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REVIEW OF PIT NUCLEATION, GROWTH AND PITTING CORROSION FATIGUE MECHANISMS

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ABSTRACT:

This paper presents a review of the state of the art developments in the pitting corrosion fatigue of aircraft structural materials. Mechanisms that govern the nucleation and growth of pitting and corrosion fatigue (CF) are briefly addressed. Some of the developments made in the author's laboratories in proposing the fretting induced pit nucleation and growth mechanisms, analysis of the hidden corrosion constituents in fuselage joints and pitting corrosion fatigue crack growth (PCFCG) model are elucidated. An epistemology of the topic is presented which will be of assistance to the community working in this area.

INTRODUCTION:

Corrosion of engineering artifacts, how corrosion nucleates and at what rate it grows, is a very important issue in the life estimation and possible life extension of engineering components. Even though design for fatigue durability and damage tolerance have improved markedly over the past 30 years or so, the design for environmental fatigue

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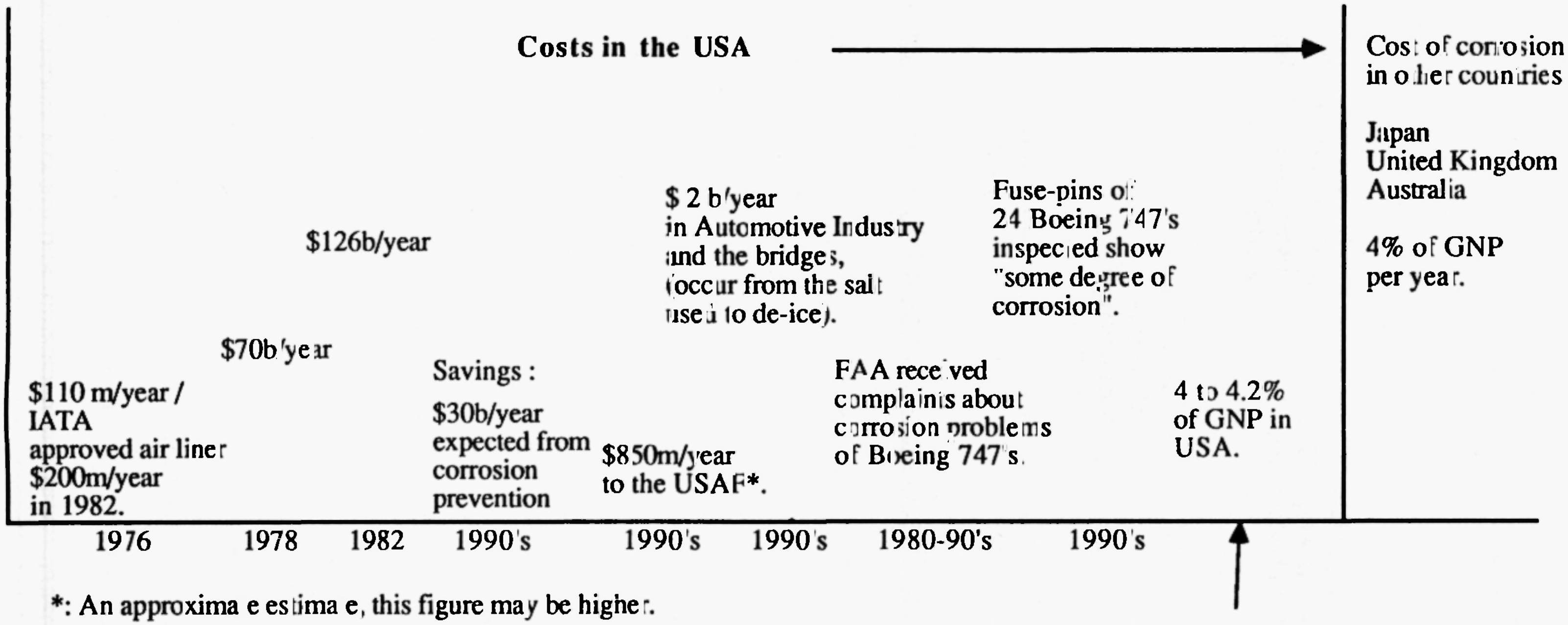
(corrosion fatigue) has received limited attention. Thus, consideration of environmental effects is a difficult question the engineering research and design communities face today. This paper attempts to unite the knowledge of chemistry of corrosion with engineering aspects of crack and corrosion fatigue crack growth in aluminum alloys.

Corrosion is a major concern in the aerospace industry today (1). The costs of corrosion and prevention methods are likely to increase with time. These are surveyed from different published sources and are shown in the "time scales" of Fig. 1. Though exact estimate of the costs of corrosion is very difficult to obtain, the estimates provided in Fig. 1 are based on published sources. Recent visits to commercial aircraft maintenance facilities have indicated that unscheduled maintenance costs of corrosion range from 10 to 50% depending on aircraft, operator, flight activity etc. It is interesting to note that the understanding of corrosion has increased in the last three decades, yet the costs of corrosion is increasing from 2 to 3 times every decade. Several means were developed to retard the corrosion damage by following prevention systems. When corrosion prevention systems failed in certain systems it resulted, in some cases, in lives being lost. Such accidents are many, however, a few examples are recorded in the "time scales" of Fig. 2. Undoubtedly, much more information will emerge on this issue. The next section briefly presents some background on corrosion.

TYPES OF LOCALIZED CORROSION:

The following types of localized corrosion are observed in the case of aircraft structures, namely;

1. crevice,
2. pitting,
3. exfoliation,
4. filiform,



*: An approximate estimate, this figure may be higher.

Fig. 1. Estimate of costs of corrosion (surveyed from published sources).

