E.3. Corrosion Allowances for Sodium Heated Steam Generators - Evaluation of Effects and Extrapolation to Component Life Time

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Abstract:

Steam generator tubes are subjected to two categories of corrosion; metal/sodium reactions and metal/water-steam interactions. Referring to these environmental conditions the relevant parameters are discussed. The influences of these parameters on the sodium corrosion and water/steam-reactions are evaluated. Extrapolations of corrosion values to steam generator design conditions are performed and discussed in detail.

1. Introduction

In setting corrosion rates for sodium heated steam generators, scatter of available experimental data and uncertainties in extrapolation of these data to component life-time result in improper design allowances, i. e. too small or too large values.
As a consequence corrosion allowances which are too large result in increased heat transfer surfaces, larger unit sizes and higher costs; a corrosion allowance too small to meet the operational requirements endangers the component life-time.
The present paper intends to give a review of those influences which result from the interaction of the steam generator tubing with the sodium and the water/steam; the available experimental background will be discussed and parameters not satisfactorily investigated will be pointedout; furthermore an extrapolation of corrosion allowances to the component design time will be given.

2. Environmental Interactions of Steam Generator Tubes

Referring to the environmental conditions to which the steam generator tube material is subjected, two categories of influences are to be discussed (Fig. 1).

The sodium/metal reactions are known to result in a general sodium corrosion, parameters of which are temperature, sodium velocity, oxygen content of the sodium etc. This general corrosion produces material loss or deposition depending on the position of the metal in the system.

The general sodium corrosion is accompanied by selective corrosion processes which give rise to an attack of specific substitutional and interstitial elemants (e.g. carbon and nitrogen transfer); this selective corrosion as well as the secondary combined effects which follow from a mechanical interaction of materials in sodium, e. g. friction and wear which could occur between the steam generator tube and the spacer are not treated within the scope this paper.

With regard to its position in the secondary sodium system, the steam generator - like the intermediate heat exchanger in the primary system - is a component whereby sodium/metal interaction metal loss and corrosion product deposition is expected to occur. While metal loss in the sodium inlet regions affects the mechanical integrity of the steam generator tube during life time, deposition of corrosion products in the areas of lower temperatures affects the heat transfer characteristics.

The chemical reactions between water/steam and metal form as a corrosion product a magnetite layer. This corrosion process is, in principle, a desired reaction; the magnetite, provided it remains intact in operation, represents a relatively dense and adherent layer at the surface which protects the material surface from further rapid corrosive attack.
The properties of this magnetite layer are reported to be strongly dependent on the operational

conditions of the steam generator; the corrosion rate under isothermal conditions has been observed to be lower than that at surfaces of high heat-transfer conditions; start-up and shutdown cycles during operation enhance corrosion rates; improperly selected water velocities in the evaporator sections of boilers are expected to produce a rippled magnetite structure which results in an increased pressure drop; chemical cleaning to remove the rippled structure would become necessery as a consequence.

2.1. Metal/sodium reactions

As was pointed out above, the steam generator unit is situated in a downstream position in the secondary sodium system where both metal loss and corrosion product deposition could be expected to occur. Whether or not the steam generator in fact is a component where a cross-over point from corrosion of metal to deposition of corrosion products happens has been examined by a literature review on corrosion behavior of steels in downstream positions of dynamic sodium systems.
Figure 2 mainly illustrates the results of sodium corrosion investigations [1 - 4] which were obtained from the downstream positions of experiments simulating the temperature sequence in a primary sodium system (700°C - cladding hot spot temperature; 400°C - IHX temperature). The figure correlates the normalized corrosion/deposition values with the temperatures downstream to a maximum temperature of 700°C. According to these tests a crossover point from corrosion to deposition would occur at a temperature of about 680°C. For temperatures which are more realistic for a secondary sodium system, i. e. the range from about 600°C down to 400°C, PNC [5] reports an experimentally-determined cross-over point of about 570°C. Additional information for this temprature range is available from the operation of a dynamic sodium loop at INTERATOM which was built as a mock-up loop of the primary sodium system of the SNR 300. The primary aim of this loop concept is to study the distribution of activated corrosion products under the specific conditions which result from the temperature distribution, geometrical parameters, coolant velocities and residence times etc. of the SNR primary sodium circuit [2]. As a part of these experiments sodium corrosion data from specimens distributed along the circuit were obtained.
The results of the weight change determinations of the specimens in the cooling section of this loop, in Figure 3, demonstrate a very marked change from material loss to weight gain mear the point where a first decrease in temperature occurs (transition - temperature about 580°C). These experiments show that one conservatively can treat a steam generator tube as having two regions, a metal loss region and a deposition area.

