WARD-3762-9

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Corrosion Analysis

Low-Capacity Pump

Large Sodium Pump Study-Phase II

Prepared for the New York Operations Office
U. S. Atomic Energy Commission
Under AEC Contract AT(30-1)-3762

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ABSTRACT

An evaluation was made of the effect of sodium on the integrity of the pump materials identified in the Large Pump Study Phase I. There were type 304 stainless steel as the constructional material and Stellites 1, 3 and Star J as potential bearing materials. These materials have been evaluated for use under expected pump operating conditions. Relevant data on sodium corrosion, the effect of sodium on mechanical properties and the effect of temperature on mechanical properties have been reviewed and corrosion losses have been extended for Type 304 stainless steel. Corrosion and wear data have been reviewed for the bearing materials.

It is concluded that no significant materials problems should arise for an operating temperature of 900°F but that confirmatory data are required in the areas of mechanical properties in sodium, mass transfer at high velocities and wear of bearing materials under operating conditions

INTRODUCTION

The study was one of six side studies performed under the low-capacity study of Phase II of the Large Sodium Pump Study. The side studies are:

Extrapolation of Hallam and Fermi Sodium Pumps to the FFTF Primary Pump Reference Design Requirements	WARD-3762-5
Drive Side Study	WARD-3762-6
Dynamics Side Study	WARD-3762-7
Radioactive Shielding Requirements	WARD-3762-8
Plant Site Pump Handling Study	WARD-3762-10

The objective of this study was to review the available data on the corrosion of materials in flowing sodium in relation to their use for pump application.

REVIEW OF CORROSION DATA ON AUSTENITIC STAINLESS STEELS

Much of the early liquid-metal data cannot be applied to the evaluation of materials for nuclear systems because the data were generated in static capsules and low-velocity thermal convection loops. Oxygen levels in these tests were either not clearly defined or were too high to be of value. Data obtained in pumped loops indicated that corrosion rates are strongly dependent on loop operating parameters such as sodium velocity, temperature differences in the system, chemistry gradients (different materials in different parts of the system), and system geometry, as well as on the local temperature and oxygen content of the sodium.

Of the corrosion data generated to date, the only applicable data are those from the UKAEA^[1] and G.E.^[2] programs. The test parameters are summarized in Table 1. In addition to the austenitic stainless steels listed, various ferritic steels ^[1, 2], and high-nickel ^[1] alloys were studied.

Summary

The principal observations were:

- a. Corrosion occurs in the hot-leg regions of pumped-loop systems, and deposition occurs in cold-leg regions.
- b. The corrosion rates for all low-nickel alloys are similar.
- c. After an initial period of a few hundred hours, the corrosion rates became linear. The UKAEA work^[1] also reported an initial small weight increase.
- d. An increase in test temperature caused an increase in corrosion rate, corresponding to an Arrhenius-type relationship.
- e. An empirical equation was developed for the G.E. data and is given as:
$$R = V^{0.884} O_x^{1.156} \exp \left[\left(12.845 - \left(\frac{23,827}{T+460} \right) - \left(0.00676 \frac{L}{D_1} \right) + \frac{2.26}{t+1} \right) \right] \quad (1)$$

where:

R = average rate of removal (mg/dm²/m to time t)

O_x = oxide level (ppm)

V = sodium velocity (ft/sec)

T = sample temperature (°F)

L/D₁ = downstream factor (number of diameters a sample is downstream in an isothermal region)

t = exposure time (months)

- f. A 'downstream effect' was observed in both programs ^[1,2]. Specimens at the inlet end of the hot leg corroded at a higher rate than specimens further down stream due, presumably, to progressive saturation of the sodium with corrosion products.

Table 1

Test Parameters

Ref.	Aust. Steels Tested	Temp. Range (°F)	ΔT (°F)	O ₂ Content (ppm)	Velocity (ft/sec)
UKAEA [1]	Type 321 Type 316 Type 316L Type M316L Type M316 FV548	1120-1300	—	Up to 30	0.5-40
G.E. [2]	Type 316	1100-1200	500 & 250	12 & 50	Up to 25

- g. The UKAEA work indicated that above about 10 to 15 fps, corrosion rates became independent of velocity for all oxygen levels investigated (Figure 1). G.E. work^[2], however, indicated an almost linear variation of corrosion with velocity (actually $v^{0.884}$) up to the test limit of 25 ft/sec.

