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Interfacial Factors in Evaporator Surface Fouling,
A State-of-the-Art Survey
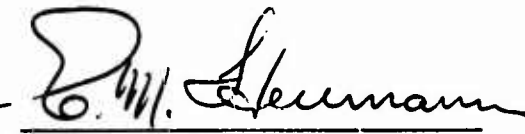
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By
B. H. Rankin and J. A. Bauman

B. H. RANKIN

J. A. BAUMAN

Approved by:

E. M. HERRMANN
Machinery Systems Division

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ABSTRACT

A survey of the literature was made to determine the
state of the art in understanding the mechanism of evaporator-
scale formation. The use of bulk conditions for the fluid and
the heating surface does not provide a reasonably accurate
model of the problem. Exploration of the variables in the
interface region should provide a logical and more fruitful
approach. Experimental examination of these interface
variables must be made under precisely controlled condi-
tions. Bench scale experiments are under way to establish
the effects of surface material composition and roughness
on scaling characteristics.

ADMINISTRATIVE INFORMATION

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INTERFACIAL FACTORS IN EVAPORATOR SURFACE FOULING, A
STATE-OF-THE-ART SURVEY

1.0 INTRODUCTION

In examining the problems of scale formation in both boilers and sea-water evaporators, it has been customary to consider the conditions of the bulk fluid and the bulk heating surface. Such techniques are not suitable for developing an understanding of the problem, because the scale forms in the changing region of surface imperfections, bubble nucleation, and thermal and fluid boundary layers that is called the interface.

1.1 Scope. This survey is concerned with scale formation on a surface where boiling of seawater occurs. Of particular consideration is the effect of the surface condition (material composition, and roughness) on the rate of scale formation and on the relative adhesion of that scale. An attempt is made to define each of the problem variables and show how it operates to control scale buildup and adhesion. The non-boiling case and other variables, such as heat flux, feed treatment, and concentration factor, are included because of their related effects.

1.2 Background. Scale control in sea-water evaporators is an extremely complex problem. A relatively good understanding of the scaling problem takes one into the disciplines of chemistry, thermodynamics, heat transfer, metallurgy, acoustics, fluid flow, and oceanography. It takes one into the newer fields of surface physics, physical chemistry, interface phenomena, and nucleation, where acceptable theories are in the process of being developed.

In the process of developing unified theory concerning the processes of adhesion, experiments based on various fragmentary theories are used to bring out evidence to support the general concepts. The interpretation of results, however, as well as the choice of experiment and procedure fall back on the fragmentary theories. Thus, a larger overview must be obtained before a unified theory can be developed and accepted. Interim practical solutions, however, appear only as partial solutions and will remain so until the final theory is accepted.

Distillation has been and presently is the only economically practical means of desalting seawater. Many other techniques and ideas, such as reverse osmosis, electrodialysis, freezing, and biological operations, have been or are being tried. Except for very special applications, none appears practical on a large scale within the immediate future. This thinking is borne out by the past and present programs of the Office of Saline Water (OSW). After carefully researching and developing to some degree almost every conceivable desalination process, the OSW built pilot plants for the new processes and then concentrated on building large evaporator-type distillation plants. Although research is continuing on the more exotic processes, almost all present OSW construction programs require large-scale evaporators. [1,2]

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[1] Superscripts refer to similarly numbered entries in Appendix A.

The thermal efficiency and the output-to-size ratio of the present distillation plants are limited by scale formation. When evaporation temperatures are raised to increase cycle efficiency and reduce plant size, or when discharge brine concentration is raised to reduce heat rejection and plant size, scale forms and reduces the heat transfer rates. This, of course, drastically reduces plant efficiency and output.

1.3 Early Approaches to Problem. A vast amount of research and experimentation has been done on controlling and/or eliminating scale formation. Two excellent reviews of the literature concerning scale formation and prevention are OSW R&D Progress Report No. 25 [3] and University of California, Department of Engineering Report 64-5. [4] A rather complete history of the research on evaporator scale can be obtained through either or both of these publications. The earliest work on scale control was generally on a hit-and-miss basis. Various types of tests were made on full-scale operating equipment under actual service conditions. Because of the large number of variables in an operational plant, the processes could not be closely controlled. As a result, information from these tests was usually inconclusive, and in some cases led to erroneous concepts.

When the Government and the universities began to see the need for more economical saline water conversion, additional funds and talent became available for attacking the problem of scale control. Laboratory-type evaporators with increasingly sophisticated instrumentation were used for the research. Much factual information was gained about the practical limits of operating conditions. The effects of the more powerful variables were determined within reasonable degrees of accuracy. However, the effects of the other variables could not be determined, so that no relatively complete understanding of the phenomena was developed.

1.4 Recent Approaches to Problem. During the last 5 or 6 years, investigators have recognized the magnitude of the problem of separating the variables in the complex problem of scale formation and are building specialized equipment to study just one or two of the variables. The important feature of this type of equipment is that all except one or two of the variables in the scale problem are held strictly constant. This type of research combined with the new developments in the theory of nucleation and interface concepts should eventually lead to a full understanding of the phenomena.

The early history of scale control was primarily a history of efforts to find a chemical additive that would effectively and inexpensively reduce or eliminate scale formation. This research has been successful to such a degree that plant efficiency has been greatly increased. It seems unlikely however, that chemical additive methods will produce any additional large improvements in plant efficiency although this research is continuing. Other means of scale control under investigation include techniques such as seeding to cause scale to deposit on preferred sites, and disturbing the scale formation by acoustics or vibration.

2.0 MEL APPROACH.

How various surface conditions affect scale formation has been investigated to a very limited degree. Some investigators [5] find that surface conditions do not affect scale formation. Such conclusions are in disagreement with theory and are probably due to lack of sufficiently accurate instrumentation or poorly controlled test conditions.

The accepted theories of nucleation show that the exact nature of the boiling conditions on any surface depends, among other things, on the surface material and its roughness. One of these boiling conditions is the degree of superheat at the surface. Another, is the movement of the boundary layer fluid. The degree of superheat at the surface determines solution temperature at the surface. The boundary layer movement determines solution concentration at the surface. Since scale precipitation is known to vary with solution temperature and concentration, scale formation must be a function of the surface material and its roughness.

An attempt is made in this study to investigate the scale formation problem in two ways, i.e., from the viewpoints of the theory of nucleation and the system variable concept. As might be expected, some of the ideas must be repeated because of interrelation of the concepts, and some which could be covered under one, may be found under the other.

3.0 NUCLEATION

The principles developed from nucleation theory must be applied when examining each of the interfacial factors and its effect on scale formation. The following discussion on nucleation is very limited; Walton [6, 7] provides an exceptionally fine description of the basic concepts and latest ideas on the subject.

Nucleation refers to the origin or birth of a nucleus of a new phase (usually) within a given material. Nucleation occurs in almost all processes; there are countless examples, such as the formation of snowflakes and raindrops. The origin of a steam bubble in boiling water and the birth of a crystal in a solution are two examples pertinent to this study. The problem is to understand exactly how and why molecules of a particular phase gather together to form a cluster that in turn becomes a nucleus of a new phase. Because of the extremely small size of the participating parts, complete experimental visual observation of the actual process is virtually impossible. Without complete observational information, the development and verification of a mathematical model for the process is extremely difficult.

