

## AN ENGINEERING APPROACH TO THE USEFULNESS OF THE LINDEMANN LAW FOR THE SETTING OF STRESS LIMITS

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### ABSTRACT

In an attempt to find a rationale ASME Code temperature limitation of metal alloys, in 1965 the writer became aware of the Lindemann Law relevance to the cohesive properties of materials. The relationship, discovered by the writer,

$$\ln \frac{\sigma}{\sigma_0} = \frac{-\nu}{\nu_M} = -\sqrt{\frac{T}{T_M}} \quad \begin{array}{l} \sigma = \text{Atomistic Load} \\ \nu = \text{Lindemann Frequency} \\ T = \text{Temperature, Kelvin} \end{array}$$

is valid to near the melting points of the elemental polycrystalline metals. From this relationship, it may be argued that an entropy change of 10.542 calories/deg. cent. is active when elemental metal is stressed beyond the threshold of indefinitely long time-to-rupture.

When the relationship is inverted it reveals the temperature of creep activation and the energetics from which various aspects of "creep damage" may be assessed. The heuristic aspect of this development is to add further strength to the "elastic billiard ball" analogy of the Compton Effect. Newtonian Mechanics might be enriched through the development of a discipline for Micro dynamics on a framework as follows:

Degree of Freedom	Definer	Entropy Change/Deg. Cent.
0	Nernst(1906)	0
I	Lindemann(1912)	10.542
II	Trouton(1884)	21.084
III	Hildebrand(1913)	31.626

## 1. INTRODUCTION

It was Lindemann himself (1) perceived a connection between the tensile strength properties of polycrystalline metal and his law of melting in a paper entitled "Some Considerations on the Forces Acting Between the Atoms of Solid Bodies." (2) In collaboration with his brother (3) a statement was appended "Note on the tensile strength of materials at low temperatures."

Lindemann hypothesized (4) that the melting temperature ( $T_M$ ) of a crystalline material is attained when the root mean square amplitude of atomic vibration becomes a certain fraction of the nearest neighbor distance. It was proposed that this fraction might be the same for all materials and Lindemann treated approximations for polycrystals. Gilvary (5) attempted to update the Lindemann Law and used data taken at the melting point. Singh and Sharma (6) applied the methods of Lattice dynamics to this problem. Shapiro (4) has made further contributions on the relation between the Lindemann Law and Lattice Dynamics.

In their "The Theory of the Properties of Metal and Alloys" Mott and Jones (7) make reference to Lindemann and incorrectly ascribe to his Law a significance not inherent to Lindemann's argument. Lindemann argues a characteristic frequency formulated by the expression:

$$\nu = C_L \sqrt{\frac{T_M}{A V^{3/2}}}$$

where  $\nu$  is the atomic volume,  $A$  the atomic weight and  $C_L$  a constant which is roughly the same for all metals. Mott-Jones argue for a characteristic temperature on the Debye concept:

$$\Theta_D = C_M \sqrt{\frac{T_M}{A V^{3/2}}}$$

Now if Lindemann's  $\nu$  is a frequency parameter and Mott-Jones  $\Theta_D$  is a temperature parameter there obviously is a dimensional discrepancy between the Lindemann view and that of Mott-Jones. Lindemann treated his oscillating atoms according to classical Newtonian dynamics, whilst Debye and earlier Einstein treated specific heats according to quantum dynamics. Lest this variance between Lindemann and Mott-Jones be treated as a wave-particle paradox, for engineering purposes we must confine our attention to the classical mechanics which motivated the Lindemann view. It would seem that the modern works of Muller by field emission microscopy and very recently by Crewe using scanning electron microscopy—that single atoms may be treated as oscillators obeying the rules of Newtonian mechanics. This should adequately qualify Lindemann's concept as a useful engineering tool. Thus it would seem appropriate to consider an approach to the usefulness of the Lindemann concept as an engineering criterion of the technical strength of materials.

## 2. LINDEMANN LAW AS A CRITERION OF ENDURANCE STRESSES

For the purposes of this discussion we will define endurance stress for the entire temperature range of engineering application of material as the maximum simple tensile stress for an indefinitely long interval of time under cyclic stressing without rupture. One hundred years ago August Wöhler studied material from that standpoint at ambient temperatures at Frankfurt-on-der-Oder. Since that time temperatures of metals applications have risen to the range for which metal structure must be engineered where Wöhler's "high-cycle" fatigue must be supplemented by elaborate and arduous studies of "low-cycle" fatigue at 10,000 stress reversals per minute may be meaningful in the so called "elastic" range it becomes meaningless in the so called "plastic" or "creep" range and "low-cycle" fatigue must supplement the understanding of "creep rate" and "time-to-rupture." The estimation of reliable "allowable stress" in the plastic range rests upon arbitrary criteria involving creep rates and stress-to-rupture time. There had been little or no philosophy to guide us over the gap between the "elastic" and "plastic" range stress ranges. Perhaps Lindemann's Law can shed some light on this problem.