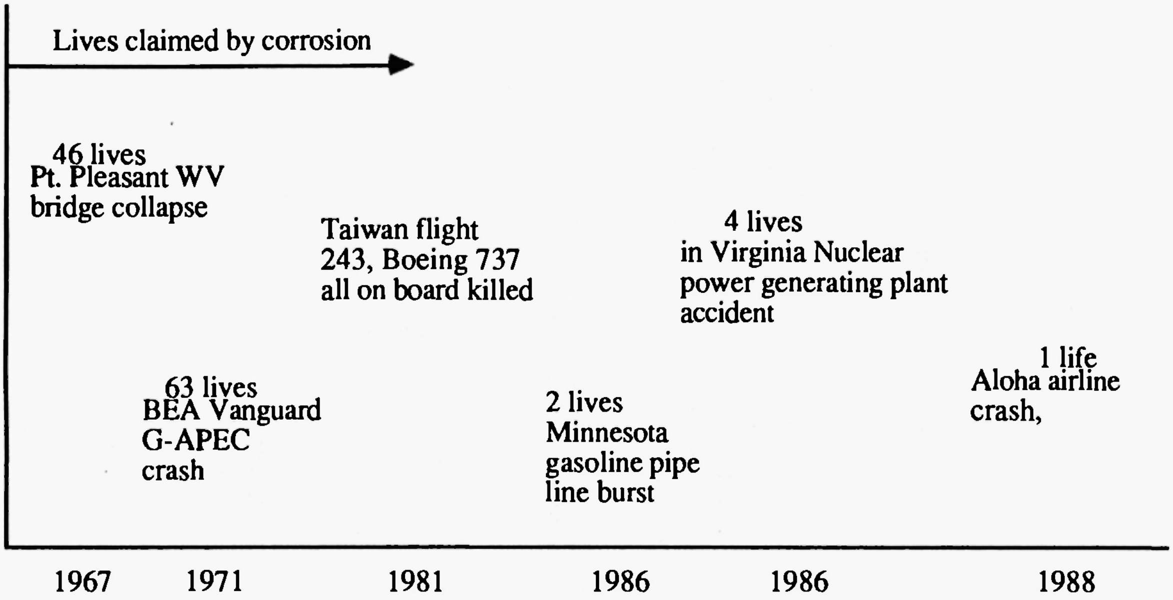


Fig. 2. A few examples of failure in the corrosion prevention systems that claimed lives.

5. etch corrosion,
6. Galvanic corrosion,
7. intergranular corrosion,
8. microbial corrosion,
9. stress corrosion,
10. hydrogen embrittlement,
11. metallic mercury corrosion of aluminum alloys,
12. fretting corrosion, and
13. other types.

Many of the above corrosion mechanisms have been investigated independently, yet not much is known on how a particular mechanism dominates in the structures of an aircraft. Since aircraft structures are loaded, how loads interact with the corrosion mechanisms is beyond the scope of this paper. Also, the nucleation and growth kinetics of the corrosion mechanisms summarized above are not yet fully investigated independently and also in interactive modes in the presence of fatigue or cyclic loads. Therefore, it is prudent to assume that the corrosion will form nuclei at a localized region of existing mechanical damage such as slip systems (they act as corrosion tunnels), hard particles, material discontinuities and where the protective films are ruptured. Corrosion also can form nuclei by Galvanic* attack. It is speculated that corrosion mechanisms transfer from one to other mechanisms e.g., a Galvanic attack may nucleate a pit and a pit may become a crack or convert to some other corrosion type e.g., exfoliation. From the regions of pits, intergranular type of corrosion may occur and from intergranular corrosion, stress corrosion cracking may result. Other interactive mechanisms may be hydrogen or other cell concentrations (chemical and/or biological) that may nucleate crevice corrosion and filiform respectively. The growth of filliform corrosion is accelerated in the presence of microbial or fungus species.

PARAMETERS INFLUENCE THE LOCALIZED CORROSION:

There are numerous parameters that influence the localized corrosion nucleation and growth rates. Some of these parameters were listed by Oldfield and Sutton (2) for crevice corrosion that were modified in this paper to make them more general for other types of corrosion. They are:

Alloy composition	major constituents, minor additions, and discontinuities their density, size and types.
Electrochemical reactions	metal dissolution, oxygen reduction, hydrogen evolution, other reduction reactions, and potential drop.

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*Luigi Galvani proposed electrochemical action of two electrical conductors, this process is known as Galvanic reaction and in corrosion "Galvanic" corrosion.

Bulk solution composition	Cl^- content, O_2 content, pH, and pollutants.
Bulk solution environment	temperature, agitation, and volume.
Composition of media solution	hydrolysis equilibria, reaction rates, activities, and corrosion products.
Passive film characteristics	passive current, film stability, parameters of passivation and repassivation, time of repassivation or otherwise, and material parameters.
Mass transport phenomenon	migration, diffusion, and convection.
Geometry considerations	exterior and interior surfaces for corrosion attack, area ratio, and number and shapes of attack.
Characterization of corrosion sites	surface, hidden, and characterization of parameters 1 through 8 at the hidden sites.
Corrosion geometry	depth, density, volume loss, and gap between sites of corrosion.
Corrosion characterization	crevice, pitting, exfoliation, filiform, Galvanic, intergranular, fretting corrosion, etch corrosion, microbial, stress corrosion, metallic mercury corrosion, and hydrogen embrittlement.
Media characterization	metal/metal, metal/non-metal, metal/marine growth, Galvanically protected, applied potential, and hydrogen characterization.

In order to develop models of localized corrosion nucleation and growth kinetics for different corrosion types, the above parameters must be known.

PIT NUCLEATION AND GROWTH MECHANISMS:

A pit is defined in the Webster's new universal unabridged dictionary "as a hollow or indentation in a surface". Since distribution of pits largely depends upon the density and number of constituent particles present on the surface, they often number in several thousands in an area of one mm^2. Therefore, localized pitted regions transform to more deeper material losses or in the form of exfoliation corrosion. In the case of aircraft structures, pitting is observed as a major contributor to the corrosion damage. Therefore, how pitting occurs, its nucleation and growth mechanisms need to be understood while developing corrosion prevention methods.

A review of pitting corrosion has been elucidated in this paper in terms of electrochemical aspects, pit nucleation and pit growth mechanisms. Later mechanical fatigue concepts were combined with the corrosion, resulting into pitting corrosion fatigue process.

Electro-chemical Aspects in Corrosion:

Luigi Galvani (3), in 1791, published a discussion of electrochemical actions of two electrical conductors, known as Galvanic reaction. For almost 100 years, no significant development occurred with "Galvanic reaction". In 1904, Tafel (4) presented an equation to describe the variation in the rate of reaction with over potential. A breakaway oxidation that resulted in localized corrosion attack was proposed by Pilling and Bedworth (5). Combining these two concepts, Evans (6) presented a linear relationship that represented the rate of corrosion by corrosion current density. Wagner and Traud (7) studied the electrode kinetics while Pourbaix (8) presented potential versus pH diagram. This diagram is well known to describe the onset of passive film formation and it is also known as isothermal phase diagram. Activities in the study of localized corrosion is presented (9-16) in the time scales of Fig. 3.