Nucleation is either homogeneous, where the nucleus is born within a homogeneous material, or heterogeneous when the nucleus is born upon or around some foreign, particle or surface. The foreign particles must be at least several molecules large before they can become nucleation sites. Heterogeneous nucleation occurs much more readily than homogeneous, and since almost all materials have some impurities of particle size present, almost all nucleation processes are heterogeneous. Of course, the foreign participants in the heterogeneous process make that process much more difficult to understand. For this reason, the homogeneous process has received much study, whereas, heterogeneous theory is almost completely lacking.

3.1 **Homogeneous Nucleation.** The Volmer or classical theory of homogeneous nucleation as modified by Becker, Doring, Frenkel, and others, is based upon equilibrium thermodynamics. The macroscopic concepts of free energy or surface tension are used to explain the critical size and growth of nuclei, but the theory does not explain the initial grouping of the clusters themselves. There is serious doubt as to the validity of this theory because nucleation is not an equilibrium process.

Further, the cluster sizes may be too small to use the macroscopic properties of free energy and/or surface tension. Nevertheless, the modified classical theory is the only usable theory available. It gives good agreement in most cases, and can be shown to provide agreement with experimental data for some heterogeneous cases. Some progress is being made in developing a microscopic nucleation theory that uses statistical procedures to equate molecular forces under dynamic conditions. [8] Application of such a theory for even the simplest homogeneous processes is extremely complex. It is doubtful if such a theory for use on most real systems will be available in the near future. Heterogeneous theory, classical or otherwise, is almost non-existent.

From the above, it is clear that a theoretical model that will predict scale formation is certainly not available, nor can one be easily developed. Nevertheless, homogeneous theory is certainly well enough advanced so that it will aid in an understanding of a real heterogeneous process: scale formation on a heating surface.

3.2 Heterogeneous Nucleation. In the case of scale formation on a heat-transfer surface when boiling occurs, two types of heterogeneous nucleation take place:

- Boiling Nucleation: Steam bubbles form on the heat-transfer surface.

- Crystal Nucleation: Crystals of scale form on the heat-transfer surface.

3.2.1 Boiling Nucleation. First, consider a boiling fluid when no heating surface is present, as in a "flash chamber". Bubble nucleii are formed by clusters of higher energy molecules that attain critical size. The cluster molecules are in the gas phase and must have a much greater thermal energy than that of the average molecules of the surrounding liquid. Since the critically-sized clusters are born within the liquid, they must be the product of random groupings of very high energy liquid molecules. When the number of high energy liquid molecules increases, the statistical possibility for the creation of vapor clusters within the liquid increases. Reasonably large number of high energy liquid molecules can be created by heating the liquid above the liquid vapor equilibrium temperature, i.e., superheated liquid. It is, then, readily seen that gas phase nucleation rate increases with superheat and that for all boiling, a certain degree of superheat is required.

The surface tension in a bubble is balanced by the kinetic pressure exerted by the steam within the bubble. For very small or subcritically-sized bubbles, the surface tension would be larger than the kinetic pressure so that clusters of subcritical size are transient and disappear. Absolutely pure water has a very high surface tension so that extremely high superheats (several hundred degrees) would be necessary to produce bubbles. Impurities serve as centers of nucleii so that relatively low superheats are required.

Now, consider the boiling to occur at the heat-transfer surface. Both the roughness and the material composing the boiling surface alter the picture and affect the superheat requirement. Pits on the surface provide vapor pockets that exist even when the liquid is subcooled. Growth of the vapor volume within the pocket then depends upon additional high energy molecules entering the pocket until the surface tension of the liquid covering the pocket is overcome and a bubble is released. The existence of vapor pockets provides very favorable conditions for the formation of the

critically-sized nucleii so that only low degrees of superheat are required to provide the bubbles. For higher heat fluxes, increased bubble release rate is required. Since this must be accomplished by increasing the number of high energy molecules that become part of the pocket, it is readily seen that higher superheats must accompany higher flux rates. Smoother surfaces provide smaller pockets which require larger energy additions before bubble release. This means that smoother surfaces require higher superheats. The surface tension that acts between the heat-transfer surface and the bubble surface causes the bubble to be released more or less readily and thus affects heat transfer and the superheat requirement. This interfacial tension is a function of the kind of material present and is, effectively, a function of the surface roughness.

3.2.2 Crystal Nucleation. Crystal nucleii are formed by clusters of molecules that attain a critical size, of much lower energy than for bubble nucleii. The actions are much the same as those of steam bubble nucleation. For these critically-sized or larger clusters to form, some amount of supersaturation must be present. If a foreign body is present, crystals will form with less supersaturation. The analogy does not appear to hold for the steam-bubble surface cavity. Other concepts appear to apply. Crystals seem to nucleate most readily on projections into the fluid, on materials that have related crystal line structures, or on previously formed crystals. Shock, electric charge, and fluid flow over the heat-transfer surface also appear to affect the crystal nucleation sites as well as the nucleation rate.

4.0 INTERFACE SYSTEM VARIABLES

4.1 Categories of Variables. The variables comprising the scale-producing system may be classified broadly into three categories; fluid variables, surface variables, and fluid-surface variables. These have been explored in the literature with a view to evaluating the effect of each. This scale-producing system is definitely nonlinear. The principles of superposition cannot be applied so that each variable can be considered alone. Nevertheless, an attempt is made to define and describe in general, the effect of each individual variable.

4.2 Individual Variables. Before considering the individual interface variables in detail, the problem must be defined. The temperatures of the heating surface and the fluid in contact with that surface are always higher than that of the fluid which is well-removed from the surface. This fluid layer, where the properties are changing, can be called the fluid interface, a graphical representation of which appears in Figure 1. Its concept is somewhat analagous to that of the fluid boundary layer in fluid flow, or thermal boundary layer in convective heat transfer.

