

Corrosion Suppression Coatings for Molten Nitrate Salt

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Abstract. The use of solar energy for power generation provides an efficient sustainable energy solution. Among a number of technologies developed for power generation using solar energy, concentrating solar power (CSP) is encouraging because it makes use of mature technology in the power block. Thermal energy storage (TES) is added to CSP, making it competitive with other power generation technologies and delivering dispatchable energy. Molten salts are one of the materials of choice for the TES. Although the use of molten salts as heat transfer fluid and thermal storage in CSP has various advantages, storage tanks and pipework can be highly susceptible to corrosion. Different approaches have been adopted to suppress corrosion including the use of specialised alloys and high purity molten salts; however, both contribute to a substantial increase in construction and operating costs.

In this study, a literature review is provided on coatings to suppress the hot corrosion of the storage vessels and pipework containing molten salts. There has been widespread use of anticorrosion coatings for numerous applications, providing guidelines to develop anticorrosion coatings for TES. Various important factors to be considered for choosing coating material are described herein. To date, several published studies discuss the corrosion resistance of different alloys and coatings for different applications. This study reports on corrosion tests and oxidation tests, while making comparison between different alloys with use of data extracted from literature. Among other materials studied the nickel aluminium alloys exhibit very promising properties as protective coating.

Keywords: Thermal Energy Storage, Molten Nitrate Salts, Corrosion Resistant Coatings

1. Introduction

The capability of TES integration in the CSP plants makes them more appealing than other renewables technologies because of its high practicality and proficiency commercially [1–8]. Dispatchability is significant to supply power through grid on demand. For continuous power generation, a reasonably priced and consistent energy storage method is required [9,10]. The LCOE of CSP is significantly reduced with the use of TES, A decrease in LCOE of about 10% has been stated for integrating a 12-hour storage capacity TES system. TES is an order of magnitude lower cost than electrical (battery) storage.

An extended literature search revealed that the scientific and technical papers dedicated to the development of anticorrosion coatings for solar thermal applications are limited. This literature review deals with the critical aspects which are related to the good performance and long-term viability of TES system. The use of coating does not only allow corrosion protection

for container material but also protects the molten salt from the ingress of corrosion products. Corrosion-resistant structural materials are quite expensive, and protective coatings allow less expensive materials to be used for the structures.

2. Corrosion suppression

Hot corrosion becomes uncontrollable by the super alloys alone at elevated temperatures for extended time duration. One of the solutions to this situation is to apply shielding coating layer to low-alloy steel [11]. Protective coating can be divided into two basic groups: diffusion and overlay coatings. Diffusion coatings are formed by diffusion of one or more elements into the surface of the metal to be protected.