Observations on Corrosion Processes

Thorley and Tyzak^[1] reported an initial weight increase with time that was eventually superseded by a linear weight loss. The weight gain was due to the formation of a sodium chromite film (NaCrO_2) resulting in a denudation of chromium in the substrate material. Once this film was removed by the flowing sodium, nickel was selectively removed. A layer of ferrite was thus formed and remained in contact with the sodium for the remainder of the test. This selective leaching of nickel and chromium was also observed in the G.E. work at low-oxygen levels^[2]. They did not observe a weight increase or the presence of an oxide. As all low-nickel materials eventually end up with a ferrite surface layer, it is not surprising that their corrosion rates are similar. At higher oxygen levels (50 ppm) G.E.^[2] reported a selective removal of iron.

Observations on Corrosion Rates

Effect of Oxygen Content

Between 5 ppm and 50 ppm oxygen content in the sodium, the corrosion rate was almost directly proportional to the oxygen content. This marked variation is shown in Figure 2. Thorley and Tyzak postulated that the iron reacted with the sodium oxide in the sodium to form a double oxide that then went into the solution.

giving $3 \text{Na}_2\text{O (in Na)} + \text{Fe (Solid)} = \text{FeO(Na}_2\text{O)}_2 \text{ (in Na)} + 2 \text{Na(Liquid)}$.

Mottley^[3] proposed that the following reaction occurred without the formation of a double oxide:

Although the exact mechanism is not yet clear, the effect of oxygen on corrosion rate is extremely important, and is probably the most critical factor in sodium corrosion.

Figure 1 Effect of Velocity on the Rate of Metal Loss from Various Stainless Steels (Bore Diameter Constant)

Figure 2 Effect of Temperature and O_2 Level on Rate of Metal Loss of Various Steels in Flowing Sodium

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The Effect of Temperature

The variation of corrosion rate with temperature was consistent with Arrhenius-type rate behavior. The UKAEA data shown in Figure 2 were best described by the equation:

$$S = 2.3 \times 10^{-6} \exp\left(\frac{17,500}{RT}\right) \text{ cm sec}^{-1}. \quad (2)$$

where S = corrosion rate.

Analysis of the G.E. Data^[2] indicated that an increase of 100°F produced approximately a three-fold increase in the rate of corrosion, consistent with the temperature term in Equation 1.

The Effect of Velocity

UKAEA Data^[1]

A velocity dependence was observed only up to about 10 to 15 ft/sec (Figure 1). Above this range, the corrosion rate is essentially independent of velocity. The initial velocity dependence agrees with the theoretical model for a diffusion controlled process given by:

$$R = 0.023 (D/d) (\bar{N}_{\text{Fe}}^{\text{ox}} - N_{\text{Fe}}) \text{Re}^{0.8} \text{Sc}^{0.33} \quad (3)$$

where:

D = the diffusion coefficient of iron in sodium

d = the pipe diameter

Re = Reynolds number ($=dVp/\mu$)

Sc = the Schmidt number ($=\mu/pD$)

p = the density of sodium

μ = the viscosity of sodium

$\bar{N}_{\text{Fe}}^{\text{ox}}$ = the mole fraction of iron dissolved in the sodium at saturation for a given oxygen content.

Since Re is a function of V, $R \propto V^{0.8}$. Above 10 to 15 ft/sec however, it was shown experimentally that R was independent of velocity, and therefore, that some other limiting process was involved. Thorley and Tyzak^[1] suggested that the slow deposition of iron in the cooler parts of the circuit and/or some chemical process involving iron removal from the sodium via a double oxide, was responsible. Hopenfeld and Darley^[4] pointed out that a transition from a mass transfer controlled process to a surface reaction controlled process would give this result.

There is some supporting evidence for this change in the rate controlling process from other systems. Kassner^[5] studied the dissolution of Type 304 stainless steel in a Bi 42% Sn eutectic alloy by means of a rotating disc. At low angular velocities, the corrosion rate was velocity dependent, but became independent of velocity at higher angular velocities, indicating a transition to a surface reaction controlled process.