From the point of view of design, this means that in order to ensure a reliable mechanical integrity at the design temperature of the steam generator tubes a corrosion allowance has to be made to compensate for wall thickness reduction. A compilation of corrosion rates for austenitic stainless steels, supplemented by data for ferritic steels which are of main interest for this paper [Table 1] is provided by Figure 4. From this figure results a mean corrosion rate for a design temperature of 530°C and 200,000 hrs (22.8 yrs) of 55 µm (79 µm for upper 1 σ - limit).

Table 2 enlists data from the literature and from own tests on the deposition of corrosion products in the cooling areas of sodium systems which impedes heat transfer.

This table gives the temperature range of the cooling section and, as far as documented, the temperature of maximum deposit.

The deposition rates arising from these different investigations differ by about a factor of 20. If one does not regard the data which is obtained from tests with temperature ranges in the cooling loop section from 650 to 480°C, i. e. from conditions that are not relevant to secondary sodium systems, the remaining results provide deposit data between 25 and 70 μm in a 22.8 years time.

These values may be somewhat misleading because of the differences in their generation. As shown in Table 2, the value of 25 μm has been determined by weight gain measurements which were then converted to a layer thickness by using the density of 7.85 g/cm^3; this means that the value of 25 μm stands for a metallic layer; on the other hand ref. [10] gives a value of 40 μm which represents a deposit layer thickness, i. e. a layer of carbides, oxides etc.; this kind of layer would have a different conductivity than the first one. The PNC-data on deposits are given in the literature [5] as thickness dimensions. It is not known what density was used to achieve this layer thickness.

Also uncertain are the differences between the loop systems from which the data are obtained; differences in parameters like temperature levels, surface ratios of corroding and depositing areas, velocity of sodium, sodium chemistry etc. are certain to influence the extent of depositions.

Ray [11] has reported on experimental determinations of a marked heat transfer degradation in sodium/sodium heat exchangers due to the deposition of corrosion products. These results show that in order to overcome a decrease of heat transfer performance as a result of deposition of corrosion products an allowance in heat transfer areas should be taken into account.

2.2 Metal/Water Reactions

Possible reactions and resulting consequences from the interactions of the metal with water and steam have been shown in Figure 1. The further discussion of phenomena is directed towards low-alloyed ferritic steels for evaporator and superheater purposes.

2.2.1 Evaporator

Corrosion allowances for the evaporator of a steam generator do not need a very extensive discussion. The water-side corrosion allowance for the 2.25 Cr 1 Mo type of steel at a temperature of 357°C (maximum straight-tube steam generator temperature of SNR 300) is about 25 to 35 μm [13] for 200,000 hrs service time. Necessary pre-sumptions for this value are that the corrosion occurs uniformly under proper water chemistry conditions and that during standby periods the magnetite layer is shielded against localized corrosion effects like pitting due to oxygen corrosion. An increase of pressure drop in the evaporator section of the steam generator has been reported in the literature [13,14]. This pressure drop increase has been correlated with the formation of a magnetite layer with a rippled surface structure which in turn depends on the water velocity. This rippled structure has only been observed in the water phase when water velocities exceed a limiting range of 1.6 to 2.9 m/s. The water velocity in the straight-tube steam generator for SNR 300 is at the lower edge of this range (1.7 m/s) and is therefore believed not likely to cause troubles.

