A Mathematical Study on Craquelure and other Mechanical Damage in Paintings

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MOLART – Molecular aspects of Ageing of painted Art – is a 5-year co-operative project between art historians, restorers, analytical chemists and technical physicists funded by the Netherlands Organisation for Scientific Research (NWO). Technical support and advice is given by Shell-SRTCA (Amsterdam), AKZO-NOBEL (Arnhem), Instituut Collectie Nederland (ICN, Amsterdam) and the Dutch art museums. The project was launched on 1 February 1995 and will end early 2002. The object of MOLART is to contribute to the development of a scientific framework for the conservation of painted art on the molecular level. The focus of MOLART is the determination of the present chemical and physical condition of works of art produced in the period from the 15th to the 20th century. Studies of historical paint manufacturing and workshop practice must give an insight into the nature of the painter’s media and the painting technique used originally. Fundamental studies on varnishes, paints and colorants are undertaken to understand the molecular aspects of ageing since this is thought to be a main cause for the continued need to treat paintings.

This report is the second in a series of MOLART reports, that will summarise all research results obtained in the course of the project. Information on this series from MOLART can be obtained from the project coordinator Prof. Dr. J.J. Boon, FOM Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands.
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MOLART is a 5-year project between art historians, restorers, analytical chemists and technical physicists funded by the Netherlands Organisation for Scientific Research (NWO). The object of MOLART is the development of a scientific framework for the conservation of painted art on the molecular level. The focus of MOLART is the determination of the present chemical and physical condition of works of art produced in the period from the 15th to the 20th century. Studies of historical paint manufacturing and workshop practice must give insight into the nature of the painter’s media and the painting technique used originally. Fundamental studies are undertaken to understand the molecular aspects of ageing since this is thought to be a main cause for the continued need to treat paintings.

Craquelure in paintings is one of the more visible aspects of ageing and is caused by the different mechanical behaviour of the various layers such as support, ground, paint etc. Moreover, this mechanical behaviour changes due to the ageing of the painting. The aim of our project is to develop a mathematical model that gives an idea of what the effects of these changing mechanical properties are on the cracking of paintings.

Cracking or, more generally, fracture is induced by mechanical stresses in a material. Tensile stress causes deformation, provided that the material is not restrained. The ratio of elongation to the initial length on an unrestrained material is called the strain. At strains below 1% most materials behave elastically, that is, the material deforms back to its original length when the load is removed. In this case the stress and the strain are linearly related. The ratio of strain to stress is called the elastic modulus and may be viewed as a measure of the material’s stiffness.

Environmental changes are an important cause of internal stress in the different paint layers. The mechanical properties of a material, e.g. the stiffness and the strength, change with temperature and relative humidity. Furthermore, the different layers will try to shrink upon desiccation and cooling. Because these layers are restrained, this produces a stress-rise in the painting.

An example of how these stresses can result in craquelure can be found in Figures 1.1 and 1.2. Theses photo’s show a detail of a painting by J. Maris. The first one was taken between 1930 and 1940, and the second was taken in 1984. The painting was exhibited in the southern wing of the ‘Rijksmuseum’ in Amsterdam, which did not have any environmental control system in that period.

The first stage of our project consisted of a literature study to obtain a more thorough understanding of craquelure in paintings and to explore the different types of research already done in this field. During this study we found that research previously carried out on the effects of
CHAPTER 1. INTRODUCTION

Figure 1.1: 'Het Jaagpad' by J. Maris (detail), photo taken ca. 1930's (Fotodienst Rijksmuseum, Amsterdam)
Figure 1.2: ‘Het Jaagpad’ by J. Maris (detail), photo taken in 1984 (Fotodienst Rijksmuseum, Amsterdam)
Environmental changes cover a wide range of methods, which makes it difficult to compare the results.

Experimental research on canvas paintings was done by Berger and Russell, measuring biaxial stress in a painting during fluctuations in temperature and relative humidity [4]. Their research showed that a small temperature rise (3°C) caused a greater stress change than a large change in humidity (38%). This result directly contradicts the commonly held belief that canvas paintings are relatively insensitive to moderate temperature changes. The stress change can be explained by thermal expansion of the paint film. This tension pushes the canvas apart, thereby relieving a portion of the tension in the canvas. They conclude that cracking and deterioration is caused primarily by the paint film, rather than by movements of the substrate.

Shrinkage of the glue size during desiccation has been identified as one of the most important sources of relative humidity-related stress development in paintings [25]. Research done by Karpowicz showed that the visco-elastic recovery of a layer of size leads to in-plane movements that are responsible for development of a common pattern of craquelure on paintings. Cracks formed at higher humidity were always sparser but wider than cracks observed in lower humidity. In higher humidity more extension was possible, and stress could be released through extension of size within an already-formed crack [20, 21]. The importance of fully understanding the structure of the painting and, in particular, the contribution of the different layers to the phenomenon of craquelure, was shown by this research.

A totally different approach was used by Bucklow [7, 8]. He developed a small set of descriptive terms for craquelure, which can be used for the attribution of paintings. He concluded from his research that changes in humidity and temperature will affect the degree to which, but not the way in which, craquelure develops.

Mecklenburg and Tumosa used a more technical approach by trying to predict the stresses in a painting when subjected to changes in temperature and relative humidity using finite element analysis [24, 25]. Finite element analysis is a numerical computing technique for solving problems in the field of continuum mechanics. Mecklenburg and Tumosa modelled a painting using experimental data on 13 year old paint.

Mecklenburg and Tumosa’s work served as the starting point for our research. The object of our project was to determine the effect of the change in mechanical properties that resulted from ageing. Research done by other MOLART participants showed that with ageing the polymeric structure of oil paint changes due to cross-linking as well as the forming of ionic bonds, where the metal ions from the pigments act as counterions [6]. Both these covalent and ionic bonds will increase the stiffness and the strength of the paint. There are several commercial finite element programs. For our computations, the modelling was performed with the MARC system, a program developed by MARC Analysis Research Corporation. Although it turned out to be very hard to determine the exact material parameter necessary for the finite element analysis, we did some simulations modelling the fresh paint as a polyacrylic acid and the aged paint as a zinc salt of polyacrylic acid.

During the project we were asked to perform some computations on the painting ‘Cathedra’ by
Barnett Newman that was cut several times in 'Het Stedelijk Museum' in Amsterdam. At the moment, restorers at the 'Instituut Collectie Nederland' (ICN) are trying to determine a way to sew the cracks and they would like to know the stress distribution around it. To determine these stress distributions the finite element analysis was used again.

Criteria from the field of fracture mechanics can be used to decide whether failure will occur when a structure is subjected to certain stress levels. One can distinguish between ductile failure and brittle failure, depending on the type of material used. Ductile failure manifests itself in plastic deformation, whereas brittle failure manifests itself in fracture.

However, even when the stress in a structure never reaches the critical level for fracture or plastic flow to occur, there still can be some resulting damage on a microscopic level. Especially, when subjected to a cyclic load, the microscopic damage resulting from each cycle can add up and finally result in fracture. This phenomenon is called fatigue. Many of the factors related to fatigue are stochastic in nature, such as initial defects and environmental conditions. The final stage of our project consisted of studying models that can compute the expected damage and the fatigue life prediction of a painting, when the environmental changes, or the stress levels, are modelled as stochastic processes. These stochastic models were developed by -among others- Sobczyk and Spencer [30, 29].