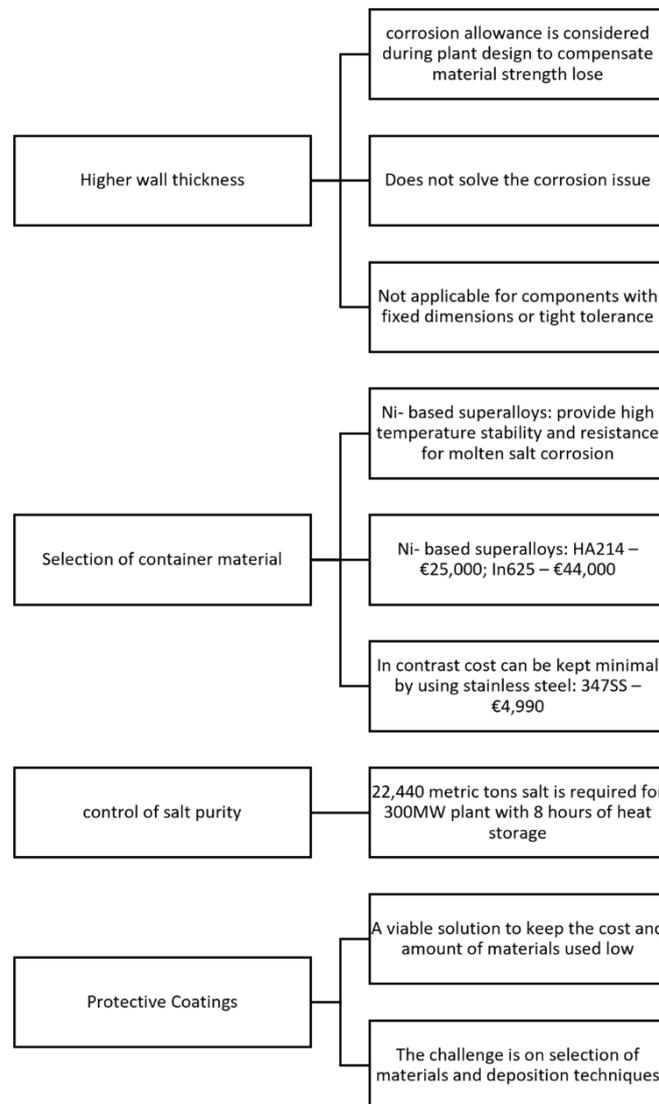


Figure 1. Current corrosion protection strategies used for molten nitrate salt TES. Higher wall thickness, use of high purity salt, selection of specialised alloys is among the different strategies currently employed for corrosion suppression.

A number of factors need to be considered while choosing coating material suitable for use in molten nitrate salts environment. The coatings should necessarily own a capacity to form a steady, slow-growing, passivating surface oxide to offer an obstruction between the coating, alloy, and environment, hence providing oxidation and hot corrosion resistance. Coatings should have microstructural strength to keep their protective properties for prolonged ser-

vice life, even at high temperatures, and have resistance to cracking or spallation under mechanical and thermal stresses induced during operation. They should offer resistance to developing fissures to preserve coating strength when under the influence of thermal and mechanical stresses. The size and the shape of the material to be coated are crucial in order to select the most suitable deposition technique. Coatings can be tailored for a specific application by controlling their elemental composition, their microstructure, and by selection of manufacturing process.

Regarding the elemental composition, only limited materials behave in a satisfactory manner in molten nitrate salt at temperatures above 550°C Table 1 [12]. It was reported that refractory metals such as cobalt and nickel, and alloys like NiMo, TiAl, austenitic stainless steel, and Ni-Cr-Fe corroded rapidly due to rapid oxidation and/or dissolution in the molten salt in a study with several metals, alloys and ceramics [13]. It was hypothesised that coatings must include a high concentration of elements such as Al, Cr, and Si which tend to form a protective scale [14].

3. Compatible materials for structural components

The suppression of hot corrosion is a major technical challenge in materials selection and structure design for TES. High temperature nickel-based alloys are the logical choice for mechanical strength, oxidation and corrosion resistance, as corrosion kinetics increase with elevated temperatures, however, the cost of nickel-based alloys are nearly four times more expensive than iron-based steels [15,16]. The corrosion behaviour of carbon steel A36 at 316°C has been studied and weight losses were found to be modest with corrosion rates of about 5 $\mu\text{m}/\text{year}$, implying they can be used in the cold parts of the plant [17]. The study by Sandia entailed a review of problems and lessons learned from operation of the Solar Two power tower after its demolition in 2009, including corrosion rates for two Fe-Cr-Ni alloys (SS321 and SS34) and two nickel alloys (HA230 and In625) in contact with solar salt at 400, 500, 600 and 680°C [15].

Corrosion is insignificant at 400°C for all the studied alloys. Material performance is also excellent at 500°C. At this temperature, both nickel alloys form protective nickel oxide, which does well in protecting the base alloy. A different scenario is observed at 680°C. Corrosion at this temperature is severe for all the alloys with metal losses greater than 450 $\mu\text{m}/\text{year}$. The corrosion resistance of steels under these conditions depends on the formation of a protective oxide scale rich in Cr, which is similar to what happens during oxidation in high temperature gaseous atmospheres. However, an important difference when using molten salt is that chromium compounds are soluble in nitrate salt and prevents the formation of a protective oxide scale (passivation) [18]. This results in non-protective and/or fast-growing oxide formation and in the increment of material degradation due to higher corrosion rates [19].