G. E. Data

Analysis of G.E. corrosion data indicated a velocity dependence as given in equation (2) throughout the velocity range investigated. This indicates that mass transfer through the boundary layer might be the controlling factor at high velocities as well as low. Recently, however, it has been reported that this velocity dependence is incorrect and that the velocity exponent is 0.2 rather than 0.884.

Conclusions

Continuous mass transfer will occur in a flowing sodium system as long as a temperature difference exists between various parts of the system. Generally, material will be removed from the hot regions and deposited in the cold regions. This process is complicated somewhat by 'the downstream effect', and material in the hot leg will show varying weight losses with position in the hot leg.

As expected, corrosion rates increase with increasing temperature. The effect of oxygen however, is unexpected. As can be seen, oxygen levels must be kept to a minimum for minimum corrosion rates.

The effect of velocity at velocities above 15 fps is apparently marginal. The UKAEA data, in fact, shows a velocity independent of corrosion rate. The more recent analysis of the G.E. data has only a slight velocity dependence of $v^{0.2}$.

Carbon Transfer

Carbon mass transfer will only be referred to briefly here, since problems should be minimal in an all-stainless steel loop.

If both austenitic and ferritic materials are present in the same loop, carbon will transfer from the ferritic to the austenitic alloy, resulting in carburization of the austenitic steel and possible degradation of mechanical properties. Carburization was observed in both programs in bi-metallic loops. In all-stainless steel loops, however, carbon transfer was insignificant^[2].

CORROSION ESTIMATES

Total metal thickness losses from the pump impeller and casing (type 304 SS and type 316 SS) after 20 years at 900°F, and 1000 hours at 1200°F, were estimated from the available corrosion data and are given in Table 2. For these calculations, it was assumed that the pump will operate as a hot-leg pump at 900°F. No corrosion would be expected if the pump were locked in the cold leg.

Remember that the estimated figures have been calculated from short-term corrosion data, and extensive extrapolation for both time and sodium velocity were necessary.

Estimates from the UKAEA Data^[1]

Above approximately 10 to 15 fps, the corrosion rate was found to be independent of the relative sodium velocity. The estimated figures, therefore, apply throughout the velocity range of interest.

The estimated thickness loss is given as a function of oxygen level (Figure 3) at 900 and 1200°F. The open circles represent the total corrosion after 20 years at 900°F, plus 1000 hours at 1200°F. At 15 ppm oxygen, which may be considered to be a typical upper limit operating level, the estimated material loss is 2 to 3 mils (Table 2).

Estimates From the G. E. Data^[2]

Corrosion rates were computed with the G.E. empirical equation for a range of velocities for times up to 20 years.

Maximum and minimum values were computed using the reported experimental limits with $\frac{L}{D} = 0$ and velocity exponents of 0.884, ± 0.160 , and 0.2 ± 0 .

The modified equation is given as:

$$R = V^{0.884 \pm 0.160} O_{x_c}^{1.156 \pm 0.364} \exp \left(\frac{12.845 + \frac{-23,827 \pm 2,584}{T + 460} + \frac{2.26 \pm 0.66}{t + 1}}{1} \right)$$

$$\text{and } R = V^{0.2 \pm 0}$$

The calculated values for 20 years at 900°F, plus 1000 hours at 1200°F are given in Figures 4 and 5; typical values are given in Table 2.

Based on these data, material loss by corrosion should not represent a problem during the pump design lifetime. High oxygen levels would only occur during start up and cleaning, at which time temperatures are kept low.

It must be emphasized that these estimates involved extensive extrapolation of published test data; however, they do represent the best available estimates at this time.

These losses are for uniform corrosion and do not take into account the possibility of intergranular attack.