Chapter 2 of this report gives a survey of the different layers of a painting and a description of the main mechanical behavior of these layers as can be found in the literature. Chapter 3 deals with the classifications of different types of craquelure and the features a craquelure pattern can have, which was mainly inspired by the research done by Bucklow. Chapter 4 gives an introduction to elasticity theory, the part of continuum mechanics that deals with stress-strain relations for materials exhibiting elastic behaviour, and fracture mechanics. A description of the changing mechanical properties of paint during ageing is given in Chapter 5. Chapter 6 explains using some examples how the finite element method can be used to compute the stress distribution in a structure. Chapter 7 gives a description of the finite element program MARC and the results of two test runs, in which we tried to reproduce some of the results of Mecklenburg and Tumosa. Chapter 8 gives the results of the computations done with MARC, both on the models of the ageing paint and on models of the 'Cathedra'. Finally, Chapter 9 describes some of the stochastic models of fatigue phenomena and the way in which these models could be used to predict the development craquelure in paintings.

Figure 1.3 shows the different scientific disciplines that were studied during this research project, and the aspects of these disciplines that are relevant to the research of craquelure on paintings. The chapter numbers indicate where the subjects can be found in this report.
CHAPTER 1. INTRODUCTION

Figure 1.3: The different disciplines and their relations
Chapter 2

The Structure of a Painting

2.1 Introduction

A painting is a multi-layered structure consisting of a support, usually canvas or wood, a glue layer to prevent the support from absorbing the paint medium, one or more layers of ground and finally, one or more layers of paint. Craquelure in paintings is the result of the differences in mechanical behaviour of these various layers.

There are a number of mechanical properties which affect craquelure forming. The first one is the dimensional response to environmental changes, that is, changes in temperature and relative humidity. When unrestrained most materials tend to shrink upon drying and cooling, but the rate of shrinkage will be different for different materials. Therefore, the layers of a painting will act as a restraint to each other and this will cause internal stresses. The elastic modulus is a measure of a material’s ability to deform when subjected to stress, and may be viewed as a measure of the material’s stiffness. It can be used to compute the stress levels in the different layers. The strength of the materials determine whether a material will crack when subjected to certain stress levels. The exact physical definition of the elastic modulus and the strength will be given in Chapter 4.

This chapter gives a survey of the different layers of a painting, with a short introduction to the mechanical properties of each layer and its contribution to the formation of cracks.

2.2 Support

There are many types of support a painting can have, but in this section only canvas and wood will be discussed. Canvas behaves in a very different manner when subjected to environmental changes than wood, therefore, the resulting stress distributions and the crack patterns in the paint layers will be completely different.

2.2.1 Panel

There is a large variety of woods used for panels. Artists used mainly the woods from their region, that is, in Portugal oak or Spanish chestnut, in Italy poplar, in Spain pine or poplar, in France oak, walnut or poplar, and in Holland and Flanders oak [15].
CHAPTER 2. THE STRUCTURE OF A PAINTING

One aspect of wood that is important for the response to environmental changes, is that it is orthotropic, that is, it has considerably different mechanical properties in the mutually perpendicular directions, longitudinal, tangential and radial. The mechanical properties of wood vary, but in general they correlate somewhat with the density of the wood [24]. Because of the orthotropic behaviour, the crack patterns on panel painting usually form a lattice.

Only the very small panels are made of just one plank. The planks are sawn radially from the trees, i.e. perpendicular to the growth rings, because these will warp the least. In most of the panels the grain of the wood is for all planks in the panel in the same direction, that is, in the direction of the longest side of the panel. But, for example Rubens, who had a spontaneous way of painting, sometimes enlarged his panels while painting and this could result in panels with horizontal as well as vertical grains [15]. The orthotropic behaviour of the wood will then lead to internal stress.

Perpendicular to the grain the rate of shrinkage upon drying as well as cooling is significantly larger than parallel to the grain. Therefore, with changing relative humidity and temperature, stress will occur in the direction parallel to the grain and one would suspect that the predominant direction of the cracks will be perpendicular to the grain. Bucklow found in his research that a characteristic of early Italian poplar panels is indeed that the cracks run perpendicular to the grain of the wood. However, a characteristic of early Flemish oak panels is that the cracks run parallel to the grain of the wood [7].

The research of Mecklenburg and Tumosa showed that relative humidity fluctuations present a real hazard for paintings on wood panels with preexisting cracks if the panel is restrained in any way. The lack of material uniformity of wood strongly suggests that dimensional response in a wood panel will vary in different areas of the same panel, resulting in localized stress development. They concluded that all panel paintings should be maintained in a very narrow relative humidity environment [25].

2.2.2 Canvas

Around the beginning of the 16th century canvas began increasingly to be used as a support. It had actually been employed earlier, but then generally for exceptional paintings, such as objects required to be transportable and thus somewhat lighter, e.g. banners and standards to be carried in processions. The diversity of fineness of the fabric used for paintings is quite large. It is measured in the number of threads in the warp direction and the number of threads in the weft direction per square centimeter. In the 17th century there was a slight tendency to use a more coarse fabric (probably for economic reasons) [15].

The dimensional response of support fabrics to relative humidity is somewhat complicated because they respond differently to the initial relative humidity cycle than to all subsequent cycles. The fibers, consisting mainly of cellulose, are hygroscopic and they will tend to swell with increases in RH, and the elastic modulus will increase. This is the direct opposite of most other materials, which increase in stiffness upon drying.
2.3. GLUE SIZING

Moreover, the dimensional response can be divided in two regions. Below about 80% RH the swelling of the fibers causes a small increase in dimension of the woven fabric, above it, the swelling of the fibers reaches the stage where they begin to interact with each other and this causes shrinkage [17]. That is, the transverse swelling of the yarns forces the mutually perpendicular yarns to increase in crimp, shortening the textile dimensions.

Elongation of the canvas is much greater in the warp direction than in the weft direction. However, when a linen is stretched on a stretcher, the crimp is considerably reduced in the warp direction and slightly increased in the weft, so that the mechanical properties tend to even out in the two orthogonal directions. Once stretched and subjected to high relative humidity, the initial fiber tension in the linen is considerably reduced due to interfiber slippage [24].

According to Keck, crackle systems in canvas paintings show a marked similarity in pattern; those in panel paintings much less [22], but Bucklow’s research seems to refute this [8]. More about the various crack patterns on canvas paintings can be found in Chapter 3.

2.3 Glue sizing

On both panel painting and canvas paintings a sizing layer of animal glue can be found between the support and the ground. On panels the glue is applied to create a smooth and stable under­ground for the ground and paint film and it also helps the binding between the support and the ground [15]. On canvas paintings the glue is applied to stiffen the support and reduce absorbency of the oil in the ground [17].