2.2.2 Superheater

Due to the fact that tubes in conventional steam generators are subjected to rather high combustion-side corrosion rates, steam-side corrosion rates have not been regarded as intensively as in sodium heated steam generators; under the conditions of the sodium-heated steam generator, however, steam-side corrosion behavior of steel is a marked percentage of the tube wall thickness.

Table 3 gives a survey on some corrosion rate data obtained from a literature review on corrosion behavior of ferritic steels under superheater conditions. This table is divided into three parts; both the upper parts compile data from laboratory-scale tests and from the post-examination of conventional steam generator tubes, respectively. The results at the bottom of this table were obtained from the post-operation examination of sodium-heated steam generators. The test temperatures range between 480 and about 600°C; test times are mainly, except the sodium-heated steam generators, between 6,000 and 200,000 hrs. The laboratory-scale values were obtained from isothermally heated specimens, whereas the results from the examination of actual steam generator tubes were from material that was subjected to heat transfer conditions [10 - 100 KW/m^2].

The determination of the magnetite layer thickness was performed either by descaling, by weight gain measurements or by direct determination of the oxide layer thickness.

For sodium-heated superheater design temperatures, i. e. 490 to 530°C, the extrapolation of the experimental results to 22.8 yrs design time by using a parabolic law gives metal loss values of between 60 and 240 µm. The data from sodium-heated steam generators, so far available, fall within the spread of the values.

It has been shown that the mechanism of magnetite formation follows a parabolic law:

$$d^X = K \cdot t \qquad (X = 2), \qquad [1]$$

where by

$$k = k' \cdot e^{-\frac{Q}{RT}} \qquad [2]$$

the temperature dependence is given (d = thickness [mm], t = time [hrs], k = oxidation constant, T = temperature [K], Q = activation energy).

Based upon this parabolic law, Figure 5 illustrates the temperature dependence of the metal loss, normalized to 200,000 hrs for a total number of 66 values. This figure reveals the scatter of the values at the different temperatures and shows a change in the slope of the curve which occurs at 580°C. This transition temperature is due to the change of magnetite Fe_3O_4 to FeO at higher temperatures. The values from sodium-heated steam generators are at the upper and lower ends of the scatter band.

From regression analysis, the following equations were obtained for the two temperature regions of different slope:

<u>T > 853 K (> 580°C):</u>

$$\ln d_{22.8\ yrs} = 22.623 - \frac{20{,}239}{T} \pm 0.2376 \ (\pm 1\sigma) \qquad [3]$$

<u>T < 853 K (< 580°C):</u>

$$\ln d_{22.8\ yrs} = 7.389 - \frac{7{,}423}{T} \pm 0.3046 \ (\pm 1\sigma) \qquad [4]$$

For the SNR 300 superheater design temperature of 528°C a metal loss in 22.8 yrs of d = 0,15 mm results from equation (4).

Deviations from the exponent 2 in equation (1) due to changes in diffusion processes have been observed. Figure 6 gives the depth of metal loss from steam generator tubes which have been operated for up to 150,000 hrs [18]. This figure shows that the regression line does not meet the parabolic law. Especially the long-time tests at the temperatures 530°C and 575°C show an exponent larger than 2.

In order to see how far the experimental values as given in figure 5 deviate from a parabolic law a regression analysis of all data as a function of temperature and time was performed. This regression analysis was based upon the general function:

$$\ln d = \frac{1}{X} \quad (\ln k + \ln t)$$

$$\ln d = \frac{1}{X} \quad (A + \frac{B}{T} + \ln t).$$

For the temperatures below and above 580°C, values for x of 2.77 and 2.33 were found, respectively. The resulting metal loss for 200.000 hrs is given by the equations [5] and [6].