No surface is absolutely smooth; there are always some structural defects in it. The irregularities cause variations in the temperature, conductivity, electrical potential, etc., so that the properties of the surface are not uniform. This solid surface layer where the properties are changing can be called the solid interface. The fluid-plus-solid interface produces the total interface that must be studied and understood as a unit.
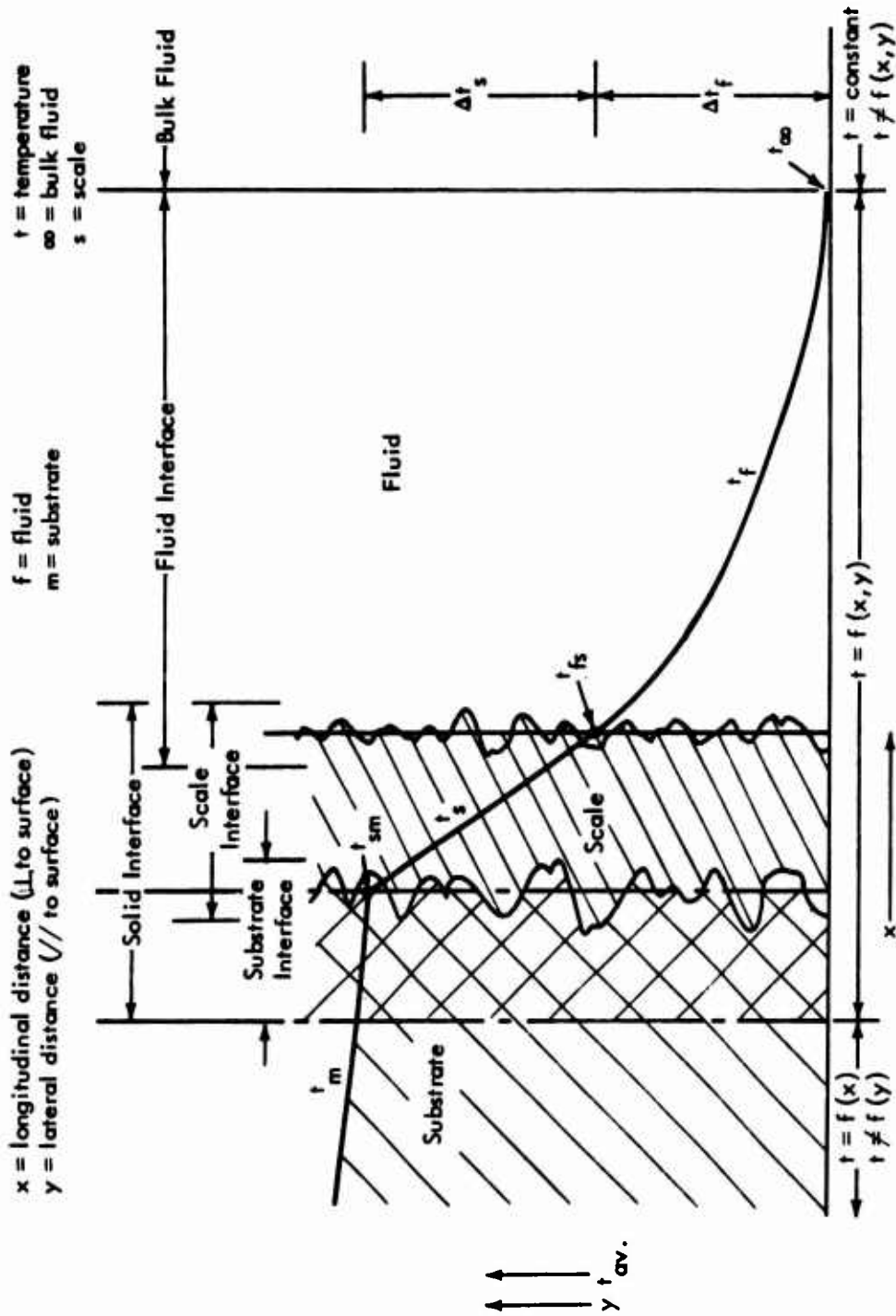
Figure 1
Interface Definitions

As indicated above, the rate of scale formation on a surface is a function of the fluid variables, surface variables, and fluid-surface variables. Rate of scaling is a strong function of some of these variables and a weak function of others. Perhaps some of these variables have a negligible effect on scale, but each variable must be investigated before it can be established that its effect is negligible.

The individual variables making up each category of the interface system are listed in Table 1. Identifying item numbers are assigned to each variable for convenience in discussing each in detail.

Many of these variables are dependent to some degree on several of the others. For example: Δt_s depends upon $\dot{q}$, scale porosity, scale roughness, fluid velocity, entrained gases, fluid bulk temperature, surface roughness, and surface material. Considering the fact that there are at least 20 odd, interrelated variables in this problem, it becomes obvious why a complete mathematical model has not yet been achieved. Probably the best that can be done is to conduct experimental studies where one variable is carefully examined while the others are all held strictly constant. This is not a simple task because some of the variables are not easily controlled; nucleation is a somewhat random phenomenon.

4.3 Strong vs Weak Influences. Scale formation is a strong function of the temperature, salt concentration, chemical treatment, heat flux, and acoustical disturbance. These have all been rather carefully investigated. Scale is also a weak function of the other variables and altering them does not produce such drastic effects. They have been studied independently only slightly, for several reasons:

• It was necessary to understand the operation of the more powerful influences first.

• It appeared that scale control should be accomplished by action of the strong influences.

• Effects produced by the weak influences may be masked by very slight perturbation of the strong influences. Test apparatus that will provide adequate process control and instrumentation to study variables where scale is a weak function will be extremely difficult to build and maintain.

4.4 Fluid Variables. In the following, a more detailed examination is made of the individual interfacial variables listed under paragraph 4.2. Wherever evaporator temperatures are discussed and not defined otherwise, the temperature concerned is the bulk boiling temperature.

4.4.1 Variable 1a: Bulk Boiling Temperature (t_∞). It has been established that the type of scale and scaling rate vary with boiling temperature. Hillier's curves, Figure 2, show that calcium carbonate, ($CaCO_3$), is the principal scale constituent at boiling temperatures up to approximately 175 F*. Magnesium hydroxide (Mg (OH)$_2$)

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*Abbreviations used in this text are from the GPO Style Manual, 1959, unless otherwise noted.

Table 1
Interface System Variables

| 1. Fluid Variables	2. Surface Variable	3. Fluid-Surface Variables
a. Boiling temperature (t_∞)	a. Substrate roughness	a. Time (velocity)
b. Salt concentration	b. Substrate material composition	b. $\dot{q}$ = heat flux
c. Sea-water characteristics	c. Substrate cleanliness, corrosion, age	c. Δ e = electrical potential between the fluid and the surface
(1) pH (acidity)	d. Scale material composition	d. Acoustic or vibrational action
(2) Entrained and dissolved gases	e. Scale thickness	e. Δt_f = temperature drop across the fluid interface
(3) Microscopic marine life	f. Scale roughness	f. Δt_s = temperature drop across the solid interface
d. Chemical feed treatment	g. Scale porosity	g. Geometry
e. Velocity (pool boiling, forced circulation)		

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A - Magnesium Hydroxide, $Mg(OH)_2$