In 1963 a paper by Bennewitz (8) on the shape of the log stress-long time curve on the log-time creep rupture tests was presented which seemed to indicate that under non-cyclic conditions of time-temperature-stress there does exist a stress-level in the creep temperature range where materials tend to stabilize for indefinite endurance. Bennewitz of Dieringhausen, Germany suggested representation of numerous such tests of exceptionally long duration (100,000 hours plus), sponsored by German steel makers, by semi-logarithmic plot of the creep-rupture time, whereby a similarity to the S-N or Wohler curve is apparent in that they both approach constant stress asymptotes. It is difficult to say whether current investigations of low-cycle fatigue will support the Bennewitz asymptote or the standard policy of the ASME Pressure Vessel Code's more conservative policy of rupture-time extrapolation on a log stress-log time basis.

In the 100 years since Wohler there has been vast employment of, and experience with, the Wohler philosophy of S-N curve determinations for dynamically stressed parts at ambient temperature. Since James Watt first employed metal under temperature and pressure two centuries ago, there has been much hapazard experience at elevated temperature operation under a downward "safety factor" trend from 10 to 3 under the current Nuclear Vessels (Section III) ASME Pressure Vessel Code.

Past experience suggests that adequate know-how on the use of "factor 3" concepts are urgently needed. Perhaps coordinations of existing information in the light of Lindemann might buy some such needed time in arriving at stress polices now sought through testing and costly incidents.

### 3. DISCOVERY OF LINDEMANN THE ENGINEER

As of September 15, 1965 the writer had been only vaguely familiar with the fact that there had flourished a scientist named Lindemann. The fact that Lindemann had been a significant figure in the history of the Twentieth Century for five decades until his passing in 1957 was yet an unopened book. For those interested in his career he was a most amazing individual who progressed from disciple to Walter Nernst at University of Berlin, to Research Director Royal Aircraft Factory, Farnborough, to Professor of Experimental Philosophy at Oxford, to Scientific Advisor to the First Lord of the Admiralty Churchill, later Knighted Lord Cherwell and finally became a Cabinet Member for Prime Minister Churchill, as Paymaster General.

On September 15, 1965 at a meeting of the Subcommittee on Properties of Metals of the ASME Boiler & Pressure Vessel Code Committee a discussion arose involving the proper permissible temperature range for alloys of differing base constituents. This discussion led the writer to diagram representative stress limit values for a number of materials as a function of homologous temperature i.e.

Allowable stress versus  $\frac{\text{Temperature, Kelvin}}{\text{Melting point, Kelvin}}$  (3)

Such a diagram, although showing a discernable trend toward limits at approximately similar homologous fractions seemed hardly convincing enough to influence the point at issue about consistent temperature limits.

At that point, it struck the writer that a homologous temperature comparison of the various elemental metals would have a greater rationale if done upon an atomistic force basis rather than the arbitrary stress unit in engineering. If the physicist compares atoms for their susceptibility to neutron capture on a mol cross-section basis, why should not an engineer concerned with intercomparing the cohesive forces between atoms do similarly? The proper correction was made upon the allowable stress data under consideration to permit a relative comparison upon a basis for load per mol cross-section. This is accomplished very simply by computing the ratio between the atomic weight and density of the metal involved, reduced to its two-thirds power, and using it as a factor thusly. (4)

Atomistic Load/mol section = Allowable Stress  $\left(\frac{\text{Atomic Weight}}{\text{Density}}\right)^{2/3}$

If we be permitted to reference expression (4) as Atomistic Load then a diagram such as shown in figura (I) may be obtained using the 100,000 hr. Life Stress Basis of the ASME Boiler & Pressure Vessel Code. The astonishing discovery in this diagram is that the values for elemental metals lie upon a smooth curve comprising the lower boundary. The upper boundary seems to present a hump about which cluster values for iron base, copper base, and aluminum base alloys. This diagram was communicated to the ASME-PVBC Sub-committee on Properties of Metal as information by a letter of October 13, 1965. Immediately after preparing this communication, it occurred to the writer, that it would be of interest to prepare a diagram depicting Atomistic Load as a function of Homologous Frequency which can easily argue should be the square root of Homologous Temperature. Again reploting the lower boundary of figure I diagram, log load/mol section versus Homologous Frequency the straight line coordination was discovered for elemental metal data (figure II).