<u>T > 853 K (> 580°C):</u>

$$\ln d_{22.8\ yrs} = 22.75 - \frac{20{,}570}{T} \pm 0.2310 \quad (\pm 1\sigma) \qquad [5]$$

<u>T > 853 K (< 580°C):</u>

$$\ln d_{22.8\ yrs} = 9.152 - \frac{9{,}113}{T} \pm 0.2574 \quad (\pm 1\sigma) \qquad [6]$$

All the data were extrapolated with the new time-relationship to 200,000 hrs and are shown in figure 7 together with the regression lines; for the design temperature of 528°C, results a metal loss of $d_{22.8\ yrs} = 0.11$ mm.

Superheater corrosion rates discussed so far have been mainly obtained from experiments under isothermal steam corrosion conditions; in addition to that, the extrapolation of the corrosion depth for the service time was based upon the assumption of a uniform magnetite growth without any spalling with time.

For a number of alloys, experimental investigations have shown that the corrosion rate under heat transfer could be considerably higher than under isothermal conditions [15, 16].

The extrapolation of the isothermal corrosion data for Incoloy 800 for three years of exposure in steam of 565°C is about one-half of the corrosion when tested under heat transfer (Table 4); a comparison for 620°C reveals nearly the same relationship. Comparable results were obtained for Inconel 625. On the other hand, a recent publication on this phenomenom did not verify these findings [16].

The alloys investigated (Table 4), did not include ferritic alloys; a correction factor to be applied on low-alloyed ferritic steels to take into account the increased corrosion rate under heat transfer is therefore not available from these experiments.

On the other side, the experimental values obtained from tubes of long-term actual steam generator operation under heat transfer [18, 25] and limited number of data from sodium-heated steam generators [17, 19] are located within the scatter based on isothermal test data.

As was pointed out before, the considerations of corrosion rates for a service time of 22.8 yrs were based on a uniform magnetite growth without any interruption. If it would become necessary to descale the steam generator unit due to a pressure drop increase the magnetite layer would be removed, and the oxidation mechanism has to be started again.

Depending on the time intervals between repeated descaling processes the metal loss consumed by the magnetite formation would be increased.

3. Discussion

The sodium corrosion behavior of low-alloyed ferritic steels for steam generator purposes is based on a number of values which were obtained at different temperatures and sodium conditions. Due to their principal similar behaviour, the data of austenitic stainless steels do corroborate these results. Curves resulting from regression analysis of the available data of austenitic stainless steels are in good agreement with the data for ferritic steels and can be used for a calculation of the corrosion rate in sodium. From this regression analysis, which is valid for sodium velocities of 3 - 10 m/s and an average oxygen content of about 10 ppm, a value of 55 µm (79 µm for 1σ) for linear time extrapolation is established for 22.8 yrs service time.

Beside the metal loss by sodium corrosion, a deposition of corrosion products occurs at lower temperatures due to a supersaturation process. This process is strongly dependant on parameters like geometry of the system, thermohydraulic characteristics, temperature conditions etc. For temperature conditions which are relevant for a secondary sodium system the corrosion deposit thickness has been linearly extrapolated to values between 25 and 70 µm for 200,000 hrs.

How the main parameters which were discussed to be of influence on the steam corrosion could affect the metal loss of the steam generator is illustrated in Figure 8. This figure demonstrates the metal loss of ferritic low-alloyed steels under uniform isothermal steam corrosion at 528°C, and in addition to that, by heat transfer and repeated descaling.
In this diagram the solid (No. 1) and dotted lines (No. 2) represent the metal loss with time for an isothermal oxidation at 528°C with a time exponent (equation 6) of 1/2.77 as average and upper 1 σ - values. If heat transfer is taken into account by applying a factor of 1.5 on the mean and upper 1σ isothermal oxidation, respectively, curves 3 and 4 are obtained. Curves No. 5 to 8 give the metal loss if the lines 1 to 4 were corrected for repeated chemical cleaning, e. g. every 40,000 hrs.