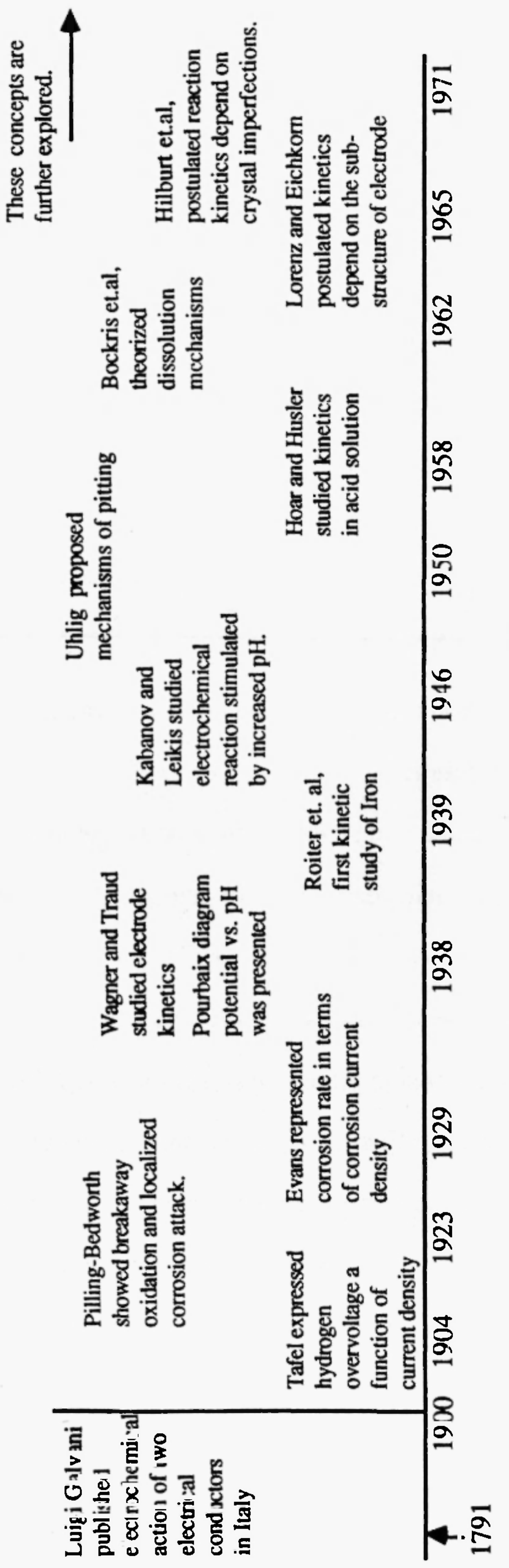


Fig. 3. Electrochemical aspects in corrosion.

Pit Nucleation Mechanisms:

The mechanisms of pit nucleation processes are shown in the time scales of Fig. 4. They are of the following types:

1. Adsorption and Adsorption Induced Mechanisms: Most mechanisms of pit nucleation consider the adsorption of aggressive anions at energetically preferred sites. Developments made in the adsorption based mechanisms (17-24) are presented in the time scales of Fig. 4. They are based on either competitive adsorption or complex ion formation on the surface. In this framework, continuous Cl^- anions and passivating agents are adsorbed. Above a critical potential, Cl adsorption was favored, which result in a breakdown of passive film on the surface, allowing pit formation.

2. Ion Migration or Penetration Theories: These mechanisms (25-27) require either penetration of damaging anions from the oxide/electrolyte interface to the metal/oxide interface or migration of cations or their respective vacancies as a decisive process. According to the penetration theory, aggressive anions adsorbed on the oxide film enter and penetrate the film when the electrostatic field across the film/solution interface reaches a critical value corresponding to the critical breakdown potential. Thus a discontinuous oxide film is produced which is much better ion conductor than the original passive layer. Rapid cation egress and pitting proceeds.

3. Mechanical Film Breakdown Theories: Mechanical breakdown of the passive film (28-29) is an independent as well as combined phenomenon with the above two frameworks. Breakdown of the passive film provides the electrolyte direct access to the base metal. Pitting occurs. Developments made in these mechanisms are described in the time scales of Fig. 4.

Pit Growth Mechanisms:

Four mechanisms by which a pit grows are described in the time scales of Fig. 5. They are

PIT NUCLEATION MECHANISMS

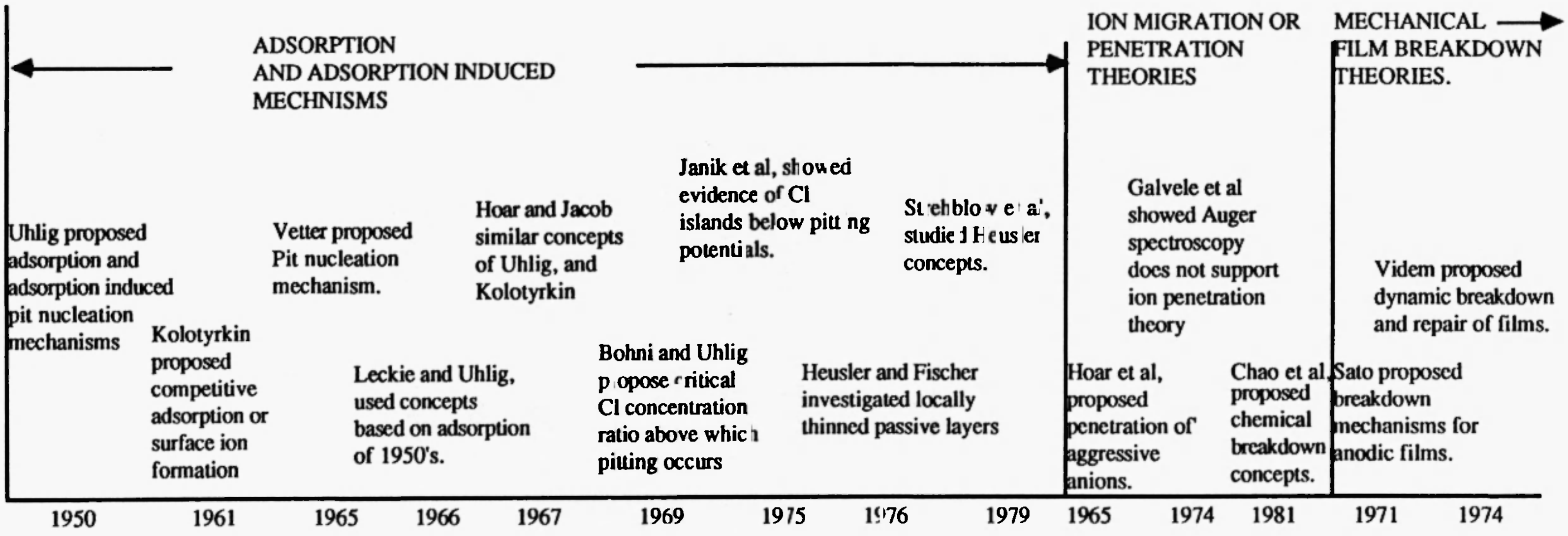


Fig. 4. Summary of pit nucleation mechanisms.