A warm solution of glue contains a large number of randomly coiled chain-like protein molecules, identical to that of the parent protein collagen, but substantially shorter in length. When applied to the support, the solution cools down to form a gel. Drying of the gel involves gradual loss of water, and a dramatic decrease in volume [20].

The thermal coefficient of expansion for hide glue is small, while its moisture coefficient is considerably higher. Karpowicz showed that after the initial expansion, size films contract at high relative humidity, thin ones at a much faster rate than thick ones. The films contracted about twice as much as they previously expanded. The degree of contraction at high relative humidity is higher for films subjected to greater strains during previous drying, and ca. 70% is the lowest RH at which the film is still capable of returning to the length it had at normal relative humidity (ca. 50% RH) [20].

In the same research Karpowicz also examined the response to cyclic changes in RH. Five-hour exposures at high RH of 83% and then five hours at 91% were too short to contract the film below its original length. Repeated cycling, however, gradually contracts the film. Further cycling will eventually stabilize the film at its new, shorter length. This behaviour is strikingly similar to that of sized canvas [20].

The glue strengths are quite high and its failure will be extremely rare at room temperature and at relative humidity level above 75%. This material has a strength of over twenty times the weaker
lead paint at room temperature and 50% RH. If it does fail, it is almost certain that the paint layer will also be damaged [24].

In the case of supports of woven fabric, the glue has a distinct influence on the mechanical behaviour of the support. Once a painting canvas is stretched and sized with glue, the resulting tautness upon drying is from the dried glue, not the canvas. At this stage the glue, not the canvas, is the primary support of the paint layers in the painting [25]. However, at high humidity the high stiffness of the fabric dominates the behaviour.

### 2.4 Ground

Once the size is dry, the actual ground layer can be applied. According to Bucklow both the composition and the thickness of the ground layer are very influential in the development of the craquelure pattern. A thin or brittle ground layer leads to cracks which follow the grain of the wood or weave of the canvas. On the other hand, if the ground is thicker or less brittle, it will reduce the local influence of the support on the paint layer and lead to cracks, the form of which is largely determined by the mechanical properties of the ground layer [7].

#### 2.4.1 Panel

On panel paintings the ground layer mainly consists of a mixture of animal glue (possibly diluted with water) and either burnt plaster (Southern Europe) or Chalk (Northern Europe). On early Italian panel paintings there are two types in several layers. First the gesso grosso, and then the finer gesso sottile [32, 15]. A characteristic of these gypsum grounds on panel is that individual cracks are jagged, and of chalk grounds on early Flemish panel paintings is that the cracks are relatively smooth [7]. After the introduction of oil paint, painters often applied an extra layer between the ground and the paint film to reduce absorbency of the oil by the ground. This layer is transparent and besides its use as isolation it could have an optical function as well [15].

Gesso’s response to relative humidity is similar to that of rabbit skin glue. The difference is that the total length change with comparable ranges of relative humidity is considerably less, and that 80% RH, not 70% RH, seems to mark the point of demarcation between the different swelling rates. Gesso swelling is influenced by the chalk-to-glue ratio. The higher this ratio the smaller the total dimensional response to relative humidity [25]. The addition of the whiting increases the modulus, but decreases the strength considerably. In comparison to most paints, the gesso is generally stiffer, but not quite as strong (i.e. it is more brittle) [24].

#### 2.4.2 Canvas

On canvas paintings oil is mostly used as the binding medium of the ground layer. The oil is mixed with chalk and often there are various kinds of pigments added as well. A considerable
2.5. PAINT FILM

number of paintings grounded with oil paints use white lead paint. This means that in many cases there is a fairly stiff paint layer between the glue size and the upper paint layers. If this paint substrate fails, then any layer above it will most likely also fail [24].

Jagged cracks with a rectangular pattern are associated with characteristically thin brittle grounds which allow cracks to faithfully follow the (plain) canvas weave. Smooth, curved cracks are associated with thick (possibly double) grounds, which allow the complete liberation of the crack pattern from the underlying structure of the canvas [7].

2.5 Paint Film

Paint consists of at least two main components, pigment and binding medium. The pigment is the material which provides the colour, the binding medium a substance which ensures that the coloured material remains in the place where it was originally applied [32]. Bucklow concludes from his research that the medium is probably one of the less influential factors in the development of the craquelure pattern [7]. On the other hand, the pigments seem to have a distinct effect on the mechanical characteristics of paint and on the way and degree in which cracking occurs [17].

The paint is the weakest material in comparison to the glue and fabric, though depending on the mixture of chalk and glue, can be stronger than the gesso. The research done by Berger and Russell led to the conclusion that cracking and deterioration is caused primarily by the paint film, which deforms the canvas and generates stress, rather than by movements of the substrate [4].

2.5.1 Binding media

At the end of the 15th century artists started using drying oil as a binding medium for paint. Before that the most important binding medium was egg tempera. Egg tempera paints dry rather fast, therefore, they have to be applied in very thin layers. The evaporation of the water during the drying of the paint leads to considerable mass loss. If the paint were to be applied in one thick layer, this mass loss would lead to shrinking and eventually the paint film would break loose from the ground [15]. In egg tempera paint the particles are kept together by proteins as the polymeric phase and by lipids, which have emulgating properties. Because this tempera paint system already starts with polymeric phases, it is fundamentally different from an oil paint [6].

Oil paint is created with polyunsaturated triglycerides as relatively small molecular units, which have to grow into larger networks by oxidative polymerisation processes. The resulting network is capable of entangling and binding pigment particles [6]. That is, the polymers will form cross-links and later also ionic bonds, in which the metal ions will act as counterions. Both cross-linking as well as the forming of ionic bonds will increase the stiffness and make the films more brittle.

In the response to changing relative humidity there are two basic regions of swelling for most oil
2.5.2 Pigments

Different pigments are often mentioned as having an effect on the mechanical characteristics of paint and on the way and degree in which cracking occurs. Many paintings have crack patterns radically different in areas containing different pigments [17].

Pigments and fillers stiffen the material. In a glassy binder the particles act as inert links between bits of polymer. Stiffness increases up to fivefold with pigmentation levels typical of artists’ paints. In the transition and rubbery regime pigment effects are much greater. Each particle absorbs a layer of polymer about 3 nm thick which then acts more glassy, only the remaining binder acts rubbery.

Although pigments radically alter the stiffness of the paint, they do not change the way in which the stiffness depends upon the relative humidity. All oil paints double their stiffness between 70% RH and 0% RH. Above 70% RH the curves diverge depending on water sensitivity of the pigment. Pigments also change the water absorption behaviour of the oil medium. Above 70% RH, the moisture clusters in the oil medium and builds up on the pigment surface, so swelling is much greater [27].

The testing results of Mecklenburg and Tumosa serve to demonstrate the considerable differences in mechanical behaviour between the slow and fast driers. The fast driers, Naples yellow or lead white paints, showed relatively less response to relative humidity when compared to the slow drier, burnt umber or other earth colors [25]. On the other hand the slow driers, burnt umber and burnt sienna, are so flexible that their ability to withstand deformation far exceeds that of the fast driers that contain lead carbonate or other driers [24]. This difference is probably caused by the fact that slow dryers contain a much higher percentage medium than fast dryers. Furthermore, pigments containing lead will produce more ionic bonds when the paint ages, which increases the stiffness. More about the effects of ionic bonds on the mechanical behaviour of the paint can be found in Chapter 5.