B - Calcium Carbonate, $CaCO_3$

C - Calcium Sulphate, $CaSO_4$

Item (a) Nominal Temperature-Difference, about 22 degrees, F

Item (b) Nominal Temperature-Difference, about 80 degrees, F
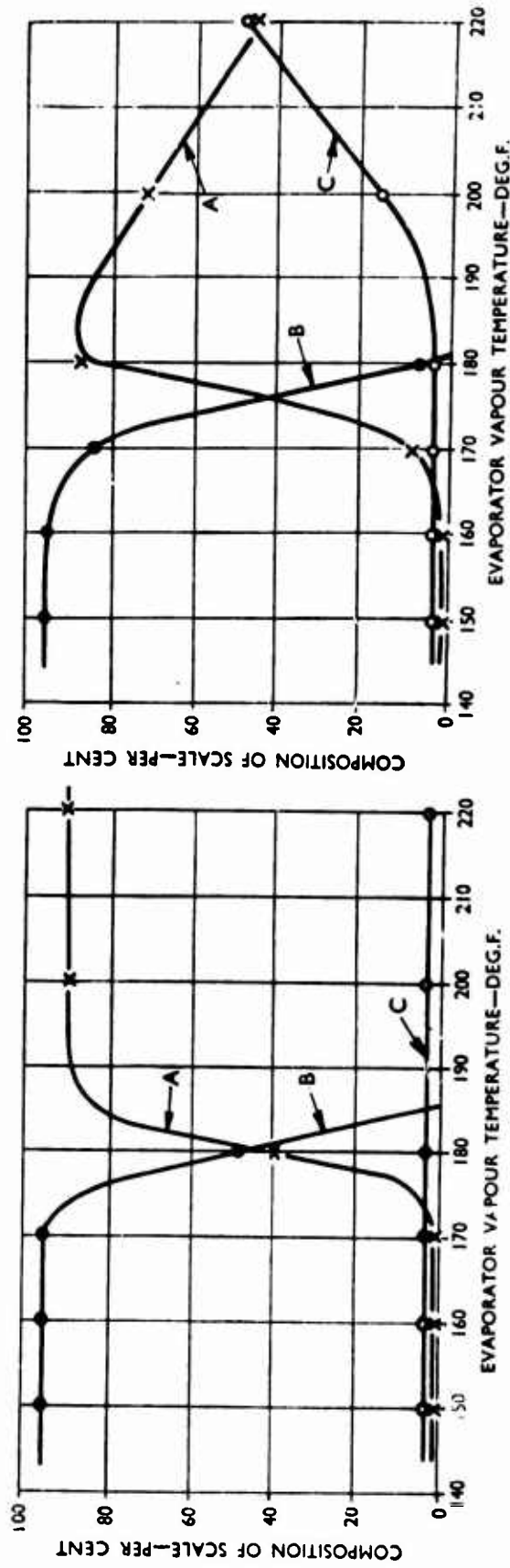
Figure 2

Main Constituents of Scale at Different Evaporator Temperatures

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becomes the principal constituent up to about 210 F, beyond which it is calcium sulphate $(CaSO_4)$. These figures are for a concentration factor of 2 and vary considerably with heat flux, concentration factor, and pH. Naval evaporators operate up to approximately 180 F [9] where $Mg(OH)_2$ and $CaCO_3$ are the principal scale constituents. The new large land-based plants use acid feed treatment, so that they operate satisfactorily up to the 250 F range. Scale is amost entirely $CaSO_4$ at this temperature.

4.4.2 Variable 1b: Salt Concentration or Concentration Factor. The solubility of $CaSO_4$ decreases with increased temperature. $Mg(OH)_2$ and $CaCO_3$ in sea water also have an effective reverse solubility curve because of the carbonate ions (the liberation of CO_2 as temperature increases). Figure 3 shows that for any particular temperature, there is a concentration limit above which scale will form. [3,5, 10, 12] This, of course, limits the proportion of the sea water that can be evaporated before the brine must be rejected.

4.4.3 Variable 1c: Sea-Water Characteristics. There are three significant variables in seawater: (a) pH, (b) entrained and dissolved gases, and (c) microscopic marine life. The proportions of the dissolved solids in sea water do not vary significantly. They are almost the same the world over [13]. Seawater from the polar regions contains slightly less salt than that from the tropic seas. This is because of the higher evaporation rates near the equator. But the relative proportions of the various salts do not change.

● The pH of seawater varies somewhat throughout the oceans. [10, 13] Figures 3 and 4 show that pH has a marked effect on the saturation of the scale materials and markedly changes the scaling rate. The technique of pH control has been used to reduce the $CaCO_3$ and $Mg(OH)_2$ scale at the OSW full-size demonstration plant at Point Loma, California. [14, 15]

● The quantity of O_2, CO_2, H_2S, and N_2, that is dissolved or entrained in sea water varies widely depending upon the sea-water temperature, depth, organic activity, and wave action. [13, 16] The amount of dissolved CO_2 has pronounced effect on the $CaCO_3$ and $Mg(OH)_2$ scaling rate because of its effect upon the pH. [4]

● Microscopic marine life activity varies widely [17] and causes changes in the dissolved gases and the pH. Marine life activity probably has many, as yet to be determined, other effects on the scaling potential of seawater. Seawater cannot even be stored without undergoing change in character. [16, 17]

Because of the variable nature of seawater, controlled experiments should be conducted with synthetic seawater or solutions containing only one or two of the sea salts. There are several recipes for synthetic seawater. [18] That given by ASTM 1141-52 is often used. It should be noted, however, that experimental results from synthetic or substitute seawater may vary considerably from those of natural seawater. [10,2] It would seem that the best approach would be to use the simpler solutions to obtain uniform experimental conditions and then to verify the results by using natural seawater.

4.4.4 Variable 1d: Chemical Feed Treatment. Scale formation can be suppressed through the use of chemical additives in the feed. Not long after Watt invented the