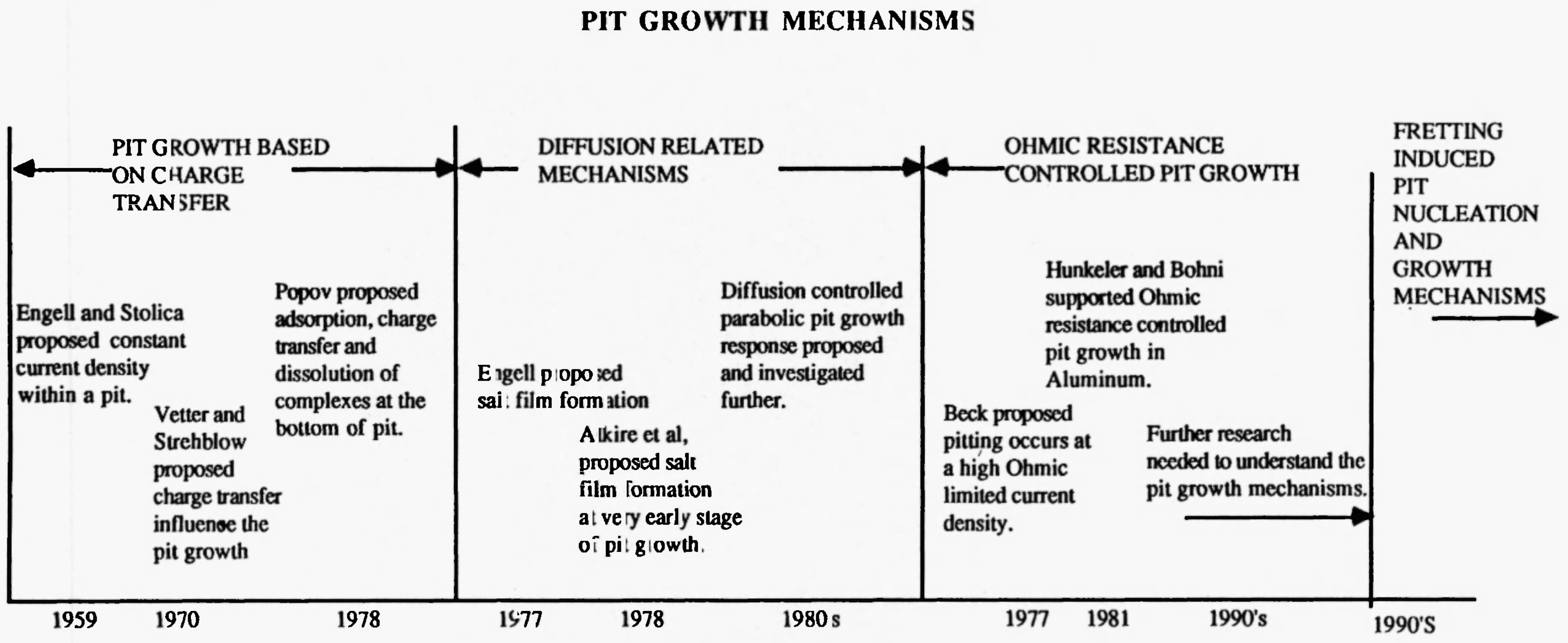


Fig. 5. Summary of pit growth mechanisms.

as follows:

1. Charge Transfer Based Pit Growth: The early stages of pitting occur due to the high current density present at the bottom of a pit. However, experimental validation of this theory has become very complex issue. As a result, constant current potentials within pits, are a widely accepted concept in the literature (30-32).

2. Diffusion Related Mechanisms: The presence of a salt layer on the surface of passive materials results in the breakdown of the passive film (33-35). Solubility of salt in aluminum alloys is quite high where salt precipitates on the metal surface. Mechanical properties of exposed, aircraft structural materials, are expected to deteriorate by this mode as active salt powder, white powder shown in Fig. 6, have been documented in the tear down corrosion investigations of a C/KC 135 aircraft. The white powder was analyzed using the energy dispersive X-ray analysis. Among various constituents, Ca, Cu, Cl and traces of Zn were observed. Photomicrograph shown in Fig. 6 is the white powder on the corroded plate, removed from a C/KC 135 aircraft, whereas Figs. 7 and 8 show the

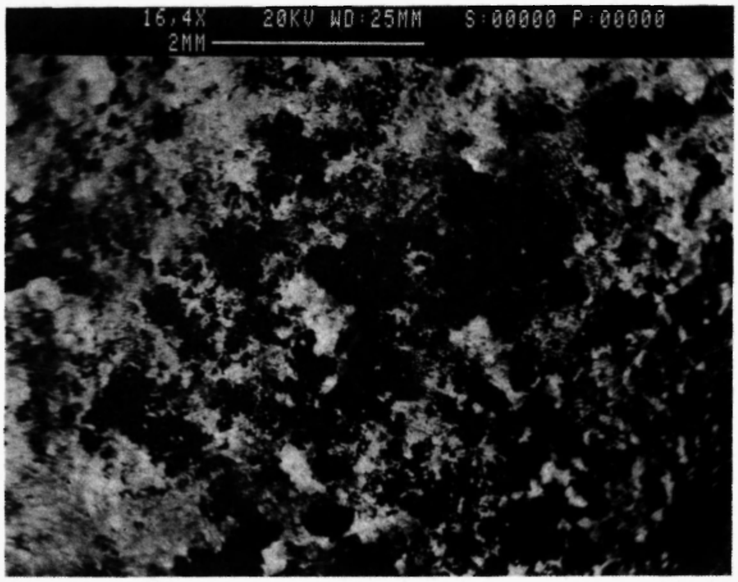


Fig. 6. White powder (salt) deposition on fuselage.

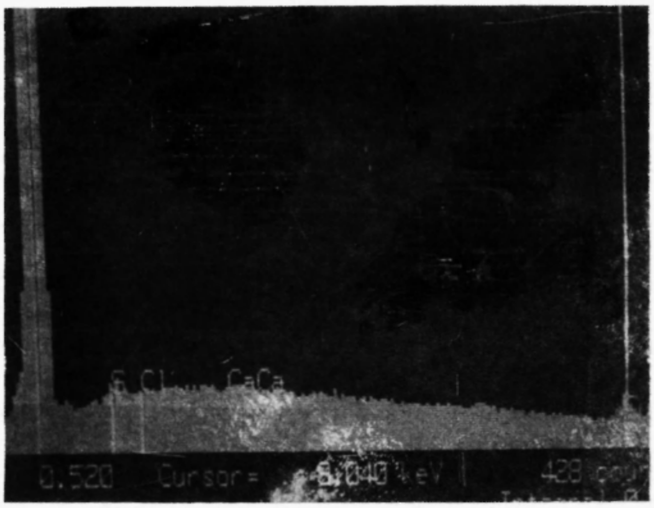


Fig. 7. Energy dispersive X-ray analysis of the white powder at a location of Fig. 6.