The values obtained for a 200,000 hrs service time vary between 0.11 mm and 0.6 mm, depending on what kind of consideration has been taken. This example shows that the metal loss is mainly influenced by the number of cleaning processes; repeated chemical cleaning, e. g. four times, raises the metal loss by a factor of 3.

As was pointed out before, contradictory results have been obtained with regard to the influence of heat transfer on the steam corrosion. A factor in maximum of 2 on corrosion rate of austenitic steels was measured at heat fluxes of about 500 KW/m^2 [15]. At a higher heat flux of 2000 KW/m^2 [16] no influence could be established. For ferritic materials no systematic relevant investigations are reported. But from references [18, 25] and [17, 19] which represent data from conventional and sodium-heated steam generators, respectively, no influences of heat flux in the range of 10 to 100 KW/m^2 could be derived. Furthermore, it should be considered that the maxima of heat transfer and temperature do not coincide; at the exit of the superheater, i. e. at the maximum temperature, a heat flux of about 70 - 100 KW/m^2 is reached whereas the maximum heat flux of about 400 KW/m^2 is obtained at the superheater inlet (360°C). It therefore seems reasonable not to take into account the heat transfer as an additional parameter for the corrosion allowance at the highest temperature.

These considerations were based on the assumption of a uniform oxidation mechanism and the application of regression analysis to existing data (equation 6). From this regression analysis the time exponent was found to be 1/2.77 instead of 1/2.0. This gives a metal loss which is about 30 % lower than on the basis of an exponent 1/2.0.

A consequent application of conservative considerations, i. e. a factor of 2 for heat transfer conditions, the assumption of a parabolic law, use of the upper 1σ - limit would lead to a value for the metal loss of 0.9 mm/200,000 hrs.

A realistic evaluation, which does not blindly accumulate all the uncertainty factors, should be based upon the time- and temperature-dependant relationship in equation (6). An additional allowance for the heat-transfer should not be required, because the major part of the evaluated values were obtained already from heat transfer conditions similar to sodium-heated steam generators and do not lie preferentially in the upper scatter band.

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SODIUM CORROSION OF FERRITIC ALLOYS IN AUSTENITIC SS LOOPS

Material	Temp. (°C)	O_{Na} (ppm)	V_{Na} (m/s)	Time (hrs)	R (μm/a)	Ref.
2,25 Cr- Nb	475	8	0,4	10,000	0,9	[6]
2,25Cr-Ni,Nb	600	5	5	5,000	2,0	[7]
2,25 Cr	600	1	5	3,000	2,2	[8]
2,25Cr-Nb,Ti	600	25	10	14,000	33,0	[9]
2,25Cr	650	8	2	4,500	2,3	[6]
2,25Cr-Nb,Ti	650	8	10	*	7,1	[9]
2,25Cr-Nb,Ti	650	25	10	*	71,0	[9]

* values from regression analysis

Table 1

DEPOSITION OF CORROSION PRODUCTS IN COOLING SECTIONS OF SODIUM LOOP SYSTEMS

TEMPERATURE (°C)		DEPOSIT (μm/a)			MAX. DEP. (μm/ 22,8a)	REF.
RANGE	MAX. DEP.	MIN.	AV.	MAX.		
650-550	(565)	2,2	24,5	-	560	[11] WARD *
650-480	593	-	1,4	5,5	125	[12] GE
~550	-	-	-	-	~40	[10] UK
600-350	530/400	-	0,2	3	68	[5] PNC
600-380	500	-	-	1,1	25	[Fig.3] IA *