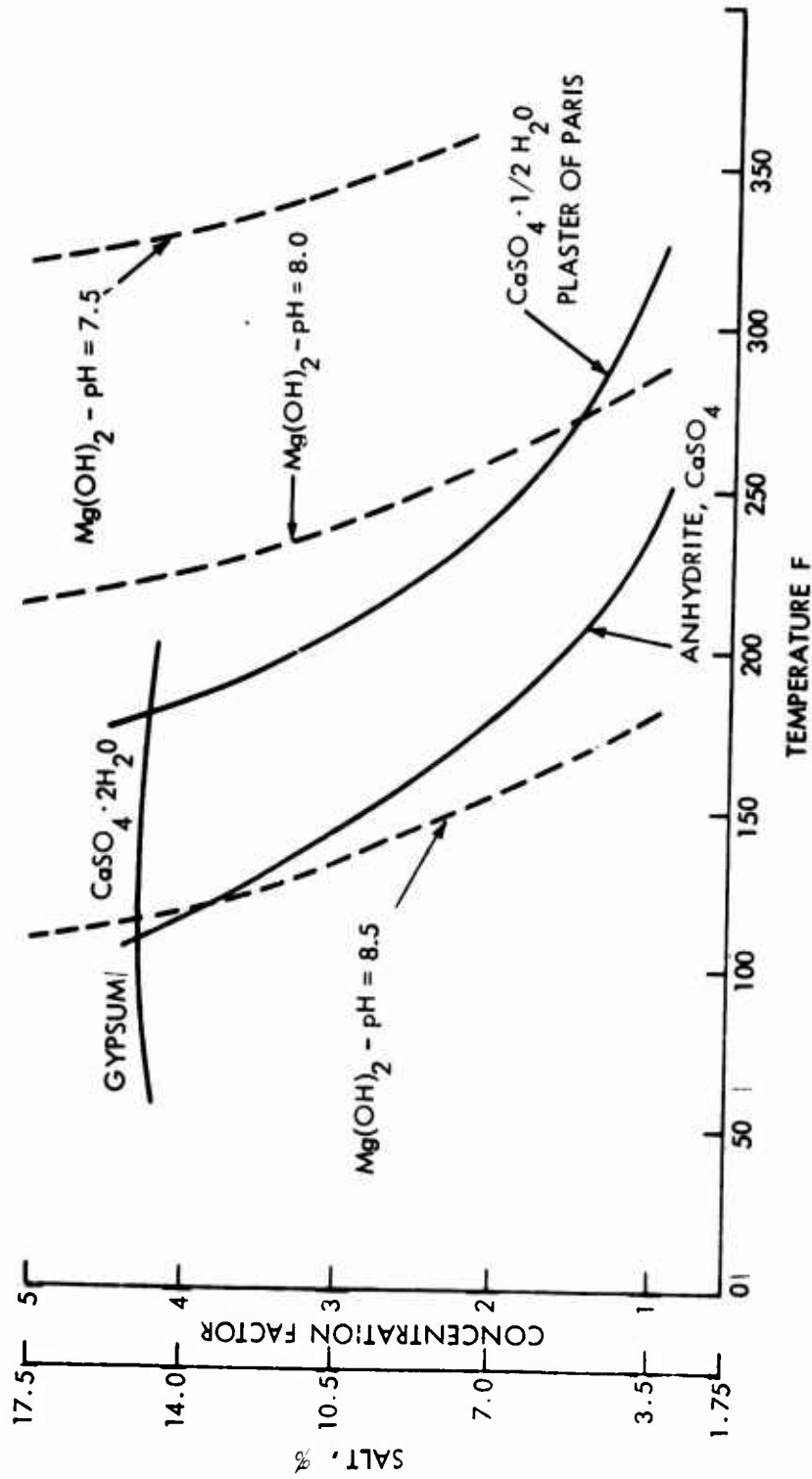USN MARINE ENGINEERING LABORATORY

Figure 3
Approximate Solubility Limits of Calcium Sulfate and
Magnesium Hydroxide in Seawater Concentrates
(Redrawn from Standiford and Sinek[11] and Mulford[9])
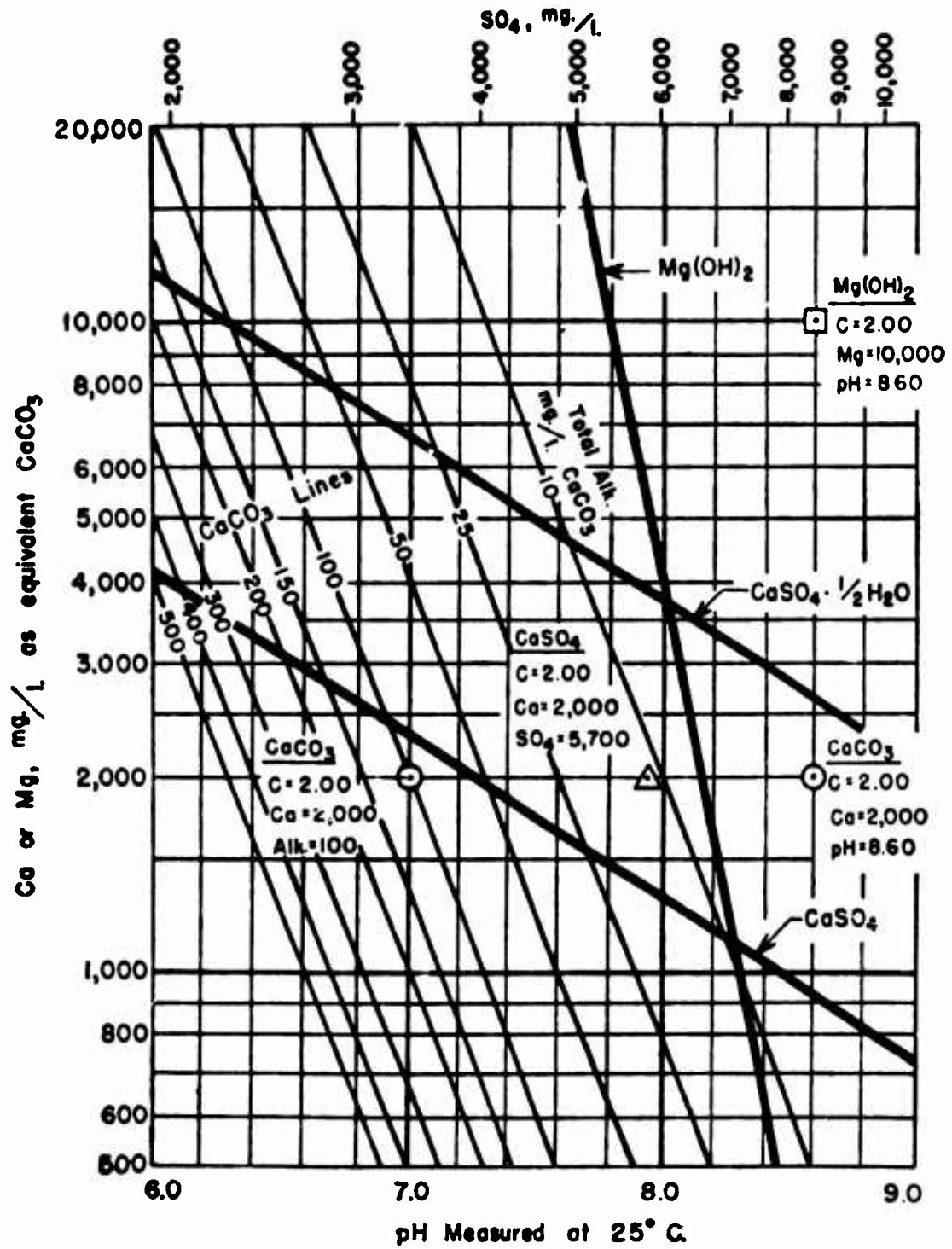
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Figure 4
Stability Diagram of Evaporator Brine

steam engine, operators knew that potatoes added to the feed would decrease scale formation. [19] Little more was learned about feed treatment until the late 1940's. During and immediately after World War II, the Navy used a starch and boiler-compound mixture as an additive. In 1949, Hildebrand and Warren [20] published results of laboratory tests that used such additives as Calgon, corn starch, sugar, molasses, calcium chloride, potassium chloride, sulfuric acid, tube coatings of wax, varnish, and resin. Although considerable work on boiler scale control was done by the U. S. Naval Engineering Experiment Station and others, the work of Hildebrand and Warren appears to be the first extensive laboratory investigation for evaporator scale prevention by feed treatment. Lurie, et al [21] gives 17 groups of additives and lists 39 references covering them.