Alloys constituent such as Cr, W and Mo can produce soluble anions and can be readily extracted from the surface oxide scales formed on the alloys [20]. HR120 and Mo332 exhibited corrosion product with similar concentrations of Ni, Fe, and Cr, while HR224 showed partial surface oxidation after 3000 hours, which was attributed to the high concentration of Al [17]. Alloys with stabilizing additions of niobium, such as SS347 or titanium, for instance SS321, are known to diminish stress corrosion cracking. The corrosion rate of SS347 has been reported to be smaller than SS321 by 30-40% for temperature up to 600°C, spallation, on surface of SS321, could be the reason[15]. The corrosion rates at 600°C have been observed to be twice compared to 500°C, as reaction kinetics usually display an Arrhenius relationship. The corrosion rates at 680°C have been reported to be two orders of magnitude larger than at 600°C [15]. The corrosion rates of 8.6 and 9.0 $\mu\text{m}/\text{yr}$ have been reported for AISI316L and AISI321H austenitic stainless steel, upon immersion in solar salt at 550°C [21]. Corrosion rate of stainless steel in molten salt is reported to speed up with the rise in temperature, the composition of oxide scales and the corrosion mechanism change with different temperatures [22].

The effect of impurities present in molten nitrate salts has been investigated in A36 and A516 at 316°C [23],[24]. No significant difference in corrosion rate was observed in A36 with an addition of up to 1.3% chloride at 316°C [23]. The effect of chloride becomes more substantial after 1000-hour exposure to the salts. After exposure to NaNO₃-KNO₃-NaNO₂, stainless steel was proposed to be better compared to carbon steel for corrosion resistance at higher temperature [25]. IN625 has been stated to have better performance in molten nitrate salts compared to other studied ferrite steel and stainless steels [26]. Stainless steels are described to behave better in molten nitrate salts as compared to ferritic steels [27].

Table 1. Beneficial and detrimental aspects of elemental constituents of alloys [12].

Elemental Constituent	Beneficial Aspects	Detrimental Aspects
Ni	Provides strength.	Prone to destructive interaction with sulphur.
Co	Provides microstructural stability and strength.	Prone to destructive interaction with sulphur.
Al	Major contributor to providing strength Contributes to oxidation resistance	Large concentration lowers melting point.
Cr	Contributes to oxidation resistance to 816°C. Reduces Al requirement for formation of alumina scale.	Lowers creep strength.
Ta	Enhances hot corrosion and oxidation resistance. Improves strength.	
Si	Enhances oxidation and hot corrosion resistance.	Large concentration leads to formation of brittle phases.
Hf, Y, Y2O3	Improve adherence of alumina and chromia scales.	Large amounts are detrimental.
Pt	Improves oxidation and hot corrosion resistance.	

3.1 Aluminide coatings

Aluminide coatings have been used widely and shown to be beneficial for corrosion protection in molten nitrate as well as other molten salts [28][29][30]. The aluminide has been reported to form a thin corrosion resistant layer of LiAlO₂ upon reaction with Li in molten K/Na/Li carbonates at 650°C [31].

A minimum of 25 (at%) of aluminium in FeAl intermetallic has been reported as required to develop a continuous LiAlO₂ oxide layer in Li/K carbonate melt at 650°C [32]. Slurry aluminide coatings, a low-cost option, has been reported to develop a thin NaAlO₂ layer in molten Solar salt at 600°C and 580°C, although the protection mechanisms still not well identified [19][33][29]. Aluminides have a great amount of interest for coating material as they provide corrosion resistance up to temperatures above their mechanical strength limit. Aluminides with appropriate quantities of aluminium forms alumina scales. They possess low densities, high melting points, good thermal conductivities, and excellent high temperature strengths.