2.6 Conclusion

Every layer of a painting has its own distinctive mechanical behaviour, and therefore, every layer contributes in its own way to the formation of craquelure. An obvious example of this fact is the orthogonal crack networks that can be found on panel paintings and are due to the orthotropic behaviour of the wooden support. On the other hand, the combination of certain materials can also result in certain crack features. An example of this is the fact that the thickness of the ground layer determines the influence of the support on the craquelure.

Environmental changes are a major cause of the stress development in a painting that can result in
craquelure or other mechanical damage. For instance, the glue sizing is very sensitive to changes in relative humidity. All layers will try to shrink upon drying and cooling, but each layer at a different rate. The fact that the layers are restraint by each other causes stress.
Chapter 3

Classification of Crack Patterns

3.1 Introduction

The previous chapter showed that different crack patterns on paintings indicate the use of different materials. Therefore, it will be necessary to have a simple but complete classification of craquelure. Such a classification could categorize the global features of a crack pattern, such as the density or the randomness of the pattern, as well as the local features of an individual crack, such as the depth or the smoothness of a crack. This chapter gives a survey of the possible ways to classify craquelure on paintings.

3.2 Types of Craquelure

We can distinguish three main types of craquelure, drying or premature cracks, age cracks and mechanical cracks. Drying or premature cracks are the result of mechanical stresses engendered by chemical and physical action. They are primarily due to internal stresses induced by the drying process, i.e. evaporation of solvent, either water or organic liquid, or departure of gaseous chemical alteration products. Since they are the result of internal stresses, drying cracks usually confine themselves to the layer or layers so stressed and do not, like age cracks, penetrate the entire structure from the support to the surface.

The width of a drying crack varies even for an individual rupture and is invariably greater than the width of an age crack, which tends to be narrow and uniform throughout. The edges of drying cracks are sometimes rounded in profile due to plastic elongation and flow before and after rupture [22].

The use of improper materials or combinations of materials can have a detrimental effect, increasing the internal stress. For example, too much fatty oil in the ground layer will make it too smooth, so that the paint film will not adhere well enough, and might flake when subjected to stress. Another problem occurs when the lower layers contain more oil and therefore dry more slowly than the upper layer [15].

Age cracks develop later in the life of the paint film and are attributable almost entirely to the mechanical stresses originating when the painting is subjected to environmental changes. The resulting fractures in the paint are called age cracks because young films normally possess sufficient ductility to withstand the stress involved, and it is only as a result of embrittlement from
CHAPTER 3. CLASSIFICATION OF CRACK PATTERNS

Age cracks appear in almost all paintings when they age. If a painting was conserved under ideal environmental circumstances, there would be a very fine, hardly visible crack pattern. Age cracks have sharp edges, and most of the time they are equally distributed over the whole surface [15].

Mechanical cracks are caused by temporary external stress. Similar to age cracks, mechanical cracks are fine and penetrate both the paint and the ground layer [15]. There are a number of forms of cracks due to mechanical loads. Cobweb patterns are caused by a force applied at one point in the surface of the canvas. Herring-bone patterns are caused by the wedges in the corner of the frames, or when the canvas is scratched at the back of the painting. Diagonal cracks in the corners result when a painting is over stretched or during keying out. Parallel cracking occurs when a canvas painting has been rolled up too tightly, causing cracks in parallel lines.

Drying cracks are the result of ductile fracture, while age cracks are the result of brittle fracture. Exact definitions of these phenomena will be given in Chapter 4.

In 1951 Boers tried to give a standardization for types of cracks and other mechanical damage in paint coats [5]. He did this by making 13 drawings of types of cracking and flaking and 6 drawings of cross-sections to indicate the depth of the cracks (Figures 3.2, 3.1). Although his classification was meant for industrial paint tests, it could also be helpful for classifying cracks on paintings, especially the cross-section drawings, even though most paintings have more layers than shown in these drawings.
3.2. TYPES OF CRAQUELURE

Figure 3.2: Types of cracking and flaking as given by Boers [5]
CHAPTER 3. CLASSIFICATION OF CRACK PATTERNS

Figure 3.3: Types of perpendicular disruption as given by Stout [31]

Drawing e in Figure 3.1 is a typical example of a brittle (age) crack, whereas e is a typical example of a ductile (drying) crack. Ductile cracks may cause a concentration of stress which later encourages brittle cracks to run through them. This can further complicate the classification of crack types which may be intermediate in character between typical ductile and typical brittle [17]. Drawing f shows such a crack. The most logical order is that the crack started as a drying crack in the upper layer and later also the lower layer cracked in a brittle manner.

However, it is also possible that first the lower layer cracked then later the upper layer, because the ground is the most mechanically vulnerable material because of overpigmentation. As the layer with the lowest elongation at break, grounds are the first to crack (drawing d) and cracking may not penetrate the paint layer until years later [27]. When this happens, however, it will not always result in a crack as in drawing f, because the upper layer might have already become brittle.

A classification of crack features for cracks on paintings was made by Stout [31]. He too used a set of drawings (Figures 3.3, 3.4), showing the difference between ductile and brittle cracks in the drawings of rifts and crevices. By combining different features, as shown in Figure 3.5 all types of craquelure can be described. The concave flake profile is an example of flaking, where the upper layers lose contact with the support. Flaking is more likely to happen after brittle fracture than after ductile fracture.
Figure 3.4: Fissure patterns [31]
CHAPTER 3. CLASSIFICATION OF CRACK PATTERNS

Figure 3.5: An example of double nibs [31]

3.3 Pattern features

Besides the features of an individual crack, we could also classify the features of the craquelure patterns. In Chapter 2 we already noted the difference between the patterns on panel paintings and those on canvas paintings. The most striking feature of craquelure on panel paintings is the rectilinear pattern, oriented along the wood grain. On canvas paintings the only universal characteristic is some relation to the stretcher bars behind. The most common pattern of cracks on canvas paintings can be found in Figure 3.6.

This pattern was also found in the simulation done by Mecklenburg and Tumosa, assuming that fracture will occur perpendicular to the direction of the maximum principal stress. Other common patterns that are caused by the frame can be found in Figure 3.7.

In 1995 Buklow gave characteristics of crack patterns of paintings for four different paint traditions: Italian fourteenth/fifteenth-century paintings on panel, Flemish fifteenth/sixteenth-century paintings on panel, Dutch seventeenth-century paintings on canvas and French eighteenth-century paintings on canvas [8]. He obtained these characteristics by showing pictures of crack patterns to 43 people asking them to rate these patterns for certain features.

The seven features Buklow used were not meant to give a complete classification of crack patterns, but they proved to be more than sufficient for a high level of discrimination between the categories of painting traditions [8]:

- Predominant direction and orientation of cracks
- Changes in direction of cracks. There are two possible scales for this feature, locally we
3.3. PATTERN FEATURES

Figure 3.6: Simplified diagram of crackle system, which develops in many oil-paint films on canvas [22]

Figure 3.7: Typical large-scale crack patterns in paintings on canvas [27]
can distinguish between smooth or jagged cracks, and globally we can distinguish between straight or curved cracks.