A commercial additive called Hagevap became very popular during the 1950's and is still being used widely with considerable success. Hagevap is basically a polyphosphate compound mixed with several other ingredients. This additive causes the scale to form more slowly, and to be softer and more easily removed. Hagavap works well up to about 180 to 190 F. The U. S. Naval Engineering Experiment Station (now the Annapolis Division, Naval Ship Research and Development Center) developed several newer additives which appear to provide good results at higher temperatures and brine concentrations. These additives have been laboratory-tested, but have not yet received complete operational testing. [22, 23] A proprietary additive is being marketed in Japan; [24] and in Australia, laboratory success has been achieved at 240 F with a polyacrylic acid additive. [25] The most recent development in feed treating is to control the pH by injecting acid into the feed. This method permitted 250 F operation of the demonstration flash plant at Point Loma, California, and 300 F operation of the 10,000 gallons per day experimental flash plant at Wrightsville Beach North Carolina. [15] Acid injection eliminates the need for the phosphate additives and seems to offer the best scale-control technique for shore based plants. This technique is obviously not suitable for use in shipboard installations because of the handling and storage problems produced by the large quantities of acid.

4.4.5 Variable 1e: Velocity. The velocity of the fluid is considered or taken relative to the heat-transfer surface. It is therefore covered under fluid-surface variables, in the discussion of Variable 3a.

4.5 Surface Variables.

4.5.1 Variable 2a: Substrate Roughness. The roughness of the heat transfer surface substrate apparently produces two effects:

● Nucleation sites for boiling are a function of substrate roughness. Scale formations vary in the vicinity of the sites because of the local large fluctuation in temperature and flow.

● Nucleation sites for the inception of crystalline deposits appear to be a function of the surface roughness because discontinuities offer preferred sites for crystal inception.

In the case of scale formation on a substrate where boiling occurs, crystal and boiling nucleation occur with three phases present: solution, vapor, and scale crystals.

In the case of scale formation on a substrate where no boiling occurs, only crystal nucleation occurs with two phases present: solution and scale. The latter case is representative of scale deposition in the salt water heater of a flash type distilling plant.

4.5.1.1 Boiling Nucleation Sites. The relationship between boiling nucleation sites and surface roughness is well documented. Wallis and Griffith [26] manufactured nucleation sites with a phonograph needle and showed that nucleation occurred more readily at the cavity. Westwater, et al, demonstrated by photography that nucleation occurs at preferred sites [27], and developed a method to locate and count the sites. [28] Kurihara and Myers [29] developed curves that related surface roughness to heat transfer coefficients. Marto and Rohsenow [30] have shown that size, shape, and age, of the surface roughness affect nucleate boiling and have developed criteria for the operation of nucleation sites. Bonilla [31] has shown that the nucleation sites occur at optimum spacing for various heat fluxes. This optimum spacing was established from tests on plates with carefully manufactured parallel scratches.

The previous discussion of the bulk boiling temperature indicated that scale formation is a function of temperature. This holds true for the microscopic scratches on a surface as well as for bulk temperatures. When heat is transferred from a surface to a fluid, the point temperature on the roughness peaks must be less than that in the valleys because the peaks are farther from the heat sources. As a result, the fluid in the valleys will be more supersaturated so that scale will form more readily in the valleys. The greater the roughness, the greater this effect should be. In the case of the boiling heat transfer, the boiling nucleation sites, which are affected by roughness, also effect scale formation. Mesler et al [32] and Hsu and Schmidt [33] proved that large temperature fluctuations occurred in the vicinity of boiling nucleation sites. These large nucleation induced temperature fluctuations have a major effect on nearby scale formation. This is established by results from many investigators who found that the initial deposits of scale were in the form of small rings. [5] Recently, several investigators have proved that these rings are located around the nucleation sites.[34] The authors of this paper have observed these rings, and that where nucleation does not occur, scale formation is uniform. Figures 5 and 6 illustrate patterns of nucleation site scale as compared with scale on nonnucleation areas.

4.5.1.2. Crystal Nucleation Sites. The relationship between crystal nucleation sites and surface roughness is less well documented. Chandler [35] and Hickman [36] have shown that well-defined projections on a heat-transfer surface are preferred sites for initial nucleation of crystals. After initial nucleation, the crystals themselves become preferred sites. [37, 38] Zettlemeyer [39] uses a theoretical approach to show how roughness, cracks, dislocations, impurities, and chemical attack, on the surface tend to alter the surface chemistry and thereby change the crystal nucleation process. Corners are more likely to participate in actions than crevices. Rough surfaces show much greater total surface and give much greater wetting energies than do smooth surfaces. Supporting views of these characteristics are presented by Hirth. [40]

It should be noted that since boiling nucleation tends to cause scale in the valleys, whereas crystal nucleation prefers the peaks, the two processes have a synergistic result. Surface roughness, boiling rate, and many other factors will determine the relative strength of each process.
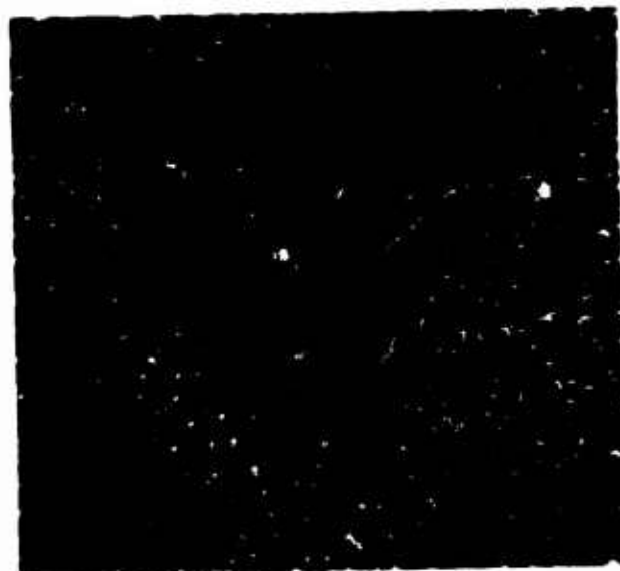
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Figure 5

Figure 6
Substitute Seawater Scale Showing
Pattern of Nucleation Sites (8X)

4.5.2 Variable 2b: Substrate Material Composition. The surface material affects the initial rate of scale formation because it affects the boiling characteristics (flow pattern) next to the heat transfer surface.

The tendency for a liquid to cling to, and spread on, a surface is called wettability. This phenomenon along with other aspects of surface tension is described by Zisman. [41] Wettability is governed by the surface tension of the water and the surface tension of the material. The size and shape of a droplet of water depends upon the surface material upon which it rests. It is easily seen then, that the size and shape of a droplet of steam formed in water at a solid heating surface, must be controlled by the surface tensions of the water, steam, and solid.

It should also be noted that the surface tension of a material is affected by its temperature, and any chemical additives (dirt, grease on a solid, chemical treatment for the water). The interfacial tension which results from the various surface tensions is also affected by the surface discontinuities or roughness. [41] From a strictly practical point of view, the concept of surface and interfacial tension gives the most useful understanding of the material to boiling relationship. The free surface energy and modern nucleation theory concepts give the most enlightening theoretical approach.

Investigation into the effect of surface materials on scale formation has been very limited. Hildebrandt and Warren [20] coated heat transfer surfaces with varnish, wax, and resin. Results indicate that the surface material affected the scaling rate. Many investigators believe that feed treatment additives alter the surface characteristics and thereby change the scaling rate. Recently there have been claims made that Teflon tubes resist scaling. [42] Although these claims appear to have some validity, the poor thermal conductivity of Teflon limits its competitive application to chemical processes where corrosion as well as scale present problems with metal tubes. Thin-film Teflon coatings, if durable, would probably show some effectiveness in limiting scale deposition.