3.2 Ni-based coatings

Performance of pure nickel coatings is very poor, but Ni-based coatings have been reportedly used to provide wear, oxidation or hot corrosion resistance [34]. Intermetallic are proposed for high temperature use because their strength increases with temperature. Therefore, the use of an intermetallic material which forms an alumina scale, with nickel as base material is very encouraging. Ni-based alloys possess higher resistance to molten nitrate corrosion compared

to Fe-base alloys, however considerably costly. The corrosion resistance of Ni-based alloys in molten nitrate-nitrite salt, has been reported to improve with Ni concentration in these alloys [18][35]. Ni based intermetallic and coatings have been explored to ease molten salt corrosion.

3.3 Nickel Chrome coatings

Nickel chrome coatings have been found to present protection against spalling and sputtering in aggressive environment of sulphate and vanadate salts at 900°C [36][37]. High chromium alloyed steels have demonstrated higher corrosion protection via formation of a slow growing continuous chromium oxide or chromate spinel layer [27]. However, solubility of Cr in the nitrate salt leads to depletion of chromium in the substrates and Cr-dissolution in molten salt [13][38]. Therefore, it is expected that materials with a significant amount of chromium will perform poorly in contact with nitrate salt.

3.4 Nickel Aluminide coatings

Nickel aluminide is proposed as a very promising material to be used as anticorrosion coating for molten nitrate salts. Favourable physical and mechanical characteristics, such as high melting temperature, high thermal conductivity, attractive stiffness, good oxidation resistance and metal-like electric conductivity [16]. Nickel aluminium coatings are reported in a number of cases with different compositions and have been seen to enhance the oxidation resistance of the substrate material. Nickel aluminium coatings are suggested to have a strong grip to substrate, offering strong adhesion and reducing the thermal mismatch between substrate material and coatings, in case of nickel or iron-based alloy substrate [39]. This type of coating has been seen to have the ability to form protective aluminium-oxide layers, resulting in exceptional oxidation resistance, which prevents further diffusion of reactants into the substrate material [40].

Greater amount of aluminium increases high temperature oxidation resistance, but it compromises the creep strength and load-bearing capability, so it is kept to less than 6% level [41]. Tortorelli observed that aluminium content must be over 30-35 at% threshold to provide protection against nitrate salts, however higher concentrations of aluminium constraint ductility and fabricability [13]. Therefore, the value of corrosion potential will be influenced by the stoichiometry of coatings and a careful control of the elemental composition will be crucial. Several studies have showed the promising properties of this alloy as anticorrosion coating [42]. Five intermetallic phases formed with nickel aluminide are NiAl_3 , Ni_2Al_3 , NiAl , Ni_5Al_3 and Ni_3Al [43][44].

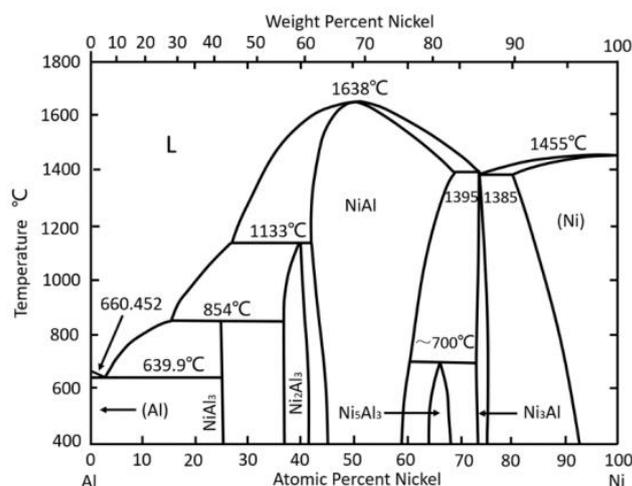


Figure 2. Binary phase diagram of nickel aluminium system [44]