Table 2

Estimated Total Corrosion after 20 Years at 900°F plus 1000 Hours at 1200°F

UKAEA Data (Velocity Independent)							
O ₂ Level (ppm)	10	15	30				
Thickness	1.3	2.3	10.5				
Loss (mils)							
G.E. Data (Velocity Dependent) at 15 ppm O ₂							
Velocity Exponent	0.2				0.884 ± 0.16		
Thickness Loss (mils)→	Mean	Max.	Min.		Mean	Max.	Min.
Na Velocity (fps)							
20	0.047	0.82	0.0027		0.22	5.6	0.008
40	0.62	1.1	0.0037		0.76	24.5	0.024
80	0.71	1.25	0.0041		1.4	54	0.041
100	0.79	1.35	.0043		1.8	62	.048

Figure 3 Estimated Corrosion after 20 Years as a Function of O₂ Level
(Data from Figure 4 TRG 1356 (C))

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Figure 4 Total Corrosion for 20 Years at 900°F + 1000 Hours at 1200°F
vs. Sodium Velocity

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Figure 5 Total Corrosion for 20 Years at 900°F + 1000 Hours at 1200°F vs. Sodium Velocity

Further metal loss may be expected at high velocities because of mechanical erosion. The impingement of the high-velocity fluid might cause localized failure at the impeller surface, resulting in metal loss. The importance of this problem is not yet known; but, an experimental program is being planned to resolve this.

MECHANICAL INTEGRITY

The Effect of Sodium Corrosion on Mechanical Properties

The effects of prolonged exposure to sodium on the mechanical integrity of structural materials are unknown; but, several possibilities exist. Briefly these can be listed as follows:

- a. deterioration of strength due to thinning of component cross sections by corrosion;
- b. structural changes caused by mass transfer, which can involve:
 - (1) selective leaching of specific elements from the hot-leg region, for example, the removal of chromium and nickel to give a ferritic surface structure and decarburization or de-nitriding of the surface,
 - (2) carburization; and
- c. grain boundary penetration.

Since the operating temperature and oxygen level will be low, it is not expected that the mechanical integrity of the pump will be significantly effected. The estimated corrosion losses given in Table 2 should not effect the pump components, since they represent an extremely small percentage of the total component thickness. Any affected layer formed by selective peaching will again be small compared with the total component thickness. Because of the 900°F temperature, decarburization should be minimized. The effect of decarburization on Type 304 stainless steel is to increase the yield strength and elongation without significantly affecting the UTS and impact properties^[6]. The short-term rupture strength and rupture life of cold worked Type 304 stainless steel thin wall tubing were found to be significantly lower after decarburization in sodium at 1300 and 1400°F^[7].

It must be emphasized that these data were generated at high temperatures (1175 to 1400°F) and under extreme decarburizing conditions (Zr hot trapping). Such changes will not occur under the pump operating conditions.

The Effect of Exposure Time on Mechanical Properties

Above 800°F, austenitic stainless steels are subject to sensitization, the precipitation of chromium carbide in grain boundary regions. Generally, the effect of sensitization on mechanical properties is to increase the ultimate strength and yield stress and decrease the elongation and impact properties. The magnitude of these changes in mechanical properties is a

function of both time and temperature.

Figure 6 and 7 give the available data for Types 304 and 304L stainless steels at 900 and 1200°F, respectively [8,9]. Assuming the changes to be additive, an estimate can be made of the changes in mechanical properties over the design lifetime of the pump. Mean starting values from Figures 6 and 7 are given in Table 3 together with the estimated changes due to 20 years at 900°F, and 1000 hours at 1200°F. The estimated final value is given in the last column.

Although measurable changes will occur they are not considered to be large enough to be deleterious, therefore, no problems are expected to arise from changes in mechanical properties.

Sigma Phase Formation

Sigma phase, an iron-chromium intermetallic compound, can precipitate in austenitic stainless steels after extended exposure at elevated temperatures. The formation of sigma is a function of composition, time, temperature, and the amount of cold work. The generally quoted temperature range for sigma formation is 1000 to 1600°F; but, it can form at lower temperatures in cold-worked materials, at least. Annealed Types 316 and 304 stainless steels, however, showed no sign of sigma precipitation after 2000 hours at temperatures up to 1500°F[11].

Welds in austenitic materials usually contain between 3 to 7% ferrite. This high chromium material is more susceptible to sigma formation than the parent metal. However, results on ferritic materials in the annealed condition, showed no evidence of sigma formation after 2000 hours at temperatures up to 1500°F[10]. On the basis of this information, sigma phase formation is not expected to present any problems under the proposed operating conditions.