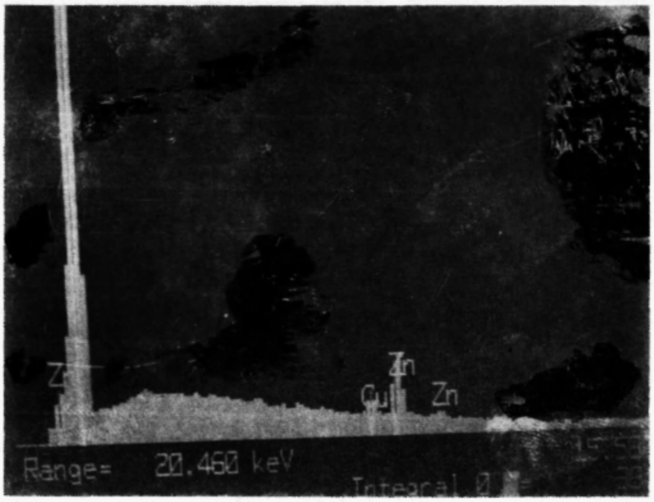


Fig. 8. Energy dispersive X-ray analysis of the white powder at a location of Fig. 6.

analysis of white powder, from a location from Fig. 6. It may be noted that this white powder is as a result of cladding which with given time results in the formation of white powder.

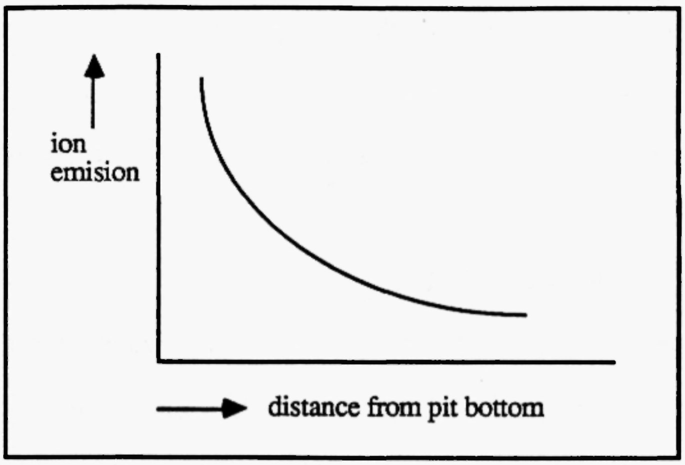
3. Ohmic Resistance Controlled Pit Growth: Pitting in titanium and aluminum alloys occurs because of high Ohmic-limited current densities (36-37). The rate of pit growth also depends upon the time of exposure and applied potential. These models are shown in the time scales of Fig. 5.

4. Fretting Induced Pit Nucleation and Growth Mechanism: Goswami and Hoeppner (38) proposed a fretting induced pit nucleation and growth mechanism of aircraft structural materials. Engineering applications such as bolts, rivets, couplings and other specific joint configurations, for example, firtree and dovetail roots in a gas turbine disk and blade attachments; relative displacement of the order of a few nanometers to as high as several millimeters occurs in the joint. Under these conditions, the contacting surfaces produce a normal pressure on the line of the actual load direction. This action results into fretting. Fretting process involves the reaction between two contacting surfaces that result in asperity removal called debris. Often these debris are corroded that nucleate pits. Once, the pits are nucleated, the repeated fretting produces pit growth.

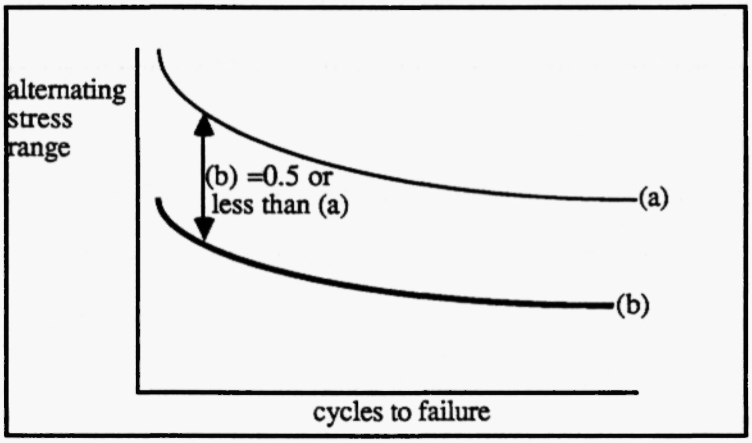
HYDROGEN EVOLUTION AND DEGRADATION:

The concentration of hydrogen increases at the sites where there are triaxial state of stresses or at the sites where there are stress or strain concentrations. Hydrogen electrons enter at three dimensional levels in transition metals and increase the concentration of electrons on that level, thus causing the lattice cohesion to weaken. This facilitates the nucleation and growth of corrosion and cracks (in sustained load conditions) even at the atmospheric hydrogen pressure or in the presence of moisture.

Innumerable studies were made to investigate the effects of hydrogen on the strength and other mechanical properties, where hydrogen was found to degrade the material properties. Hydrogen also was found present at the bottom of a pit. Presence of hydrogen at the bottom of a pit was recorded in terms of secondary ion emission. A trend is shown schematically in Fig. 9 (a) which shows that as the distance from the pit bottom increases, the emitted intensity of the ions decreases. Fig. 9 (b) shows effect of hydrogen



(a) schematic representation of secondary ion emision from pit bottom.



(b) fatigue behavior of off-shore steel, in (a) no hydrogen and (b) hydrogen.

Fig. 9. Schematic behavior showing effect of hydrogen in a off-shore steel.

on steels where alternating stresses, for the same cyclic life, reduced more than one half in the presence of hydrogen than that of the material without hydrogen.

When the temperature was increased, even at lower ranges (39) from 20 to 90°C, the solubility of hydrogen increased in typical metals. Quickly, the hydrogen concentration became critical in the region of triaxial stress states. Higher crack growth rates, even at this temperature range, was due to the activation energy for stress corrosion crack propagation which became equal to activation energy for hydrogen diffusion. Therefore, these hydrogen mechanisms cause a deleterious effect on the corrosion and/or corrosion fatigue crack growth rates.

AIRCRAFT CORROSION FATIGUE:

The corrosion fatigue is a process in which gradual accumulation of damage takes place in a material subjected to repeated stresses in corrosive environments. Thus, corrosion fatigue is a failure mechanism of engineering components in which the damage as a result of synergisms between fatigue and environment is accelerated. Therefore, failure of components in such situations occurs prematurely in the low cycle fatigue regime. The growth of damage may be in terms of cracks, embrittlements and localized material losses (e.g., pitting, exfoliation, etc.).