Experimental results that compare the scaling rates for various materials seem to be nonexistent. Swailes [43] states that Monel evaporator baskets gave less scale buildup in tests run at the Admiralty Distilling Experimental Station, Portland, British Isles, but these results could have been caused by other factors. Leister [44] has shown that the surface material has a large effect on the scale adhesion. Scale adheres poorly to Teflon, glass, and rubber. A very important contribution to this problem of surface material effects has been made by Young and Hummel, [45] who showed that boiling occurs much more readily on Teflon (low surface energy) surfaces than on stainless steel. For the same bulk water temperature, the water temperature at the interface is lower on a Teflon surface. The resulting lower interface temperature should result in less scale formation.

4.5.3 Variable 2c: Substrate Cleanliness. Small amounts of dirt or film deposited on a surface may vastly alter its surface energy. As discussed under Variable 2a, this will change the scaling picture because the preferred crystal nucleation sites will be altered. [39] Dirt films will also change the boiling nucleation sites. This will result in a change in the scale buildup. Similar effects are produced by corrosion or aging of the substrate surface.

Although there appears to be no known work on the direct effect of surface cleanliness on scale formation, the above theory is supported by work in feed additives. Many investigators believe that the surfaces are coated by the additives so that scaling rate is reduced. The theory of surfactants (materials that alter surface tension) presented by Schwartz and Reid [46] can be extended to show how surface cleanliness would affect scaling. Zettlemeyer et al [47] show that altering surface layers with monomolecular coatings or by just heating the surfaces for a time will cause the nucleating qualities of that surface to be altered.

4.5.4 Variables 2d, e, f, and g: Scale Characteristics. Scale alters the substrate surface so that the solid side of the interface becomes a new and different material and therefore has its own characteristics. The substrate originally controls the scale formation, but as the scale builds up the properties of the deposited scale begin to control the scale formation process and the substrate contributes less control over the scaling. Scale thickness, material roughness, and porosity will determine the relative contribution of the substrate to the fluid interface conditions that determine the scaling rates. The authors of this paper have noted that scale does not completely form over strong boiling nucleation sites, even though appreciable scale may be present over most of the substrate.

It is believed by some observers that $Mg(OH)_2$, will not readily form unless $CaCO_3$, crystals are already present. Such interactions between scale constituents might be expected because of the difference in their surface energies.

4.6 Fluid Surface Variables.

4.6.1 Variable 3a: Time and Fluid Movement. Nucleation is statistical in nature because random groupings of lower energy molecules form the critically-sized clusters that become crystal nucleii. In other words, time is required for the statistical chance that a right sized cluster be formed. Power and Fabuss [38] have demonstrated that there is a very definite relaxation time for the initial formation of scale crystals on a substrate. [1] Any local concentration of the salts should decrease this time, and any fluid disturbance that tends to destroy local concentrations at the heating surface should increase this relaxation time. The authors know of no investigations that provide a relationship of flow to crystal nucleation time. It is felt that a correlation of relaxation time for crystal inception, surface roughness, and fluid velocity would shed considerable light upon this subject. Investigations of acoustic disturbance to destroy the salt concentrations at the heating surface are presently being conducted. [1]

4.6.2 Variable 3b: Heat Flux (q). The effect of heat flux on scaling rate is well known. Many investigators have shown that as the heat flux increases, the scaling rate increases. Fourier's equation for heat transfer shows that as the heat flux through a layer is increased, the temperature drop across that layer must increase. For the case being considered, as q increases, the temperature difference across the fluid interface, Δt_f, increases. This means that the surface temperature will be higher for a particular bulk fluid temperature, t_∞, and scale will form more readily. Of course, disturbance of the fluid layer by steam bubbles, fluid flow, and acoustics, will alter the system and make development of a mathematical model for such a system extremely difficult. Madijski [48] and others developed mathematical models for the case of nucleation and pool boiling for a defined surface. Han and Griffith [49] developed a model that

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included surface roughness concepts. Davis and Anderson [50] have extended the effort
to include forced flow. Many simplifying assumptions have been made in those models
so that although they appear to match most experimental data, they are not applicable
to many specific cases. The authors know of no model that includes scaling rate, sur-
face roughness, and heat flux.

4.6.3 Variable 3c: Electrical Potential Between the Fluid and the Surface, (Δe).
Some investigators [10] believe that scale formation is closely related to the electrical
potentials involved; other investigators consider the ionic nature of the chemistry
only; still other investigators seem to ignore the electrical potential aspects completely.
The chemical operations involved are most certainly ionic in nature and therefore should
have a relationship with the electrical fields at the scaling surface. Less than 20 years
ago several devices were marketed that operated on the principle that electrical treat-
ment of evaporator feed would reduce scale, but no conclusive scientific evidence of
their effectiveness is available. Although such a scheme hardly seems plausible, there
is good reason to suspect that electrical potential on the evaporator tubes would have
an effect on scale formation. Mulford [9] discussed another effect: several types of
electrolytic cells have been used that released sacrificial ions to prevent scale buildup.
Schmid, et al, [51] showed that electrostatic fields alter the surface tension of salt
solutions. Schmut [52] discussed the theory of using electrical potential to control
dispersions in fluids as applied to the paper making industry. Sennett and Oliver [53]
discuss the theory of microelectrophoresis which is concerned with dispersions having
a size on the order of those of nucleation clusters. Pouring [54] showed that ex-
ceptionally high potentials are generated during homogeneous condensation nuclea-
tion in nozzles. The electrical potential effect upon nucleation in cloud chambers
is well known. Silverman [1] is conducting basic research on potential effects on scale
formation.

It seems that inception and growth of crystal nucleation and inception of boiling nuclea-
tion must be related to electrical potential. Suitable experiments to determine the
effect on scale formation by electrical potentials must be carefully planned because
the electrical potential distribution on a surface is a function of the surface roughness
(both the substrate and scale, and their relative conductances), fluid flow, local salt
concentrations, and bubble formations, and each of these effects must be carefully
evaluated.

4.6.4 Variable 3d: Acoustical Disturbances. Acoustical disturbances cause small
but rapid movements and very sudden pressure fluctuations in the fluid. Both of these
affect the scaling rate in several different ways. Because water has been evaporated
from the solution next to the heating surface, the salt concentration is greater in the
interface region. The small but rapid movements of acoustically-disturbed fluid cause
mixing within and between the fluid interface and the bulk fluid. This mixing results
in a lower salt concentration next to the heating surface and decreases the thickness of
the fluid interface so that the surface temperature is reduced. As a result, the scaling
rate decreases. Fand and Cheng [55] have demonstrated that acoustic action decreases
the fluid interface thickness and promotes better heat transfer. Additional work on
reduction of sea water scale though the use of acoustics is in progress. [56, 57] The
acoustically-induced pressure fluctuations affect the scaling rate by changing the
saturation temperature of the fluid. When the pressure drops, the saturation tempera-
ture drops, and this causes the liquid phase to be supersaturated so that it tends to boil.