It was at this point that the writer's attention centered on the Lindemann reference in the Mott-Jones (7) book which led to an appreciation of the Lindemann Law and Lindemann himself. In his book on Quantum Theory (15) he argued that a difficulty lay in the wrong things having been chosen as fundamental concepts. He argued that one of two conjugate qualities, as position and momentum or time and energy is meaningless without the other. He thought that the difficulty lay in the wrong things having been chosen as fundamental concepts—we ought to have used charge and action, not position and time. Possibly Lindemann could have succeeded in working this out had Churchill not found it necessary to make excessive demands upon his talents, then Mott-Jones (7) might have understood the Lindemann Law, and its engineering implications not so long have gone unappreciated.

But let us look further into what can be deduced from the now abundant information not available to Lindemann and which he could only surmise inductively:

Figure (3) can be expressed by an equation

$$\ln \sigma_o = \sigma_m = -\sqrt{\frac{T}{T_N}} \quad (5)$$

Now Entropy Change by definition is

$$\Delta S = \frac{\Delta H}{T} = (\ln 10)^2 R \quad (6)$$

If we write  $\sigma = \sigma_o e^{-\sqrt{\frac{RT}{RT_H}}}$

Where  $\sigma$  is allowable load

And  $\Delta S$  is Entropy Change/deg. = 10.542 cal/deg c.

Thus we now have a Trouton (16) rule that seems universal for elemental metals in long-time equilibrium with stress up to their melting points. But for the unfortunately obscure publication of the Lindemann work as a contribution to the Nernst Festschrift, page 261 (1912) it might long have been a tribute to the genius of Lindemann that he had deduced by his law and meager data an entropy value from his "considerations" on the forces acting between the atoms of solid bodies," averaging about 10 cal/deg c. very close to that which can now be deduced from the plethora of information we now have. In Lindemann's own words "constant which reminds one of Trouton's law."

The Trouton Rule originally stated that the entropy increase per mol for all normal or non-polar liquids at their boiling point to be 21 cal/deg c. However, it is modernly known entropies for liquid of polarized molecules range around 32 cal/deg. But now we are confronted with a law that represents, not the entropy of transition from liquid to gas but that of transition from the solid "elastic" state to the plastic range associated with the creep of metals. The fifteen elemental polycrystalline metals comprising the comparison of figures I and II establish a thermodynamic argument for the entropy increase of 10.542 cal/deg. c. Thus there seems to be a basic constant defining the state for which we tediously and individually establish stress limits for

codes by "long-time" tests. May a shrewd appraisal of the Lindemann principle eventually "short-circuit" some of the tedium and the departures from the base line lead to a better understanding of strengthening by alloying!

That an entropy change of 10,542 cal/deg. c. exists for 1 degree of freedom for solid metals may be surprising to those who believe the only available energy is the amount manifest upon melting. We must consider the cascaded thermo-electric systems that achieve 10% efficiency compare favorably with 30% efficiency averaged from the 3 degrees of freedom available to the power industry from steam. Moreover, it is acknowledged in thermo-electric literature, that the whole field of "size dependence" on electronic mean free path has not yet been explored.

At this point, attention must be directed to the Compton Effect, discovered in the mid 1920's. D'Abro (21) records "Compton in his original interpretation of the effect which bears his name, assimilated the collision of a photon and electron to that of two perfectly elastic billiard balls." - "This theory of the Compton effect has since been refined by the application of Heisenberg's principle of uncertainty. Nevertheless; the crude mechanical explanation on the basis of the analogy of billiard balls gives a good approximation, and we see from this example that even in the remote levels of the microscopic, far different from the level of commonplace experience, mechanical analogies may still retain a certain measure of validity."

Had Compton been aware of the interpretation that can be placed upon Lindemann's work (1910-12) he might have persisted in his initial view.

#### 6. METALLURGICAL IMPLICATIONS OF LINDEMANN'S LAW

It is clear that the thermodynamics of strengthening by alloying as manifested in figures 1, 2 and 4 may be quite complex by virtue of the containment of interstitially dissolved ingredients restricting the vibrations of the basic lattice points of the base material. Furthermore, we know that such interstitial ingredients tend to diffuse and yield phase changes in "solutionized material" upon long, persistent elevated temperature exposure in the "creep range." Generally such strengthening may be taken advantage of in the so called "elastic range" but becomes increasingly less dependable as temperature rises in the "creep range." As deformation tends to be increasingly permitted by "diffusional" adjustments the interstitial materials tend increasingly to yield phase change, generally causing deterioration and failure.

It seems reasonable to surmise that a useful "slip interference" strengthening exists on the "high-cycle" range and diffusional deterioration for the "low-cycle."