Thermal Shock

Thermal shock damage at high temperatures is cumulative, especially if the dwell time after the temperature transient is of sufficient duration to allow relaxation[11]. The resistance to thermal shock is a function of the material fatigue behavior, and allowable stresses are estimated with high-temperature fatigue property data as given in Code Case 1331-1. Some cyclic strain fatigue data (10^5 cycles) are available for Types 316 and 304 stainless steels at 1200°F (650°C) in sodium of varying impurity contents[12,13]. The fatigue limit in clean sodium is higher than in air; high carbon causes little change; and a high-oxygen sodium causes a decrease in the fatigue limit.

It is believed, therefore, that the established fatigue data given in Code Case 1331-1 should be adequate for calculating allowable stresses under pump operating conditions. It would be advisable, however, to establish new fatigue curves for a sodium environment for future use.

Figure 6 Change in Room Temperature Mechanical Properties after Exposure at 1200°F

Figure 7 Change in Room Temperature Mechanical Properties
after Exposure at 900°F

Table 3

Estimated Room Temperature Mechanical Properties of Type 304 Stainless Steel After Exposure for 20 years at 900°F plus 1000 Hours at 1200°F

Property	Mean Starting Value	Change after 20 yr at 900°F	Change after 1000 hr at 1200°F	Final Value
UTS (psi x 1000)	81	+5.5	0	86.5
YS (psi x 1000)	31	+5	0	36
Elong. (%)	71	-7	-4	60
R. A. (%)	78	-6	-2	70
Charpy Impact (ft/lb)	89	-17	-23	49

CORROSION AND WEAR DATA FOR BEARING MATERIALS

Bearing materials suitable for operation in liquid sodium between 900 and 1200°F, were identified in the Large Pump Study Phase I^[14] as Haynes Stellites 1, 3, and Star J. Selection was based on adequacy of properties and past experience. Corrosion and wear data for these materials in liquid sodium are limited and consist mainly of screening test data and short-term, low-temperature, pump operating experience.

Friction and Wear

An extensive evaluation of the friction and wear characteristics of numerous material combinations is currently under way at the Liquid Metals Engineering Center^[15]. Discs of one material are rotated against flat-faced pins of a second material, at wear path velocities of about 2.5 to 4 cm/sec, while the pressure between the disc and the pins is maintained at 8.65 Kg/cm² (123 psi). Tests are performed in flowing sodium at 1200°F (650°C) with an oxygen level of 20 to 30 ppm. Preliminary information from this work is given in Table 4. The recommended materials are shown in various combinations with themselves and other Stellites, and some data for the Colmonoy alloys are also included for comparison. The specific wear rate values, reported as the volume of material removed per unit length of wear path per unit load, cm³/cmkg, show that Stellite combinations are an order of magnitude better than Colmonoy combinations.

A similar program at UKAEA^[16], using a radiused pin held against a rotating cylinder, showed that the amount of wear experienced by Stellite 1 - Stellite 1, Stellite 3 - Stellite 1, and Stellite 6 - Stellite 1 combinations increased with increasing temperature (Figure 8). Between 392°F (200°C) and 1112°F (600°C), the specific wear rate increased from 8×10^{-13} cm³/cmKg. Bearing loads of 2 and 4.5 Kg were used; but, actual stresses were not reported. Tests were again run at low speeds (3 to 5 cm/sec).

Corrosion Data

Sodium corrosion data for Stellites, or indeed any other bearing materials, are extremely limited, since unlike structural materials, Stellites have not been the subject of extensive corrosion studies. The data available have been generated in screening tests of short duration, such as tilting furnace experiments.

Baumgartner^[17] reported data for Stellites 1, 6, 12, and 21 in 20 to 40 ppm oxygen sodium up to 1400°F. Stellites 6 and 93 (iron base) hard facings were tested in NaK at 1200°F^[18]. Stellite 6 performed well with no metallographically observable attack in 100 hours and a recorded weight loss of about 0.6 mg/cm² under the same conditions.