The net result is that the acoustic pressure produces a very high degree of supersaturation at the heat transfer surface and thereby improves the boiling nucleation. This results in decreased scaling.

Messino, Sette, and Wanderlingh [58] provide a discussion of the fundamental theory of acoustically-induced nucleation. The acoustically-induced pressure fluctuations should also affect the crystal nucleation rate. Crystals tend to form more readily when the metastable supersaturated solution is shocked by pressure impulses.

4.6.5 Variable 3e: Temperature Drop Across the Fluid Interface: (Δ t_f) The temperature drop, Δt_f, is the direct measurement of the supersaturation of the fluid in contact with the solid surface. Since the scaling rate increases with fluid temperature (see Variable 1a), a very small value of Δt_f is desirable. This occurs when the heat flux is small or when the interface thickness is small for a particular heat flux. The fluid interface thickness can be reduced by increasing fluid transfer between the fluid interface and the bulk fluid, through one or more of the following mechanisms:

- Acoustical disturbance causes mixing into the boundary layer. [55]

- Higher fluid flow velocities over the heating surface reduce boundary layer thickness. [59]

- Boiling nucleation causes the steam bubbles to "pump" or stream fluid into the boundary layer. [60]

The fluid interface thickness will also be reduced if the ratio of heat transferred by vapor transport to the heat transferred by conduction and fluid transport is increased. This can be accomplished by improving the boiling nucleation sites through the use of surfaces having optimum roughness [31] or surfaces having low surface energies. [45]

4.6.6 Variable 3f: Temperature Drop Across the Solid Interface: (Δt_s). Surface roughness of the substrate and scale on the substrate surface make definition of the exact limits of the solid interface very difficult. However, for practical purposes, arbitrary limits can be defined and measurements can then be made and used to provide a quite worthwhile understanding of the roughness and scale contributions to the heat transfer problem.

The temperature drop, Δt_s, is not an independent variable, but it is included here because it is a function of almost all of the other variables. When considering a mathematical model for scale formation, Δt_s must be included in the equations because it has a large effect upon the scaling and heat transfer problem. It can be eliminated from the final mathematical model by combining equations. It may be possible to relate the effects of the scale conditions, substrate conditions, heat flux and other variables through use of this parameter.

4.6.7 Variable 3g: Geometry. The entire boiling and flow pattern is radically changed as the geometry of the fluid volume changes. The convective flow patterns vary markedly for horizontal, vertical, and inclined heating surfaces. [59] The height and area of the active liquid as compared to the heating surface area will also alter the convection patterns. Round and rectangular surfaces, both vertical and horizontal,

where heating occurs on all or parts of the surface could also be considered. The interface thickness, the concentration within the interface, the fluid velocities, and the interface temperatures are all dependent upon a whole group of variables that are required to adequately describe all of the possible geometric situations. It would seem that each geometrical configuration must be considered as a particular case so that investigations of many cases would be required to produce a complete mathematical model.

5.0 DISCUSSION

5.1 <u>Analytical Approach</u>. An analytical model for scale buildup must consider two distinct but interacting problems:

• Nucleation and phase change of water (liquid) to steam (gas) at the evaporator surface (solid).

• Nucleation and phase change of the soluble sea salts (liquid) to scale (solid) at the evaporator surface (solid).

At the present time mathematical models for the simplest of the above systems have been developed and agree with experimental data. Incomplete and special case models, of each of the above systems have been developed. No description of a model for the combined systems has been published. It is not expected that a complete model can be developed by using the presently available technology. It is to be expected that the simpler system models can be applied one at a time so as to develop an understanding. However, additive effects may produce gross errors in this type analysis. Experiments must be conducted where each variable is isolated and independently controlled. The results when compared to the models should provide considerable insight into the interface-scale problem. The results may indicate the next steps to be taken to decrease the scaling problem.

5.2 <u>Experimental Approach</u>. An experiment that would tell how surface roughness and surface material will affect initial scale formation, the formation rate, and scale adherence, would give valuable insight into an understanding of the process. Such an experiment could also provide very practical benefits. If it were known for sure that titanium tube evaporators could operate for a period between cleanings of, say, only 10 percent longer than copper-nickel tube evaporators, a changeover to titanium would be warranted. If it were found that smooth tubes would give two weeks' extra service between cleanings, it might provide advantageous to alter the cleanining techniques to keep the tube surfaces smoother. If thin film Teflon or epoxy coatings show real advantage from a scale-forming point of view, a new thorough investigation on their ability to wear well and provide good heat transfer might be warranted. For such an investigation to provide positive results, the variables must be controlled exceptionally well. Scale is unquestionably the very weak function of surface material and roughness. Perturbations or instrument errors in the other variables will easily mask out the desired results. Since concentration factor, and salt water chemistry are exceptionally difficult to maintain over any reasonable period of time, it seems that the only way to ensure that these variables are precisely duplicated for more than one specimen, is to put several specimens into the same heat bath. Heat flux must be measured very accurately. The flow pattern must be carefully controlled so that it is

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the same for all tests of the same heat flux. Since boiling nucleation is present, the gas bubble pattern must be held uniform. This will also tax the experimenter's ingenuity.

6.0 CONCLUSIONS

From the review of literature on the state-of-the-art, it is apparent that a better understanding of the interface system variables affecting evaporator scale formation is needed. This system consists of a multiplicity of variables of the bulk fluid, the heat transfer surface, and the fluid-surface interface. Because of the large number of more or less interrelated variables, the problem of attempting an analytical model of the complete system appears to be too complex at the present time.

It is indicated that an experimental approach to a better understanding of scale formation and ahesion can be more fruitful, provided that:

● Each variable is isolated and controlled independently.

● Provision is made for adequate precision in both the control and the measurement of the variables.

● Duplication and/or uniformity of the interface (fluid-surface) variables, such as heat flux, velocity, interface temperature differences, can be assured.

● The latest developments in nucleation theory and the interface concept are considered in determining what experiments are necessary and how they should be conducted.

7.0 FUTURE WORK

Laboratory studies, along the lines of the experimental approach described above, have been initiated to establish a correlation of the system interface variables under carefully controlled conditions. Precision apparatus of bench-scale size has been setup. In the initial phase of the experimental work, the effects of surface material composition and roughness are being investigated. Other variables will be explored as the program develops.

Appendix A

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13. ABSTRACT

A survey of the literature was made to determine the state of the art in understanding the mechanism of evaporator-scale formation. The use of bulk conditions for the fluid and the heating surface does not provide a reasonably accurate model of the problem. Exploration of the variables in the interface region should provide a logical and more fruitful approach. Experimental examination of these interface variables must be made under precisely controlled conditions. Bench scale experiments are under way to establish the effects of surface material composition and roughness on scaling characteristics.

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14. KEY WORDS	LINK A		LINK B		LINK C	
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Evaporator scale						
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Interface surfaces						
Surface material composition						
Surface material roughness						