- Junctions or terminations of cracks
- Relationship between crack directions
- Distance between cracks
- Thickness of cracks
- Organization of cracks

In the survey ‘Cracking and Crack Networks in Paintings’ Hodge gave possible causes for the different features of certain crack patterns [17]. Wavy or irregular cracks have been attributed to inhomogeneity in the material causing the crack to meander, following the path of least resistance. In paintings large materials such as chalk in grounds might have this effect. Irregular cracks can also be found commonly in drying crack patterns. If cracks are wavy, the intersection will tend to occur at the convex part of the meander, where there is a stress concentration. Markedly curved cracks in paintings usually occur in those on canvas. Cracks in an isotropic stress field may curve rather than grow in a straight line. Isotropic stress will tend to produce regular spacing of cracks rather than random spacing.

The majority of junctions of cracks are ‘T’ rather than ‘X’-shaped. Most of the cracks also meet at approximately 90°. The stress release perpendicular to an existing crack, combined with its inability to carry stress over it, means that stress at right angles to the crack is eliminated. This means that any cracks formed at points of weakness along its edge will initially grow at 90°. Cracks approaching an old crack may be bent around to 90° (how near to it may depend on the amount of stress released by the opening of the first crack). In an isotropic stress field, cracks will tend to nucleate at 120°. The tendency of a network towards the orthogonal or the non-orthogonal seems to depend on the order of nucleation and the speed. There will be a difference in the patterns resulting from simultaneous nucleation and those nucleated sequentially. These include purely topological differences and differences in the predominant junction angle. If cracks grow simultaneously from several 120° angle points of nucleation, a network with a relatively high proportion of 120° angles will be formed. If cracks grow one after another, a more orthogonal network will be formed. This is especially the case if growth is rapid, so that almost as soon as a crack has nucleated, it has propagated across the surface. Later crack formation will then be greatly affected by the presence of an existing crack. Amongst paintings it seems that 18th and 19th century paintings on canvas are most likely to have orthogonal patterns. Bucklows research seems to contradict this last remark. He concludes that 17th century Dutch paintings have an orthogonal network, whereas 18th century French paintings do not [8].

In sequentially nucleated networks, there may be obvious generations of cracks. Secondary cracks may form a different pattern since they are produced by stress conditioned by the presence of the primary cracks. They may be formed as the film becomes more brittle with age and the stress released by the first set of cracks is not sufficient to prevent further cracking. As noted by Hodge, a T-junction in cracks must have formed the top of the T first. Thus, for a rectilinear pattern with two generations of cracks in two rectangular directions, the primary cracks will have been formed first and then later the strips of paint broke from side to side.
3.4 Conclusion

The enormous amount of different crack features that can be found on a painting, makes it very hard to give a good standardization of these features. This problem becomes clear when studying the work of Stout [31]. The descriptive framework he gave for classification does probably cover all possible crack features, but the set of terms becomes so big, that it will be hard to work with. Bucklows research shows that to obtain a discriminating set of features, a much smaller set would be sufficient.

But when dealing with mathematical damage modelling it would be nice to have a mathematical way to classify craquelure patterns. A possibility would be to describe the pattern by means of Bezier curves, a method that is used in handwriting recognition. At Cambridge this method was implemented for crack patterns, but it took a computer 3 days to give a good representation of specimen which was less than 5 by 5 cm. Therefore, this does not seem a very practical method. Better results might be expected by calculating the fractal dimension of the patterns, by determining the ratio between certain crack features, for instance, the ratio between ‘T’ and ‘X’-shaped crack junctions, or by determining the distribution of certain crack features throughout the surface. The latter two of which are used in the description of crack patterns in clay soils [9, 13].
CHAPTER 3. CLASSIFICATION OF CRACK PATTERNS
Chapter 4

Fracture Mechanics

4.1 Introduction

Craquelure is caused by mechanical stresses, which in turn are the result of the difference in mechanical behaviour of the various layers of a painting. In Chapter 2, a description was given of the different layers of a painting. This chapter gives an introduction to elasticity theory, which can be used to compute the stress levels that occur in the different layers of a painting when subjected to environmental changes. Furthermore, this chapter gives some criteria from the field of fracture mechanics that are used to decide whether a material will crack.

The phenomenon of the fracture of solids depends on a wide variety of factors, including macroscopic effects, the microscopic phenomena which occur at the locations where the fracture nucleates or grows, and the composition of the material. The study of the fracture process depends on the scale level at which it is considered. At one extreme there is a rupture of cohesive bonds in the solid, and the associated phenomena take place within distances in the order of $10^{-7}$ cm. For such studies the principles of quantum mechanics could be used. At the other extreme the material is considered as a homogeneous continuum, and the phenomenon of fracture is studied within the framework of continuum mechanics and classical thermodynamics [14].

The theoretical strength of a perfect crystalline material can be calculated approximately by considering the atomic structure. The strength is proportional to the interatomic force, whereas the force is a function of the interatomic distance. However, it turns out that many structural failures occur at stress levels considerably lower than the theoretical strength of the material [26, 34].

The discrepancy between predicted and actual strength can be explained in terms of stress-raising flaws which give rise to local stresses greatly in excess of those nominally borne by the body under test. These flaws may be physical discontinuities such as cracks or surface scratches, discontinuities of elastic moduli (as when a particle of one other material is embedded in a matrix of a second) or irregularities in the ordering of atoms such as dislocations in a crystal lattice.

4.2 Elasticity Theory

Stress, denoted by $\sigma$, is defined as force, $F$, divided by the cross-section area, $A$, upon which the force acts (see Figure 4.1), that is,

$$\sigma = \frac{F}{A}.$$
and hence, the unit used to measure stress is Pascal (1 Pa = 1 N/m²).

Under tensile stress, that is, when the force acts on an area perpendicular to it, the intermolecular bonds of a material will resist elongation to a greater or a lesser degree. Let \( L_0 \) denote the original length of a specimen, and let \( L \) denote the length after the force is applied, then the ratio of elongation \( L - L_0 \) to the initial length is called strain, and will be denoted by \( \varepsilon \), i.e.,

\[
\varepsilon = \frac{L - L_0}{L_0}.
\]

Tensile stress also causes a specimen to shrink in the directions perpendicular to the applied force. The transverse strain \( \varepsilon_T \), i.e., the ratio of the elongation to the original length in the directions perpendicular to the applied force, will normally be a negative quantity. The ratio between the longitudinal strain and minus the transverse strain is called the Poisson ratio, and is denoted by \( \nu \):

\[
\nu = -\frac{\varepsilon_T}{\varepsilon}.
\]

Then the change in volume is given by

\[
\frac{\Delta V}{V_0} = (1 - 2\nu)\varepsilon + \text{higher order terms}.
\]

For most materials the Poisson ratio is in the range 0.2 and 0.5 and in elasticity theory the strain is usually less than 1%, therefore, we can neglect the higher order terms. And hence, when the Poisson ratio is 0.5, the volume of the specimen will remain constant under tensile stress. This condition of constant volume holds for liquids and ideal rubbers. In general, however, there is an increase in volume [28].