One of the causes of coating failure might be differences in thermal expansion coefficient among coating and substrate material. Bond coating of some material can be used for overcoming this mismatch. Applying a NiCrAlY bond coat prior to nickel aluminide coatings to increase the bond strength and decrease thermal expansion coefficient mismatch, provides good protection to the substrate in air and molten salt environments at 900°C [45]. It has been reported that addition of rare earth metal as for example yttrium can increase the strength of the coating and also enhances nucleation of Al₂O₃ growth during the high temperature oxidation tests [16]. Formation of NiO, NiAl₂O₄, and Al₂O₃ oxides on Ni₅Al coating at 900°C for 100 cycles provided better hot corrosion resistance in comparison with bare super alloy [46][47]. Ni₃Al coatings have shown a reasonable corrosion rate in NaNO₃-(KNO₃)-Na₂O₂ environments while NiAl functioned quite well [13].

Electrodeposited slurry aluminide and nickel-aluminide coatings have been found to perform better than uncoated material, for corrosion resistance in solar salt at 580°C. Ni₃Al coatings are much preferred to be used as corrosion resistant coatings as they possess strength at raised temperature, oxidation protection and creep properties. Ni₃Al coatings on stainless steel substrates have been found to be protective in the presence of NaNO₃:KNO₃ salt environments, with the potential to extend the lifetime of components such as the storage tank systems [48]. Laser re-melting of Ni₃Al coating has also been observed to enhance the hot corrosion resistance [49].

3.5 Cost estimation

Material cost calculated for a molten salt container of size ~35m diameter, ~12.5m height (30 mm thick wall) made with SS347 costs ~€ 3.5M, while for HA214 ~€ 8.6M and ~€ 15.8M for HA214 [50]. Total cost of same size container including 30mm SS347 and 100 µm thick, air plasma spray deposited Ni₃Al coatings with 25% coating efficiency was given as ~€ 4M, implying Ni₃Al coatings enable low manufacturing cost with accommodated wall thickness [50].

4. Conclusion and outlook

The main conclusion of this literature review is that the suppression of corrosion in TES is a challenging task. The different solar thermal technologies have been reviewed, leading to the conclusion that CSP coupled with molten salt heat transfer fluids represents the perfect combination of high efficiency and heat storage capability. However, to make the most of this combination it is crucial to reduce hot corrosion by molten salt in component systems. The literature review revealed that there is still a lot to be done in order to find the ultimate solution to this problem. There is much to learn from other technologies in the field of hot corrosion.

Among the molten salts used as TES and HTF, Solar Salt NaNO₃ (60wt%) KNO₃ (40wt%) and related compositions are the most employed till now because of its low melting point and low cost. There is a need to study anticorrosion coating to be used with Solar Salt (NaNO₃ (60wt%), KNO₃ (40wt%)) for thermal energy storage components. In contrast to system components of other technologies dealing with hot corrosion, recoating of molten salt storage and distribution components is not straight-forward. Therefore, it is crucial that the coating survive many thermal cycles and performs its protective role for many years during plant operation. It would be interesting to see if future work focuses on different coating deposition techniques. Corrosion tests in molten salt and oxidation tests in air should be carried out to understand the underlying mechanisms and to distinguish between oxidation and corrosion results. Field tests should be carried out to observe the coatings in the real environment with fluctuating solar flux, wind and dust, compared to the steady temperatures in laboratory tests.

In conclusion this review document shows the potential for Plasma sprayed Ni₃Al protective coating on SS347 to suppress corrosion and defines the future directions in this field.

However more research is needed for investigating the Ni₃Al coatings on other substrate materials.

Data availability statement

The authors confirm that the data supporting the findings of this study are available within the article. Additional information on data can be made available upon reasonable request from the corresponding author.

Author contributions

Sarah Yasir: Methodology, investigation, visualization, writing—original draft, data curation, formal analysis.

Adrianus Indrat Aria: Methodology, conceptualization, validation, visualization, supervision, formal analysis, writing — review and editing.

Chris Sansom: writing — validation, review, and editing.

Competing interests

The authors declare that they have no competing interests.

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