* calculated from mg/cm^2h by $\gamma = 7{,}85 g/cm^3$

Table 2

CORROSION RATES OF 2,25 Cr-1Mo IN STEAM

Base	Temp. (°C)	Time (hrs)	HeatFlux (KW/m^2)	Metal Loss* (μm/ 22,8yrs)	Measurement	Ref.
laboratory scale	593	15,000	-	526	descaling	[20]
	593	17,000	-	461	?	[21]
	480	6,000	-	112	weight	[22]
	500	7,000	-	105	descaling	[23]
	510	7,000	-	239	descaling	[24]
conv. SG	490	200,000	10-100	63	oxide thickness	[18]
	530	200,000	"	134		[18]
	515	26,000	"	96		[25]
Na SG	497	4,000	-50	116		[17]
	510	3,100	-15	60		[19]

* Extrapolation based on parabolic law

Table 3

STEAM CORROSION UNDER ISOTHERMAL AND HEAT TRANSFER CONDITIONS

MATERIAL	HEAT FL. (KW/m^2)	ISOTHERM. (μm/3a)			HEAT TRANSF. (μm/3a)			Ref.
		565°C	600°C	620°C	565°C	600°C	620°C	
Incol. 800	400/	3.8	-	10	6.2	-	19.5	[15]
Incon. 625	550	3	-	3.5	5.2	-	9.5	
Incol. 800	2000	-	17	-	-	17	-	[16]

Table 4

ENVIRONMENTAL INTERACTIONS OF STEAM GENERATOR TUBE (CHEMICAL REACTIONS UNDER NORMAL CONDITIONS)

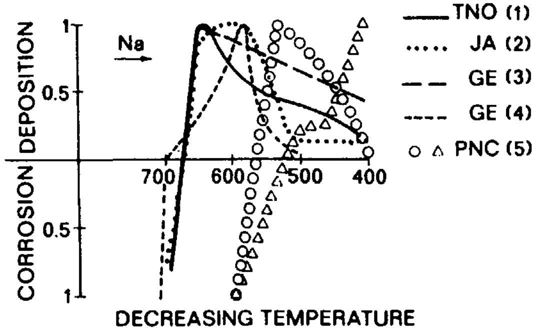
● WITH SODIUM

EFFECT	CONSEQUENCE
- CORROSION	
- UNIFORM CORROSION	- TUBE WALL THICKNESS REDUCTION
- SELECTIVE CORROSION	- CHANGE OF MECHANICAL PROPERTIES
- DEPOSITION OF CORROSION PRODUCTS	- ALTERNATION OF HEAT TRANSFER CHARACTERISTICS

● WITH WATER/STEAM

EFFECT	CONSEQUENCE
- CORROSION	- METAL LOSS, TUBE WALL THICKNESS REDUCTION
	- RIPPLED MAGNETITE, PRESSURE DROP INCREASE AND CHEMICAL CLEANING