In general, the stress will have tensile as well as shear components. The tensile components are denoted by \( \sigma_x \), \( \sigma_y \), and \( \sigma_z \), and the shear components are denoted by \( \sigma_{xy} \), \( \sigma_{yx} \), \( \sigma_{xz} \), \( \sigma_{zx} \), \( \sigma_{yz} \), and \( \sigma_{zy} \) (see Figure 4.2). Mathematically, this means the stress is given by a tensor.

A more formal approach is to first define a stress vector \( \sigma_n \) as the surface density of the applied force [11]. That is, \( \sigma_n \) is the limit of the ratio of the surface force \( F \) acting on a surface element with normal \( \mathbf{n} \) to the area \( \mathbf{A} \) of this element when \( \mathbf{A} \to 0 \) and the element shrinks to a point, i.e.,

\[
\sigma_n = \lim_{\mathbf{A} \to 0} \frac{\mathbf{F}}{\mathbf{A}},
\]  

(4.1)
4.2. ELASTICITY THEORY

Figure 4.2: Three-dimensional stress state

where \( A \) is the surface area and \( n \) is the normal to this surface. The components of the stress tensor are then defined as the quantities \( \sigma_{ij} \) such that

\[
\sigma_n^i = \sum_j \sigma_{ij} n_j
\]

for all \( n \).

Hence, we speak of tensile stress if we can choose a coordinate system such that only one of the components on the diagonal of the matrix \( \sigma_{ij} \) is non-zero, and all the off-diagonal components are zero. When there is no internal angular momentum, the tensor is symmetric, that is \( \sigma_{ij} = \sigma_{ji} \).

Hence, for the shear components of the stress we have the symmetry rules \( \sigma_{xy} = \sigma_{yx} \), \( \sigma_{xz} = \sigma_{zx} \) and \( \sigma_{yz} = \sigma_{zy} \).

Now, we can derive the equilibrium equations for the internal forces (stresses) and the external forces:

\[
\sum_j \frac{\partial \sigma_{ij}}{\partial x_j} + F_i = 0 \quad \text{for } i \in \{1, 2, 3\},
\]

where \( F_i \) are the external forces per unit volume. In the general case it is impossible to determine six unknown quantities \( \sigma_x, \sigma_y, \sigma_z, \sigma_{xy}, \sigma_{xz} \) and \( \sigma_{yz} \) from the three equilibrium equations and
the forces at the boundaries. In order to determine the state of stress in such a case one must also invoke stress-strain relationships such as, for example, Hooke's law for elastic solids [26].

Strain is a dimensionless symmetric tensor. If we consider the three-dimensional deformation of a solid, where the displacements of a point is \( u = (u_1, u_2, u_3) \), the strain of an element of the solid is described by six quantities: three tension-compression strains along the coordinate axes and three shear strains.

\[
\varepsilon_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \quad \text{with } i, j = 1, 2, 3.
\]

The stress-strain relationships can differ significantly for different types of materials (see Figure 4.3). However, when the strain is small (less than 1%), most materials will behave elastically, that is, after removing the load, the material will deform back to its original shape. In this case, the strains are uniquely determined by the stresses (at constant temperature) [28].

Now, Hooke's law for elastic solids states a linear relation between tensile stress and strain, i.e.

\[
\sigma = E \varepsilon,
\]

where \( E \) is called the elastic modulus. Or, more generally,

\[
\varepsilon_{ij} = \sum_{k,l} A_{ij}^{kl} \sigma_{kl}.
\]

For isotropic linear elastic solids, the coefficients \( A_{ij}^{kl} \) only depend upon the elastic modulus \( E \) and the Poisson ratio \( \nu \). The stress-strain relations in three dimensions then become:

\[
\begin{align*}
\varepsilon_x &= \frac{1}{E} (\sigma_x - \nu(\sigma_y + \sigma_z)), \\
\varepsilon_y &= \frac{1}{E} (\sigma_y - \nu(\sigma_x + \sigma_z)), \\
\varepsilon_z &= \frac{1}{E} (\sigma_z - \nu(\sigma_x + \sigma_y)), \\
\varepsilon_{xy} &= \frac{1 + \nu}{E} \sigma_{xy}, \\
\varepsilon_{yz} &= \frac{1 + \nu}{E} \sigma_{yz}, \\
\varepsilon_{zx} &= \frac{1 + \nu}{E} \sigma_{zx}.
\end{align*}
\]

### 4.3 Failure Criteria

#### 4.3.1 Tensile strength and plastic flow

When looking at the stress-strain curve of a ductile material, we can see that with an increasing strain the stress will reach a local maximum, called the yield point or yielding stress, \( \sigma_Y \). From here on the material will change from elastic behaviour to plastic behaviour. Plastic deformation is irreversible, that is, when the load is removed the material will not deform back to its original shape. If we keep increasing the strain, the material will eventually break. The stress level \( \sigma_F \) at which fracture occurs, is called the ultimate strength [28, 26].
4.3. FAILURE CRITERIA

Figure 4.3: Strain-strain curves for different types of material [28]
When paint is still young, it is a ductile material, and therefore premature cracks are the result of a plastic deformation followed by fracture. On the other hand, age cracks are the result of brittle fracture. Brittle materials break before the yield point is reached. Hence, the only failure criterion is the ultimate strength.

Because the stress tensor is symmetric, we can choose a coordinate system such that $\sigma_{ij} = 0$ if $i \neq j$. The coordinate axes of this system are called the principal axes and diagonal components of the stress tensor, i.e. $\sigma_{ii}$, are called the principal values. On the surface elements perpendicular to the principal axes, the stress vectors as defined by (4.1) are directed along the normals to the elements. When put in order of magnitude, the first principal value is also the maximum normal stress, and the last the minimum normal stress. Brittle fracture will occur when the maximum principle stress exceeds the ultimate strength.

To decide whether plastic deformation will occur however, we do not compare the yield stress $\sigma_Y$ with the maximum principle stress, but other failure criteria can be used. The best known yield criteria are the Tresca equivalent stress [11]

$$\frac{1}{2} \max\{|\sigma_1 - \sigma_2|, |\sigma_2 - \sigma_3|, |\sigma_3 - \sigma_1|\},$$

and the Von Mises equivalent stress

$$\frac{1}{2} \left\{ (\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2 \right\}^{1/2},$$

where $\sigma_i$, $i = 1 \ldots 3$, are the principal stresses. Hence, if the Tresca equivalent stress or the Von Mises equivalent stress exceeds the yield stress, plastic deformation will probably occur.
4.3. FAILURE CRITERIA

4.3.2 Energy balance in fractured bodies

When a solid is fractured new surfaces are created in the medium in a thermodynamically irreversible manner. Material separation is caused by the rupture of atomic bonds due to high local stresses. The energy balance in a fractured body is given by [14, 34].

\[ W = U + \Gamma + K, \]

where

\[ W \] is work done,
\[ \Gamma \] is the dissipated energy used to increase the crack area
\[ U \] is internal energy,
\[ K \] is the kinetic energy.