At a Gordon Research Conference, July 7-11, 1969 centered around the topic, "Creep as a Diffusionally Controlled Phenomena," an early paper by the writer was acknowledged as the first proposal that creep might be a diffusionally controlled phenomena (17) (18) (19) and that diffusional activation might be calculated from an Arrhenius type of equation using creep data (17). The equation proposed was based upon an expression used in a previous paper (20) in which a "zero stress" viscosity is estimated by plotting stress versus log flowability obtained, dividing creep rate by the stress producing it and extrapolating toward intersection to the flowability ordinate. This procedure produced parallel isotherms and was tantamount to estimating the end-point of non-Newtonian behavior, thus an end point for thixotropic behavior and of some usefulness for estimating the activation energies of self-diffusion.

It soon became apparent that for certain types of alloys the simple Arrhenius scheme (developed for liquids) yields, in some cases, activation energy calculations substantially higher than can be justified within the "vaporization" energy concept. This has led the school of Arrhenius parameter students to the factor called "activation volume" around which a controversy centered at the 1969 Conference.

The simple Arrhenius equation seems hardly adequate to explain the elevated temperature strength properties of alloys into which are solutionized "interstitially" located ingredients. Such ingredients are known generally to migrate and precipitate phases and/or allow allotropy at temperatures well below those for substantial activation probability of the lattice constituents. This suggests the need for studies systematically planned to learn the rules of deviations from the "base line" curve which we have seen fixes a natural entropy constant, - associated with the temperature ranges where degree of freedom increases from one to two.

It will readily be recognized that a polycrystalline material will have a somewhat diffuse deployment of "activation requirements" dependent upon the proximity of atoms to grain boundaries.

When the degree of freedom averages near liquidity, the Arrhenius equation yields reasonable activation energy calculations. When, as is the case with "Super Alloys" the degree of freedom hovers just above one the "activation volume" difficulty arises.

Charts are exhibited for total representation in dimensionless variables expressing cohesive aspects, as follows:

Homologous Temperature	$(T/T_M)$
Homologous Load	$(\sigma/\sigma_u)$
Homologous Frequency	$\sqrt{T/T_M}$
Homologous Wavelength	$\sqrt{T_M/T}$

Thus the question of dimensions is disposed of in a total "relativity" system conceived in terms of measurements and statistics suitable for engineering evaluations of materials.

It bears emphasis that the present paper is a proposal primarily for engineering consumption, where illustrative situations are shown in terms of Homologous Load as function of the relative aspects of temperature,

- (1) The base-line of "Metal-tivity" derived for the array of polycrystalline elemental metals as previously shown representative of sustaining 100,000 hours of "stress damage", (where the material is on the verge of a change-of-state characterized by "one-degree-of-freedom" Entropy change.)
- (2) Soft aluminum alloy (3003), a material with some manganese in solid solution generally stable and corrosion resistant up to 400°F.
- (3) A mild carbon steel pipe alloy rated to 1000°F.
- (4) Duraluminum (2024), a strength alloy up to 400°F.
- (5) Austenitic Stainless (304) rated up to 1500°F.

The plots are based upon ASME Code stresses generally used for Unfired Pressure Vessel services, presuming 100,000 hours of life before "creep damage" might dictate replacement.

A second relativity chart is appended showing "Homologous Wavelengths" versus "Metal-tivity." This transformation emphasizes the inroads of 100,000 hours of environmental effects. Only the 3003 aluminum alloy presents an aspects similar to previous charts. Curves 3, 4, 5 transform into two practically straight lines with sharp juncture, the 304 stainless steel being the sharpest. The high temperature branches are amenable to activation

energy calculations on the Dulong-Petit Law principle, yielding unlike Arrhenius Law calculations, reasonable values not to be reduced from the controversial "activation volume" consideration. After all, why should Arrhenius' equation derived to interpret 2 or 3 degrees of freedom situations, be effective when applied to 1 to 2 degrees of freedom data? In studying these charts, one discerns the instability that starts in the approach to 650°F, we note a rounding off denoting the approach to strain-aging embrittlement precipitation. Above 650°F, the graphitization" range is clearly discerned. In the case of 18-8 a sharp break defines the on-set of slow "sigma phase" formation. While true structural features should be taken into account, the curve certainly reveals the range in which activation energies may be derived and for estimating the time needed to produce various degrees of damage by introducing cracks based upon shorter or even longer reductions than 100,000 hours.

It will be appreciated that "Homologous Wavelengths" correlation suggests the driving force of time to revert metal systems to polarized matter. Since the preponderant nature of the Earth's crust is 92% oxygen by volume, this becomes a most intriguing topic for study by charting the character of "Polar-tivity" as functions of "Homologous Wavelengths" and "Homologous Frequency." Lindemann (1910) actually presented data on his Melting Point versus Frequency studies of non-metallic minerals. The analysis of "Polar-tivity," however is a subject beyond the scope of the present paper.