An aircraft structure is likely to deteriorate as the flight cycles are applied. The loads and environments in which an aircraft operates decides the rate of damage growth. Growth of corrosion damage depends upon material as well as many other issues related to manufacturing, design procedures, tooling, and assembly practices employed. If the corrosion attack occurs on structurally significant items (SSI's), the consequences can be very difficult to address. Since corrosion either accelerates other forms of damage such as fatigue, fretting and other processes, this results in lowering the structural integrity of the aircraft structure. The corrosion problems will appear in an aircraft depending upon the type of aircraft, operating environment and usage. A typical example of corrosion prone areas in a transport aircraft is shown in Fig. 10. These are under de-icer boots, floor support and flooring, passenger, cargo and crew doors, galley areas, areas in the path of exhaust gases, integral fuel tanks, toilet areas, and battery areas.

Corrosion fatigue studies of aircraft structural materials commenced in the late 1960's.

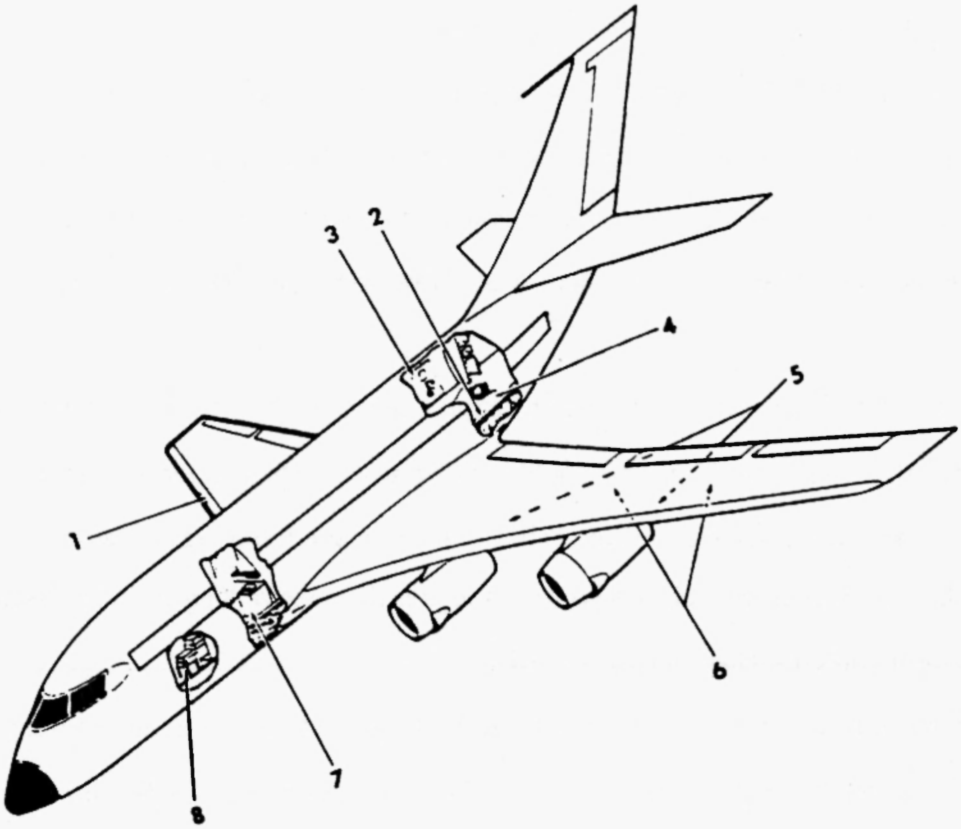


Fig. 10.Corrosion susceptible areas in a small transport aircraft.
1. Under deicer boots. 2. Floor supports and flooring,
3. Passenger, cargo and crew doors, 4. Galley areas, 5. Areas in the path of exhaust gases. 6. Internal fuel tank. 7. Toilet areas. 8. Battery areas.
(Courtesy of Canadian Department of National Defence)

Fracture mechanics modeling methods in corrosion fatigue were first proposed by Brown (40). Later Wei and Landes (41) proposed a superposition model (linear summation of crack growth by plasticity driven fatigue and crack growth by chemical reaction). Within this model, the rate of hydrogen assisted fatigue cracking was predicted by adding two contributory components; 1) crack growth by mechanical component in an inert environment, and 2) that of the crack growth by static load, at similar stress intensity, below:

$$(da/dN)_{mechanical} = A\,(\Delta K)^n$$

$$(da/dN)_{environment} = 1/f\,(da/dt)_{environment} \qquad (1)$$

Austin and Walker (42) argued the superposition model and suggested that the two processes are competing with one another and are not superposition, instead, competition mechanisms. Wide body of the corrosion fatigue data were generated and assessed with the superposition model, however, there were mixed opinions due to the following (43-44):

1) It did not account for the corrosion fatigue crack growth at K_{max}, below K_{ISCC}. Where the latter term is the threshold limit of mode I stress intensity factor range in stress corrosion cracking. In some alloys, the opposite trend was observed where the crack growth at levels below K_{ISCC} was higher than above K_{ISCC}. This discrepancy was more difficult where $K_{ISCC} = \Delta K_{IC}$.
2) According to the superposition model, at the K_{max} level corresponding to K_{ISCC}, the diagram should show a shift in the curve. However, such shifts were seen very seldom, only under the hydrogen gas testing. Such a behavior, where occurred, the shifting in the curve resulted below K_{ISCC}.
3) As the superposition model provides qualitative evaluation of the effect of loading frequency, such evaluations were unreal of a component loading waveform. As a result, effect of different waveforms were not explored.

Therefore, no single model was statistically proven to the degree where it can be used in design for life assessments of aircraft structures. However, several attempts were made to develop such concepts. Several defense and civil aircraft operators formulated a discussion of corrosion problems at this time (45) and the first international conference on corrosion fatigue held in 1971.