Table 4

Performance of Various Material Combinations in Sliding Contact

Contact pressure 8.65 Kg/cm^2 (123 psi) up to 200 hours

Pin Material	Plate Material	Na. Env.	Na. Temp. (°F)	Pin Sliding V (cm/sec)	Coeffs. of sliding frict.		Specific Wear on 3 Pins/ (cm ³ /cm-Kg)
					Initial	Final	
Stellite 3	Stellite 6B	LCT	1200	2.3, 3.0, 3.5	0.31	0.35	5.73×10^{-11}
Stellite 273 HF	Stellite 1	LCT	1200	3.1, 3.9, 4.7	0.30	0.28	6.79×10^{-11}
Stellite 1	Stellite 6B	LCT	1200	3.1, 3.9, 4.7	0.36	0.50	5.73×10^{-11}
Stellite 3	Stellite 6H HF	LCT	1200	3.1, 3.9, 4.7	0.50	0.35	4.58×10^{-11}
Stellite Star J	Stellite 6B	LCT	1200	3.1, 3.9, 4.7	0.20	0.21	4.8×10^{-11}
Stellite 6B	Stellite 1	LHT	1200	3.1, 3.9, 4.7	0.28	0.31	5.3×10^{-11}
Stellite 273	Stellite 1	LHT	1200		0.26	0.22	4.3×10^{-11}
Colmonoy 6 HF	K-95	LCT	1200	2.3, 3.0, 3.5	0.61	0.83	75.1×10^{-11}
Colmonoy 4 HF	Colmonoy 5 HF	LCT	1200	2.3, 3.0, 3.5	0.37	0.63	25.8×10^{-11}
Colmonoy 4 HF	Colmonoy 75 HF	LCT	1200	2.3, 3.0, 3.5		0.35	80.5×10^{-11}

LCT = cold trapped

LHT = hot trapped

Figure 8 Wear Rates and Friction Coefficients for Stellite 1 and Stellite 3 in Sodium

Previous Operating Experience with Bearing Materials

The Rapsodie^[19], Hallam, Fermi, and EBR-II Reactors^[20] used Colmonoy-faced sleeves and shafts in the hydrostatic sodium lubricated pumps.

Bearing operation of the Hallam, Fermi, and EBR-II pumps has been satisfactory; but, all operations have been at temperatures below 1000°F. A seizure occurred on one of the Rapsodie intermediate pumps before attaining an operating temperature of 1022°F. The cause of the seizure was not determined. A Rapsodie primary pump seizure occurred sometime later and its probable cause was lack of wear resistance in the bearing material. The pump's operating temperature is now limited to 842°F. The same report states that all previous prototype bearings were made of Stellite, but for the Rapsodie pumps, a change to Colmonoy was made. The use of the proven material, Stellite, might have eliminated the seizures.

Stellite 6 bearing material has been successfully used in the PFR pump development program at temperatures of about 750°F for about 7000 hours^[21, 22]. This included 1800 hours at 10% power, during which significant wear would be expected. Examination of the pump at that time revealed no major problem.

It is impossible to give any estimate of bearing lifetime on the basis of existing data. Fortunately, however, both wear and corrosion results reported for Stellites to date are encouraging and indicate that the low-operating temperature and oxygen level will aid the reduction of wear and corrosion of the bearing materials.

Conclusions and Recommendations

Because of the pump operating conditions, 900°F, 15 ppm O₂, no significant corrosion problems are envisaged for the pump constructional materials. At these temperatures, the estimated corrosion thickness losses are extremely low and not likely to present any problems. There is a possibility of thickness loss by erosion in regions that experience a high relative sodium velocity, e.g., the impeller tip, (80 fps). No estimate of the extent of such damage can be given; but, a program is now being designed to determine this.

As far as can be ascertained from available mechanical property data, at this temperature and oxygen level, mechanical properties should remain unchanged from the available in-air or inert atmosphere properties. While no problems are expected in this area, there is an obvious need for mechanical property data in sodium over the entire temperature range experienced by plant components.

Data on Stellites indicate that, as far as corrosion and wear are concerned, they are the best choice and should perform adequately. However, the existing wear data are strictly comparative, and before estimates can be made of wear during operation, testing must be carried out under simulated startup and low-load conditions.

In conclusion, it can be said that a sodium pump operating at 900°F for 20 years, plus 1000 hours at 1200°F should not experience any materials problems. However, because of the shortage of data for this particular temperature range some work should be done in the areas of:

- a. mechanical properties in sodium on all constructional materials,
- b. mass transfer at high velocities on impellet materials; and
- c. wear of bearing materials under operating conditions.

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