Let \( A \) denote the fracture area, then the energy balance during crack growth can be given as a function of \( A \):

\[
\frac{dW}{dA} = \frac{dU}{dA} + \frac{d\Gamma}{dA} + \frac{dK}{dA}.
\]  \hspace{1cm} (4.5)

The internal energy \( U \) can be divided into the elastic strain energy \( U^e \) and the plastic work \( U^p \). For an ideally brittle material, the energy dissipated in plastic deformation is negligible. Fracture resistance \( R \) is defined as the energy dissipated in propagating a fracture over an increment \( \delta A \) of the fracture area. In the limit this gives

\[ R = \frac{d\Gamma}{dA}. \]

And the energy release rate \( G \) is defined as the energy available for crack growth, i.e.

\[ G = \frac{dW}{dA} - \frac{dU^e}{dA} = \frac{dK}{dA} + R. \]

If \( \gamma \) represents the energy required to form a new unit of material surface, then the Griffith criterion for crack growth is given by [34]

\[ G = 2\gamma, \]  \hspace{1cm} (4.6)

where the factor 2 appears because two new material surfaces are formed during crack growth. Therefore, the right-hand side of Equation 4.6 represents the resistance of the material that must be overcome by crack growth.

A crack is called stable, when it is at rest or grows with constant or decelerating speed. When the growth rate is accelerating, the crack is said to be unstable. This is always the case with crack initiation. For fracture initiation, we have that \( K = 0 \), and \( \frac{dK}{dA} \geq 0 \), hence, \( G - R \geq 0 \) [34].
4.3.3 Stress intensity factor

Instead of considering the global energy balance, it is also possible to attain a fracture criterion based on a local analysis of the stresses around a crack tip. Consider a plane crack extending through the thickness of a flat plate. There are three independent kinematic movements of the upper and the lower crack surfaces with respect to each other, as shown in Figure 4.5.

Now, consider a crack of length $2a$ in an infinite plate subjected to uniform equal stresses $\sigma$ along the $x$ and $y$ directions at infinity. To describe the stresses around the crack tip, we will use polar coordinates $r$ and $\theta$, with the middle of the crack as the origin (see Figure 4.6) [14]:

$$\sigma_x = \frac{K_I}{\sqrt{2\pi r}} \cos \left( \frac{\theta}{2} \right) \left( 1 - \sin \frac{\theta}{2} \sin \frac{3\theta}{2} \right),$$  

(4.7)

$$\sigma_y = \frac{K_I}{\sqrt{2\pi r}} \cos \left( \frac{\theta}{2} \right) \left( 1 + \sin \frac{\theta}{2} \sin \frac{3\theta}{2} \right),$$  

(4.8)

$$\sigma_{xy} = \frac{K_I}{\sqrt{2\pi r}} \cos \left( \frac{\theta}{2} \right) \sin \frac{\theta}{2} \cos \frac{3\theta}{2},$$  

(4.9)

where

$$K_I = B \sigma \sqrt{\pi a}$$  

(4.10)

is the stress intensity factor. $B$ denotes a factor that accounts for the geometry of the specimen and the crack. In the example given above of an infinite plate with uniform stress at infinity $B = 1$. The subscript $I$ indicates the fact that in this case we have an opening fracture mode. Similar equations can be derived for the other fracture modes. For analyzing the shearing mode fracture, the plate should be subjected to uniform in-plane shear stress at infinity, and for analyzing tearing mode fracture, the plate should be subjected to uniform out-of-plane shear stress at infinity.

The stress intensity factor is a fundamental quantity that governs the stress field near the crack tip. It depends both on the geometrical configuration and the loading conditions of the body. However, it can be shown that the Equations 4.7–4.9 apply to all crack tip stress fields independently of crack/body geometry and the loading conditions [14].
Figure 4.6: A crack of length $2\alpha$ in an infinite plate subjected to a uniform stress $\sigma$ at infinity
Consider the case of an opening mode crack that extends along its own direction in a self-similar manner, i.e., the shape of the crack near the crack tip does not alter. Due to symmetry, only normal stresses will be present in elements along the crack direction (see Figure 4.7). Assume the crack extends by a length \( \delta \), then the energy released during crack extension is the work performed by the stresses \( \sigma_y(\delta - \beta^*, 0) \) acting through the displacements \( u_y(\beta, \pi) \). For \( \delta \to 0 \) the conditions \( u_y \to u^*_y \) and \( \beta \to \beta^* \) are satisfied, and the work performed is given by the energy release rate \([14]\)

\[
G_1 = 2 \lim_{\delta \to 0} \frac{1}{\delta} \int_0^\delta \frac{1}{2} \sigma_y(\delta - \beta, 0) u_y(\beta, \pi) \, d\beta,
\]

where the subscript I indicates the fracture mode and the displacement \( u_y \) is given by

\[
u_y = \frac{K_I}{E} \sqrt{\frac{\pi}{2\delta}} \sin \frac{\theta}{2} \left( (3 - \nu) - (1 + \nu) \cos \theta \right).
\]

Then, with Equation 4.8 we get

\[
G_1 = \frac{K_I^2}{E} \frac{2}{\pi} \lim_{\delta \to 0} \frac{1}{\delta} \int_0^\delta \sqrt{\frac{\beta}{\delta - \beta}} \, d\beta.
\]

The limit of the integral can be written as

\[
\lim_{\delta \to 0} \int_0^\delta \sqrt{\frac{\beta/\delta}{1 - \beta/\delta}} \, d\beta = \int_0^1 \sqrt{\frac{\pi}{1 - x}} \, dx = \frac{\pi}{2}
\]

and therefore we have

\[
G_1 = \frac{K_I^2}{E}.
\]
Hence, we can write the Griffith criterion (Equation 4.6) in terms of the stress intensity factor and it follows that in the elastic case the global energy analysis, using the energy release rate $G$, is equivalent to the analysis of the local stresses around the crack tip, using the stress intensity factor $K$.

Although the theories on the fracture criteria such as the stress energy release rate and the stress intensity factor will not be used in the rest of this research, they do help to understand the physical phenomena that take place in a painting.

### 4.4 Conclusion

When we assume the materials in a painting to behave linear elastically, we can use the theories as described in the first section of this chapter to compute the stress in a painting that is subjected to environmental changes. For instance, when the temperature rises, the paint layer will try to expand. The expansion rate is called the thermal expansion coefficient. In general, the other layers, such as the support and the glue sizing, will have a different thermal expansion coefficient, and therefore, the paint will endure a certain strain. When the stress-strain relation of the paint is known, we can compute the stress in the paint layer. And then we can use the fracture criteria, such as the ultimate strength to determine whether the paint layer will crack.
Chapter 5

Mechanical Properties of Polymers

5.1 Introduction

Most paint media (and glue) are amorphous or semi-amorphous polymers and have some distinctive mechanical properties. A linear polymer consists of long molecular chains of covalently bonded atoms, each chain being a repetition of much smaller chemical units. These chains can be joined by other chains at points along their length to make a cross-linked structure.