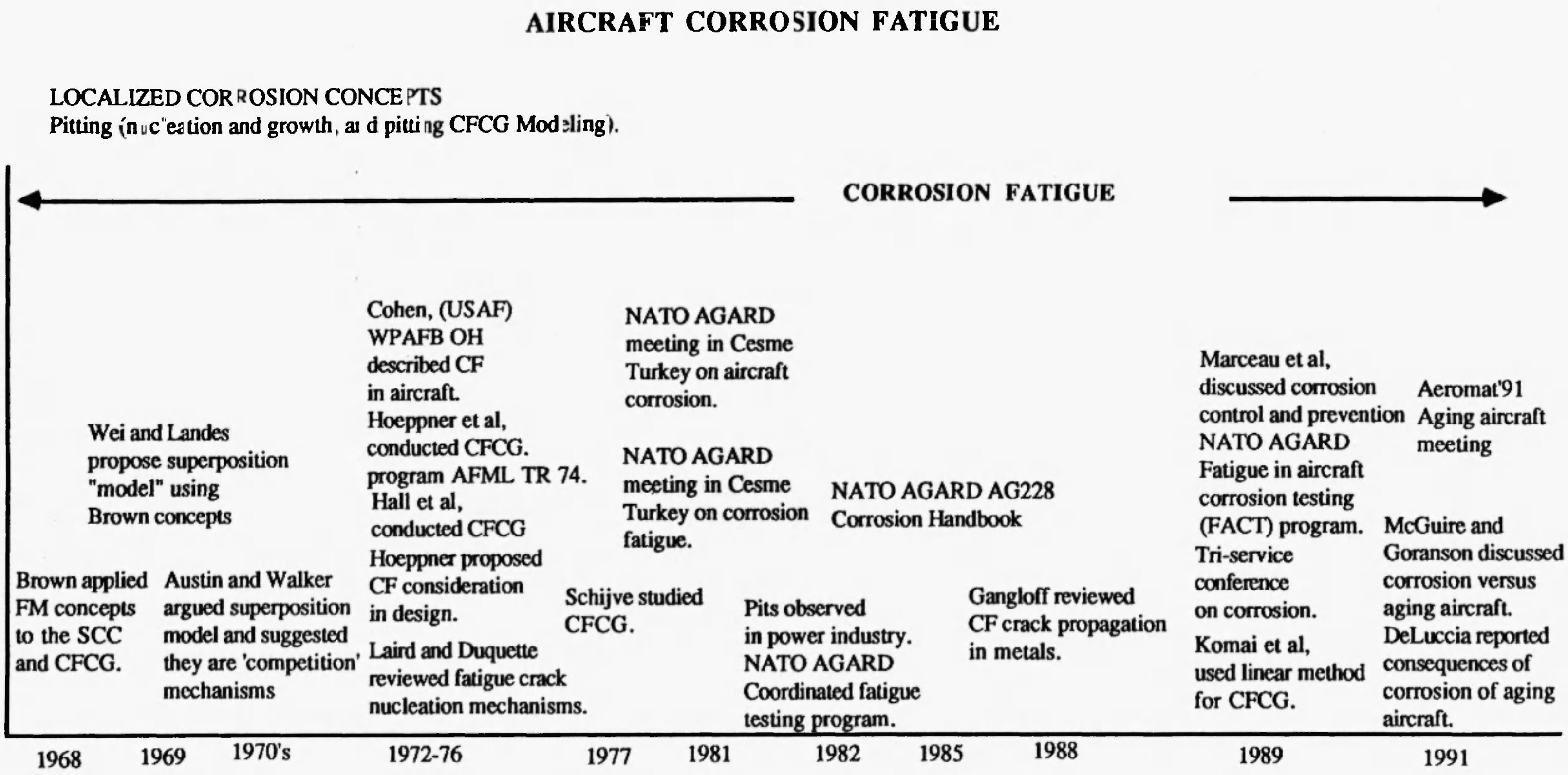
The role of localized corrosion in the acceleration of fatigue crack growth (FCG) was conceptually presented by Hoeppner (46-48) and corrosion fatigue crack growth studies were conducted on a series of aircraft structural materials. A parallel effort in the corrosion fatigue crack growth rate (CFCGR) determination was made by Hall et al (49). Several review articles (50) were written during this time as shown in the "Time Scale" of Fig. 11. This work was a major undertaking by the USAF in the late 1960's and early 1970's to increase our understanding of environmental effects on fatigue crack growth.

Several attempts were made by the (46-48) Advisory Group for Aerospace Research and Development, Structures and Materials Panel in a NATO effort to investigate corrosion fatigue. Several round robin test programs were initiated as shown in Fig. 11. In recent years more and more concerns were expressed by aircraft operators who either experienced premature failures or intend to extend the design life of their aircraft fleets. Several such interests are reflected in references (51-62).

When the areas of aircraft corrosion fatigue (CF) and CFCG were developing, practically no attempts were made to address the effect of localized corrosion such as pitting and its transformation into CF or fatigue crack growth processes. Hence, there is a need to further develop localized corrosion mechanisms and concepts such as pit nucleation and growth. Once these mechanisms are understood, they can be combined with the fatigue and crack growth concepts. Such data can be synthesized in the further development of probabilistic models that can be used in the design, maintenance and setting the inspection intervals for existing fleets. The consideration of corrosion and corrosion fatigue can be included in the aircraft structural integrity or damage tolerance analysis.

PITTING CORROSION FATIGUE:

The pit nucleation, growth and corrosion fatigue mechanisms are very important in developing localized corrosion fatigue concepts. Pit growth rate is accelerated by many factors such as microstructure, discontinuities present, film composition, (coherence and mechanical properties of the film), previous plastic deformation etc., the quantitative



AIRCRAFT CORROSION FATIGUE

LOCALIZED CORROSION CONCEPTS
Pitting (nucleation and growth, and pitting CFCG Modeling).

CORROSION FATIGUE

Brown applied FM concepts to the SCC and CFCG.

Wei and Landes propose superposition "model" using Brown concepts

Austin and Walker argued superposition model and suggested they are 'competition' mechanisms

Cohen, (USAF) WPAFB OH described CF in aircraft.
Hoeppner et al, conducted CFCG. program AFML TR 74.
Hall et al, conducted CFCG
Hoeppner proposed CF consideration in design.
Laird and Duquette reviewed fatigue crack nucleation mechanisms.

Schijve studied CFCG.

NATO AGARD meeting in Cesme Turkey on aircraft corrosion.

NATO AGARD meeting in Cesme Turkey on corrosion fatigue.

Pits observed in power industry.
NATO AGARD Coordinated fatigue testing program.

NATO AGARD AG228 Corrosion Handbook

Gangloff reviewed CF crack propagation in metals.

Marceau et al, discussed corrosion control and prevention
NATO AGARD Fatigue in aircraft corrosion testing (FACT) program.
Tri-service conference on corrosion.
Komai et al, used linear method for CFCG.

Aeromat'91 Aging aircraft meeting

McGuire and Goranson discussed corrosion versus aging aircraft.
DeLuccia reported consequences of corrosion of aging aircraft.

Fig. 11. Summary of corrosion fatigue research since 1968.