Fig. 1



NORMALIZED DEPOSITION VERS. LOOP TEMPERATURE

Fig. 2

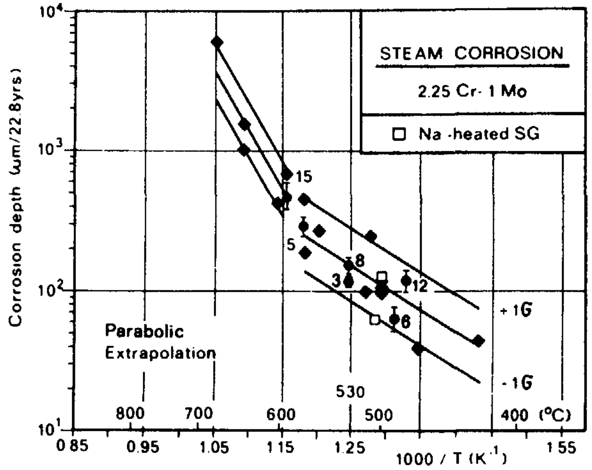


Fig. 5

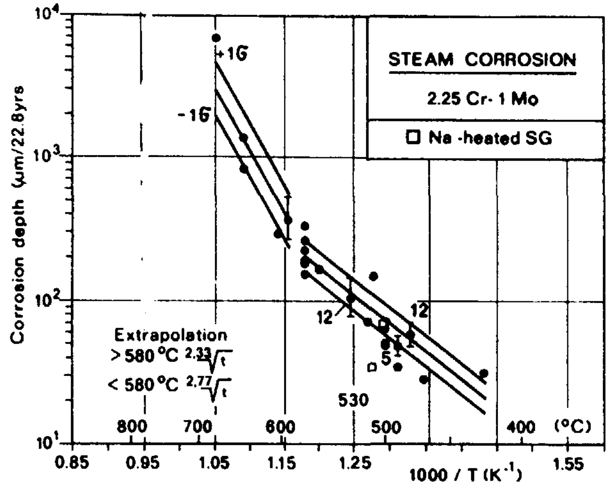
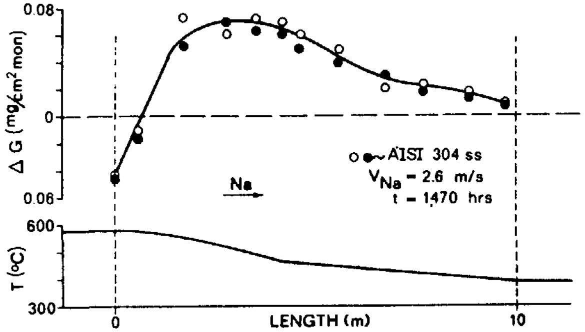
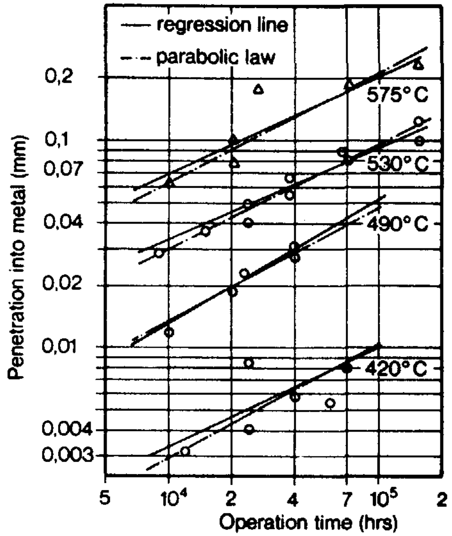


Fig. 7



CORROSION/ DEPOSITION IN COOLING SECTION OF SODIUM SYSTEM

Fig. 3



CORROSION OF 2,25 Cr-1 Mo AND 1 Cr-0,4 Mo IN STEAM

Fig. 6

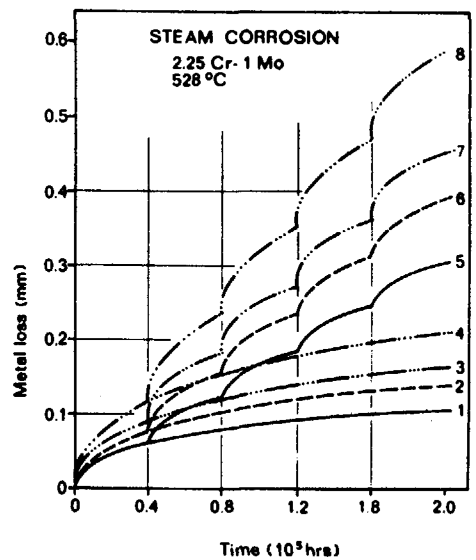


Fig. 8

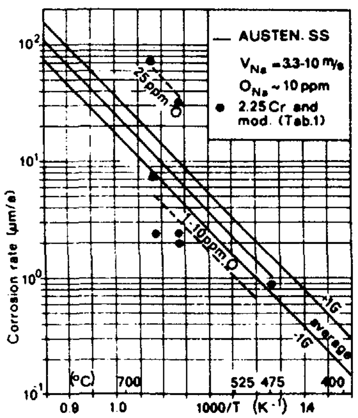


Fig. 4

CORROSION RATE OF FERRITIC AND AUSTENITIC STEELS IN SODIUM