## 7. THE HEURISTIC QUEST

The heuristic quest of experimental philosophy for at least the century past seems beset with a teleologic "hang-up" attributable, perhaps, to a difficulty in the understanding of its own language and definitions. This is actually the situation to which Professor Lindemann addressed the only book which he ever published (15).

When we speak about frequency, we think "time per second", - conversely for Wavelength we think "seconds per time". In the case of the first instance we are unconsciously annoyed by the imminence of the disorder implied by interruption of energy flow, and entertain the concept of Entropy. In the latter instance, we feel assurance in the period-of-continuity aspect and call it available energy.

It is becoming increasingly apparent that a basic source of thermodynamic difficulty is the retention of the Absolute Zero Ideal.

More than a century ago (1854) Lord Kelvin proposed a thermodynamic temperature scale based upon the idea that a perfect gas composed of particles of zero size, when not kinetically excited would collapse into zero volume. While this might be a nice mathematical abstraction, Kelvin did not intend it to be taken literally. With reference to gases and not cryogenic solids, Kelvin pronounced his Absolute Scale "good enough for all practical purposes," - speaking for his time.

Max Planck in Treatise on Thermodynamics (22) half a century later states, "The zero of absolute temperature is that temperature at which a perfect gas of finite volume is a statement, which when applied to actual gases, has no meaning since by requisite cooling they show considerable deviation from one another and from the ideal state. How far an actual gas by average temperature changes deviates from this ideal cannot, of course be tested, until temperature has been defined without reference to any particular substance." Thus it would appear that any scheme of extrapolation to absolute zero involves in effect, an untenable infinite extrapolation.

Cryogenic studies of certain alloy systems diagram superconductive transitions at minus 0°Kelvin. An eminent laboratory studying superconductive transition of alloy systems was asked about the meaning of the concept temperature minus to "absolute zero." It was rationalized by an idea which implies the cessation of all relative kinetic activity, but that a remanence of subatomic energized states persist preventing transition to a cooperative

superconductive structure - this is tantamount to saying that although the peas are banging around inside the rattle they make no noise! - an assumption rather difficult to reconcile with the first law of thermodynamics. Thus, so far as solid state properties are concerned, the first, second and third laws are each rendered somewhat nebulous and the "black-holes" of superdense matter in space make Kelvin zero seem torrid.

So let us take a look at the score:

<u>Observer</u>	<u>Degree of Freedom</u>	<u>Entropy Change/Degree</u>
Nernst (1906)	Zero	0
Kammerlingh-Onnes (1913)	0+	0+
Kelvin (1854)	0++	0++
Lindemann (1912)	One	Approx. 10
Kanter (1965)	One	10.542 x 1 = 10.542
Trouton (1884), (non-polar-fluid)	Two	about 21 x 2 = 21.084
Hildebrand (1913) (polar-fluid)	Three	about 32 x 3 = 31.626

Thus, it may be argued that the Dulong-Petit Law (1819) yields the requisite degree of freedom only above the Debye characteristic temperature. But by the same token, similar argument should apply to Boltzman (1871) constant variability and even the variability "h" in Planck's Law which might be established if it were pursued to "absolute" zero; but it was Nernst (23) who clearly concluded that absolute zero is unattainable, wherever it is.

In the perspective of a thermodynamic reference to four degrees of freedom rather than the outmoded Kelvin Absolute Zero of Temperature, we would have a scheme as follows:

	<u>Degree of Freedom</u>
Perfect Order	0
Range of Elastic Behavior	0+ to 1
Range of Non-Newtonian Flow	1 to 2
Range of Newtonian Flow	2 to 3
Polarized Vapor	3

At current reckoning, little justification is apparent for Quantum Dynamics in the sphere of Mechanical Engineering Applications. Thus, we struggle on with Newtonian Mechanics, assured that the macro and possibly even the micro aspects of materials may be meaningfully engineered. Perhaps a new discipline may emerge from the four degrees of freedom (0, I, II, III), which could be called Micro dynamics, with no commitment to absolute zero.

<u>Degree of Freedom</u>	<u>Definer</u>	<u>Entropy Change/Deg. Cent.</u>
0	Nernst (1906)	0
I	Lindemann (1912)	10.542
II	Trouton (1884)	21.084
III	Hildebrand (1913)	31.626



