cr content based on data obtained for various special steels and superalloys.

temperature. At 700°C, due to the formation of low-melting point eutectic, molten sulfates form and cause accelerated attack after about 1000 h of exposure. No internal sulfidation occurred at 550°C, while sulfides precipitated at scale/alloy interface and in Cr-depletion zone at 700°C. The spinel phases occur in corrosion product at 700°C, while at 550°C that was not the case.

7.4. Effect of aggressive species in the environment

The type and extent of hot corrosion are a function of the composition of flue gas, particularly its SO_2 content [19]. It has been demonstrated that the extent of hot corrosion of Inconel 740 in the flue gas with SO_2 was higher than that in the flue gas without SO_2 [19]. The extent of sulfidation attack was much higher in the flue gas with SO_2. SO_2 in flue gas accelerated the corrosion damage of the alloy in coal ash environment. The metal loss was found to be doubled after 123 h of exposure on switching over from flue gas without SO_2 to flue gas with 1% SO_2. The sulfidation was predominant in the propagation stage.

8. Conclusion

The subject of hot corrosion of superalloys in coal-fired boilers in advanced power plants is a highly complex phenomenon, with a large multiplicity of variables coming into picture. The chemical composition of the alloy plays an important role in this context. High levels of cobalt and molybdenum may have a detrimental effect. While high levels of chromium were found to be useful in combating hot corrosion, a simple relationship between chromium level and resistance to hot corrosion does not seem to hold good when environments contain sulfur-bearing species. Clearly, further work is warranted to optimize the chemical composition to combat hot corrosion while maintaining the stress rupture strength and microstructural stability over long periods of power plant operation. Increasing levels of sulfur-bearing species in the environment increase the extent of sulfidation and susceptibility to hot corrosion. Temperature is also an important parameter in influencing the hot corrosion process; the progress of corrosion reaction and the phases present in the corrosion product could be very different, depending on the reaction temperatures.

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