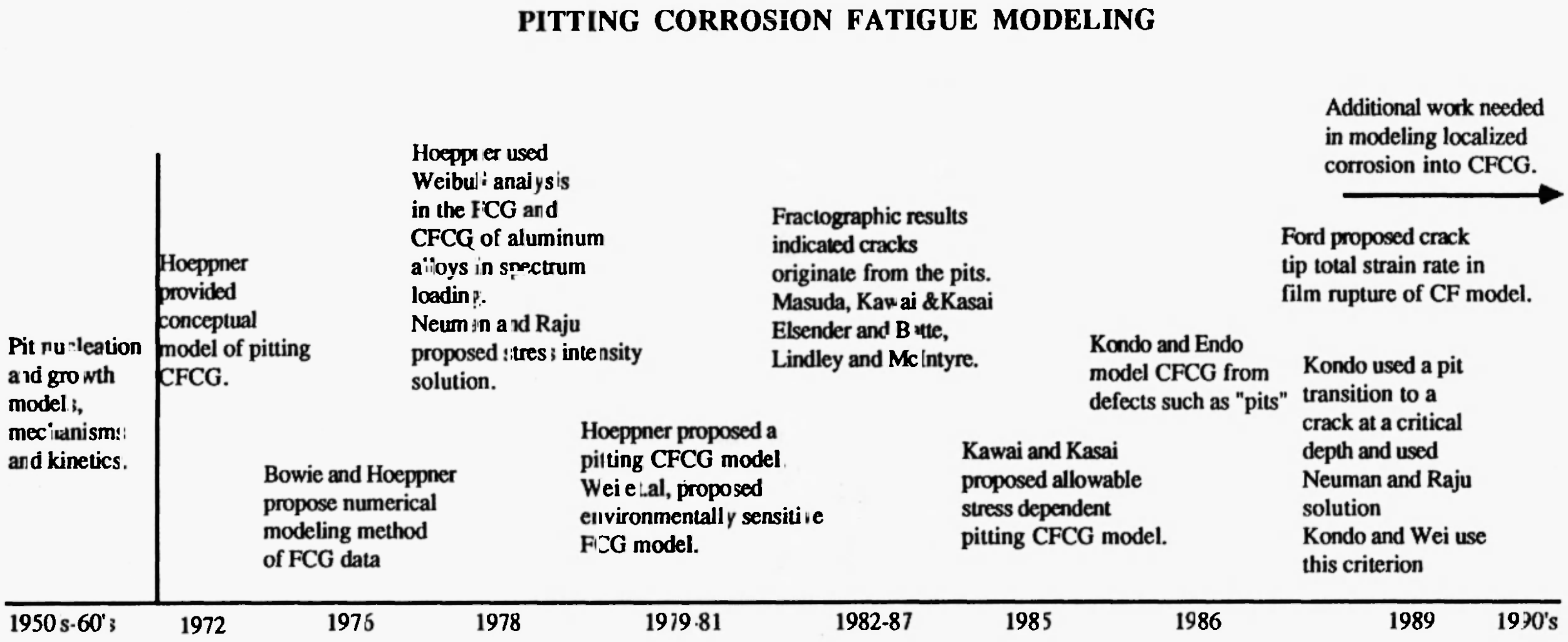
estimations of growth rates are very difficult.

Once the pits form, their shapes are an additional consideration. In the literature pits were assumed as hemispherical. However, microstructural influences may generate pits in different shapes. In the authors' laboratories deeper channel shaped pits were documented in some aluminum alloys that may act as a crack. Currently, the criteria for a pit transition to a crack is much in debate and no single, analytically valid, criterion has emerged. Development of a criterion depends upon many factors e.g., the constituent particle distribution, their size and density, stress range, frequency and waveform used. Hence no criteria can be generalized and used to model pitting corrosion fatigue crack growth rates. Various developments made in this area of research has been presented in the time scales of Fig. 12 and in the references (63-75). The physics of pitting, fatigue, and pitting corrosion fatigue (PCF) are elaborated in much detail else where (76).

Hoeppner (66), proposed a pitting CFCG model of aluminum alloys. As the pit growth kinetics are not yet understood properly, when to consider a pit a crack is still a point of great concern. Hoeppner, used a power law pit growth model, where the exponent described the linear, parabolic and cubic growth rates respectively. A four parameter Wiebull equation was used to fit the CG data. The equation employed in ref. (66) has the following form:

$$1-\frac{\Delta K}{K_{Ib}} = \exp.\left[-\left\{\frac{\log_e(\frac{da}{dN}+1)-e}{v-e}\right\}^k\right] \qquad (2)$$

For a decade no other model was put forward in the pitting CFCG modeling. Figure 12 compiles the models (63-75) that have been developed. Recently Kondo (73-74) used a pit transition to a crack criterion and crack growth modeling within the linear elastic fracture



PITTING CORROSION FATIGUE MODELING

Pit nucleation and growth models, mechanisms and kinetics.

Hoeppner provided conceptual model of pitting CFCG.

Bowie and Hoeppner propose numerical modeling method of FCG data

Hoeppner used Weibull analysis in the FCG and CFCG of aluminum alloys in spectrum loading.
Neuman and Raju proposed stress intensity solution.

Hoeppner proposed a pitting CFCG model.
Wei et.al, proposed environmentally sensitive FCG model.

Fractographic results indicated cracks originate from the pits.
Masuda, Kawai &Kasai
Elsender and Batte,
Lindley and McIntyre.

Kawai and Kasai proposed allowable stress dependent pitting CFCG model.

Kondo and Endo model CFCG from defects such as "pits"

Kondo used a pit transition to a crack at a critical depth and used Neuman and Raju solution
Kondo and Wei use this criterion

Ford proposed crack tip total strain rate in film rupture of CF model.

Additional work needed in modeling localized corrosion into CFCG.

Fig. 12. Summary of pitting corrosion fatigue models.

mechanics. However, conventional pit shapes (hemispherical) were assumed due mainly to the complexities in developing analytical approaches.

CONCLUDING REMARKS AND RECOMMENDATIONS:

This paper has briefly reviewed corrosion, electrochemical aspects, pit nucleation and growth mechanisms, corrosion and pitting corrosion fatigue of aircraft materials over the last 200 years. Recent interest in the extensive detection of corrosion and corrosion induced fatigue cracks has increased the concern of the community related to our ability to model the pitting corrosion fatigue process to incorporate life prediction into structural integrity assurance procedures. Models proposed in 1960-70's allow these predictions to be made under simplistic conditions. However, additional developments are needed to allow greater confidence in the applicability of the models to assure residual strength of structure will not be jeopardized. The greatest needs are related to the following:

1. Continued development of pitting corrosion prevention systems with verification of their validity.
2. Consideration of pit size, shape, density and interaction on structural integrity.
3. Evaluation of need to identify structurally significant items (SSI) related to pitting corrosion fatigue.
4. Development of understanding of pit growth kinetics, link up potentials of MPS (multiple pitting sources) and conversion of pits to cracks modelable by fracture mechanics.
5. Crack growth data in the structurally dependent regime and LEFM regimes to expand the data base of aged and unaged aircraft materials.

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