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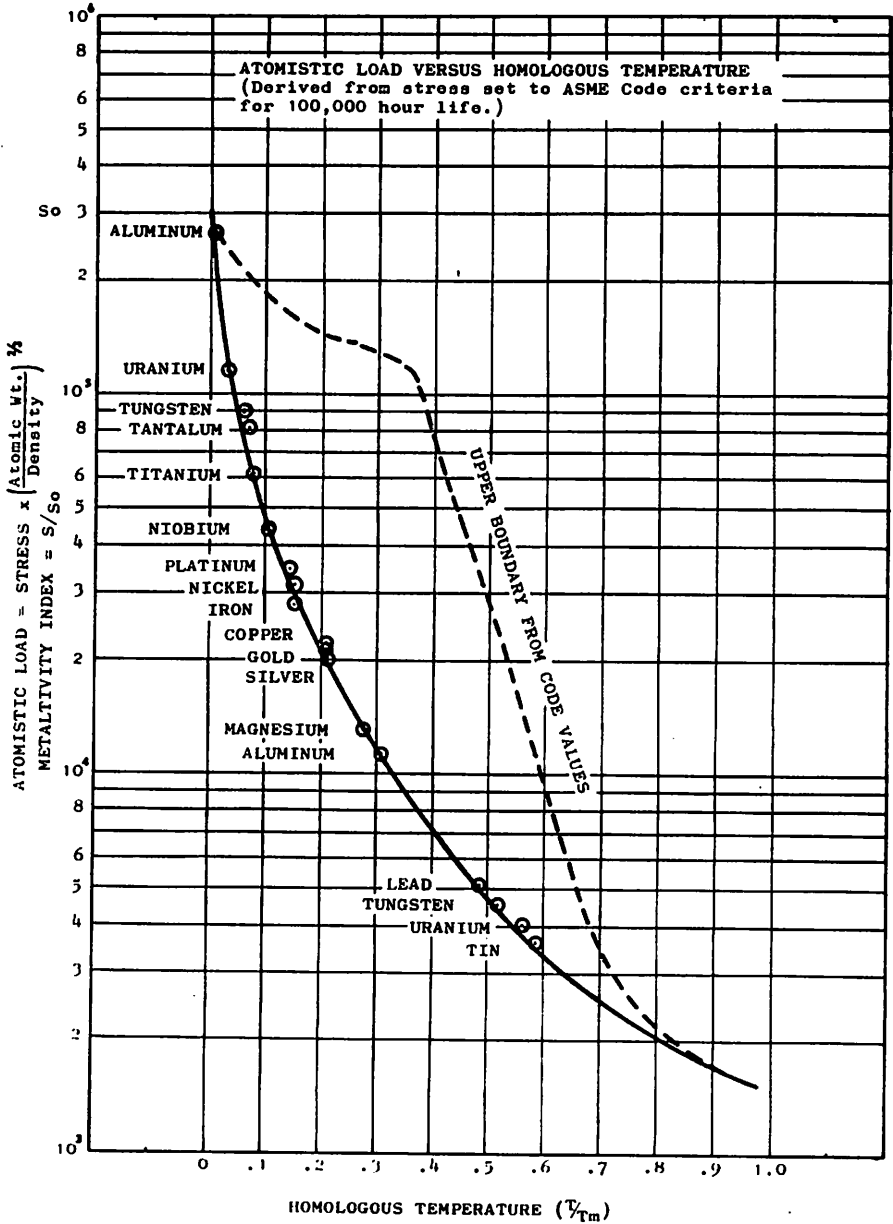


FIG. I

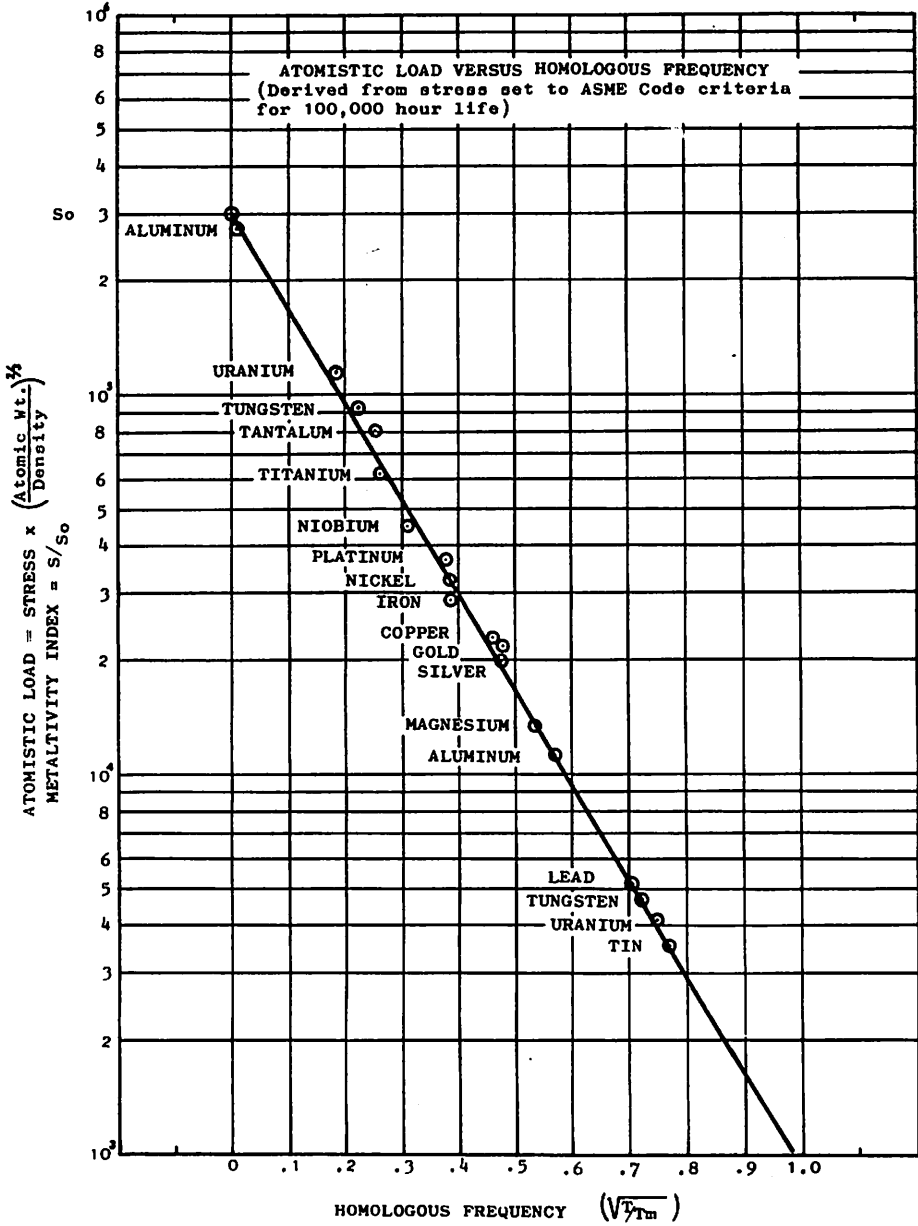


FIG. II

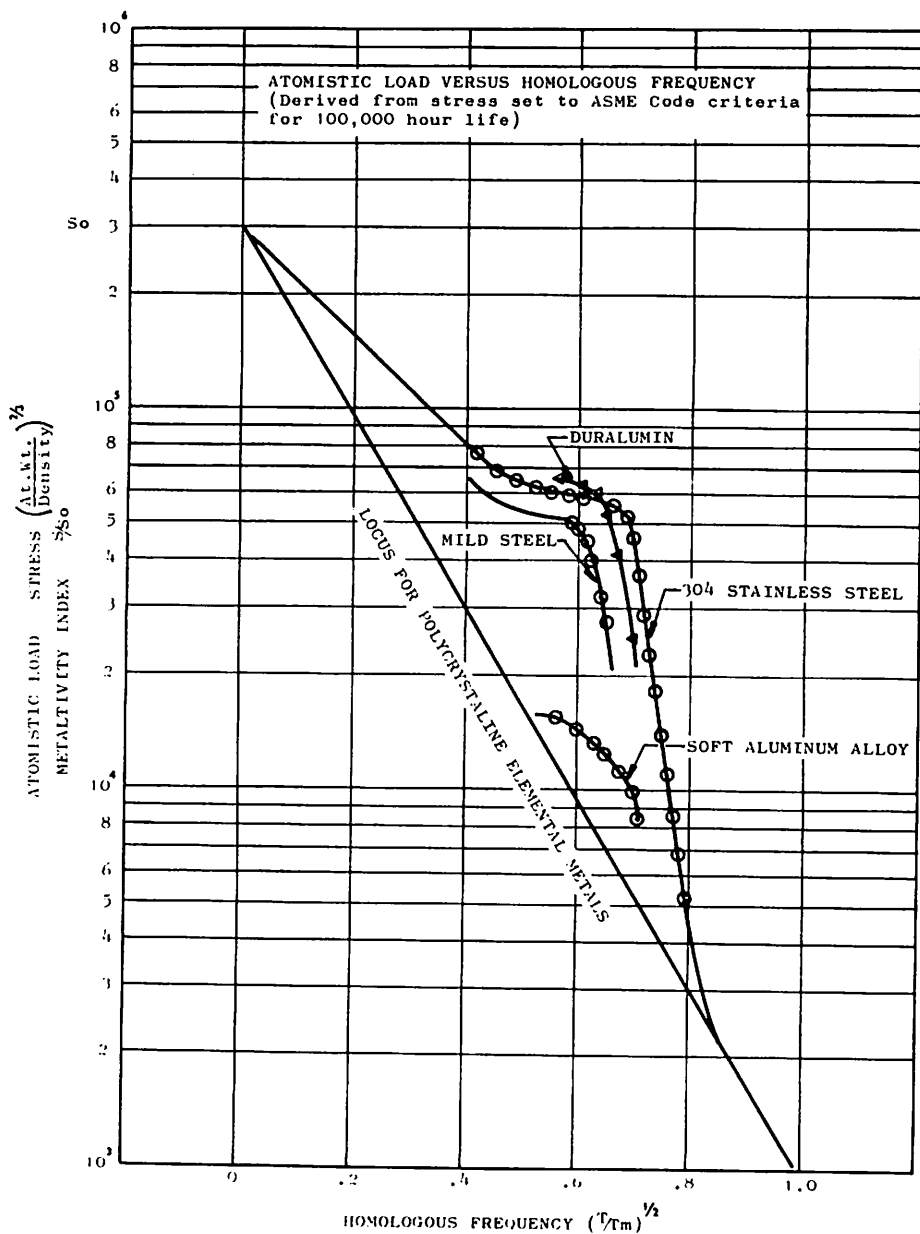


FIG. III

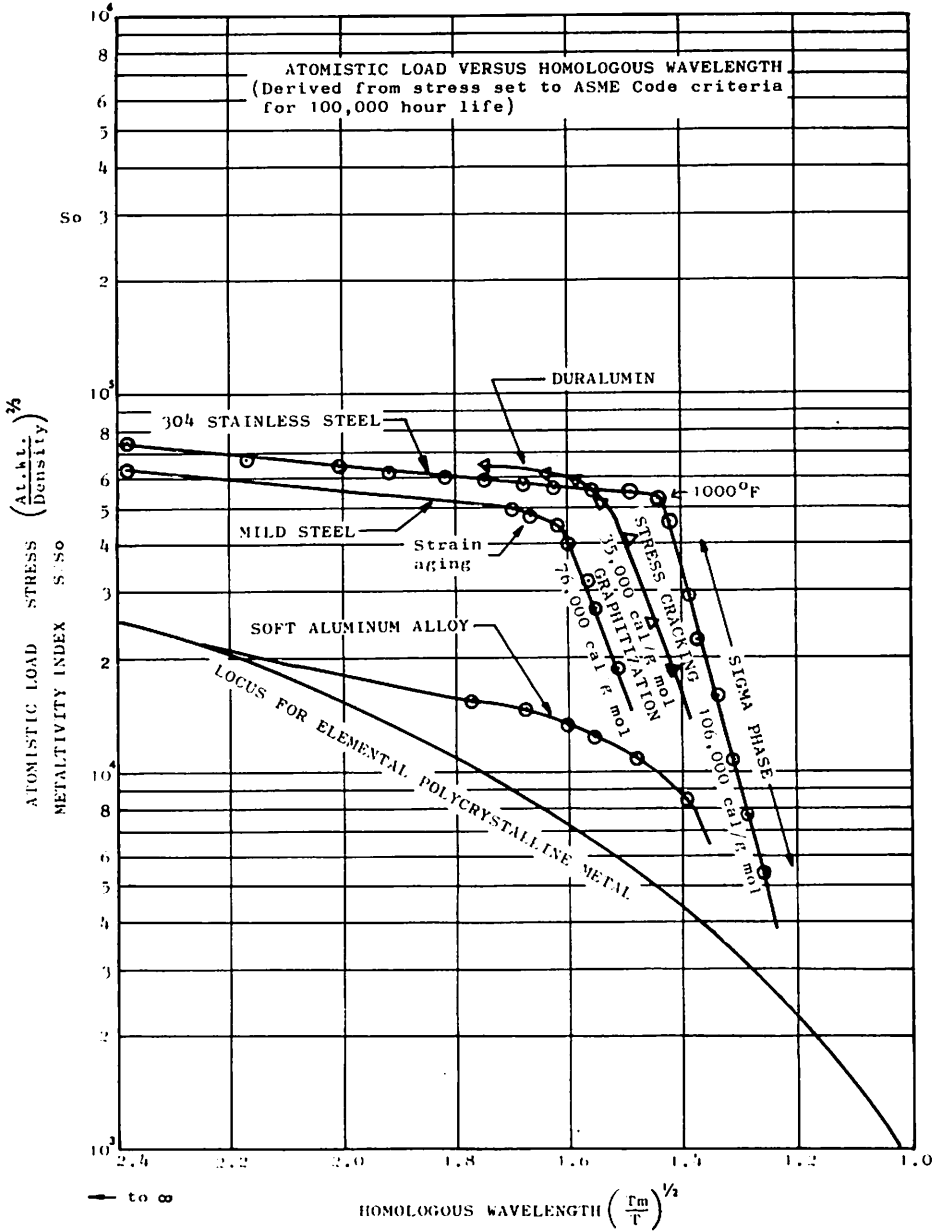


FIG. IV