When cooled down from the melt many polymers form a disordered structure called the amorphous state. Some of these materials, have a comparatively high modulus at room temperature, but others, have a low modulus. These two types of polymers are often termed as glassy and rubber-like respectively. The form of behaviour exhibited depends on the temperature relative to a glass-rubber transition temperature \( T_g \) that is dependent on the material and the test method employed.

If the polymer is warm enough and tension is applied slowly enough, the molecules bump past each other enough to let their kinks straighten out. The network can be pulled to twice its length without breaking, hence rubbery behaviour. Glassy behaviour occurs when the molecules do not vibrate enough to bump past each other during the application of stress. Note that time and temperature are intertwined: glassy behaviour can arise from either too low temperatures or too little time.

Part of the chemical research done within the MOLART project concerns itself with the chemical ageing of oil paint [6]. The model recently proposed states that the triglycerides in an oil based paint auto-oxidise to reactive monomers, which in turn form cross-links to a glycerolester based polymer. The glycerol ester bonds in this polymer will then hydrolyse and the oil paint becomes a polyanionic polymer. Metal ions of the pigments will act as counter ions to balance the charge.

This chapter describes the effect of cross-linking as well as ionic bonds on the mechanical properties of polymers, and gives a short introduction to the theory of time-dependent mechanical behaviour of polymers.

5.2 Cross-linking

The low-strain elastic moduli of polymers are functions of the rate of tests well as of temperature. The modulus-temperature curve for an amorphous polymer is almost horizontal in the glassy
Temperature region, then will drop steeply in the glass-to-rubber transition region (Figure 5.1). The response then levels out to a nearly constant plateau in the rubbery region, the length of which increases with the molecular weight and finally drops to zero. If the polymer is cross-linked, the response levels off at the true rubbery modulus. In this case the height of the final plateau increases with increasing degree of cross-linking (Figure 5.2). The degree of cross-linking is measured by the number-average molecular weight between cross-linked points, $M_c$.

In addition to raising the rubbery modulus, cross-linking produces two other effects. First, when the cross-link density becomes fairly high, the glass transition temperature is increased, so the drop in the modulus becomes shifted to higher temperatures or longer times. Second, the transition region is broadened, with the modulus dropping at a lower rate and plateauing at a higher level. Cross-linking has rather little effect on the magnitude of the modulus in the glassy state [28].

The brittle strength of a polymer is greatly influenced by material variables such as molecular weight, crystallinity, additives and cross-linking. The strength increases rapidly with increased molecular weight with a tendency to level off as this becomes very large or very small [2].

Figure 5.1: Modulus-temperature curves for different molecular weights [28].
5.2. CROSS-LINKING

Figure 5.2: Effect of cross-linking on modulus-temperature curves. The numbers on the curves are approximate values of $M_c$. The value 30000 refers to an uncross-linked polymer with molecular weight of roughly 30000 between entanglement points [28].
5.3 Ionic Polymers

An ionic polymer is a polymer—either inorganic or organic—which contains both covalent and ionic bonds in its chain or network structure. A typical example is a metal salt of polyacrylic acid. By convention, ionic polymers exist in the salt or ionised form; that is the counterion—usually a cation—is not hydrogen. The ionic bond is a strong bond, like the covalent bond; however, it can be thermally labile under circumstances, which may be a useful property.

The chains of a conventional polymer are made up of covalent, i.e., strong directional bonds formed by joining together atoms with a valency of two or higher. An important variable in polymer science is the way in which a network is arranged or packed in space. In contrast to the polymer structure, there exists ionic solids, made up of ionic, i.e. strong non-directional bonds, between atoms, with a valency of one or higher. Since the ionic bond is non-directional, there is no network in the polymer sense, and the ions tend to cluster or pack as closely as possible.

The segmental mobility of the molecules will depend upon the inter-chain forces, and in the case of ionic polymers upon the degree of ionization and the presence of ionic bonds. Experimental work confirms this and shows that the electrostatic forces between the bound ions in the polymer chain, and the counterions reduce the segmental mobility and, therefore, increase $T_g$ [18].

The effect of cross-linking is to increase the transition temperature, as is in general the case with ionic bonds. In Figure 5.3 an example is given of the effect of ionic bonds on the stiffness. Strength is a more complex property than stiffness, since it is greatly affected by the specimen imperfections, and involves larger strains. Although it is difficult to make broad generalizations about strength, it is usually true to say that increase in stiffness also increases the strength. However the increase in strength, in the case of ionic bonds, is less than the increase in stiffness.

The thermal expansion coefficient is related to the elastic modulus of a material. Barker [3] has shown that the following very approximate relationship is valid for a large number of polymer materials:

$$E\alpha^2 \approx 15\text{Pa }{^\circ}\text{C}^{-2}.$$  \hspace{1cm} (5.1)

However, the thermal expansion coefficient for zinc salt of polyacrylic acid is much less than would be expected based on this relationship (see Table 5.1).
5.4 Creep and stress-relaxation

The time-dependence of the elastic modulus of polymers gives rise to the phenomena of creep and stress relaxation. Creep is the increase of strain with time observed in a specimen under constant stress and, conversely, stress-relaxation is the decrease of stress with time at constant strain.

The behaviour of materials of low relative molecular weight is usually discussed in terms of two particular types of ideal material: the elastic solid and the viscous liquid. The former has a definite shape and is deformed by external forces into a new equilibrium shape; on removal of these forces it reverts instantaneously to its original form. The solid stores all the energy which it obtains from the external forces during the deformation, and this energy is available to restore the original shape when the forces are removed. By contrast, a viscous liquid has no definite shape and flow irreversible under the action of external forces. One of the most interesting features of polymers is that a given polymer can display a gliding scale of properties between an elastic solid and a viscous liquid depending on the temperature and the experimentally chosen time scale. Mathematically, we can describe elastic behaviour with Hooke's law and viscous behaviour with Newton's law:

1. Hooke's law: \((\sigma)E = E\epsilon\), where \(\sigma\) is the stress, \(\epsilon\) is the strain and \(E\) is the elastic modulus.

2. Newton's law: \((\sigma)_V = \eta \frac{\partial \epsilon}{\partial t}\), where \(\eta\) is the viscosity.

![Graph showing shear modulus vs. temperature for different materials](image-url)
CHAPTER 5. MECHANICAL PROPERTIES OF POLYMERS

In the first law, stress is linearly related to the strain, in the second the stress is linearly related to the strain rate [33].

A simple possible formulation of linear viscoelastic behaviour combines these two equations, making the assumption that shear stresses related to strain and strain rate are additive:

\[ \sigma = (\sigma)_E + (\sigma)_V = E\varepsilon + \eta \frac{\partial \varepsilon}{\partial t}. \] (5.2)

This formulation is used to model a stress-relaxation experiment with a Maxwell unit, which consists of a Hookean spring with elasticity modulus E and a Newtonian dashpot with viscosity \( \eta \) in series (see Figure 5.4). The model is given a definite strain while the stress is measured as a function of time, i.e.

\[ \frac{d\varepsilon}{dt} = \frac{1}{E} \frac{d\sigma}{dt} + \frac{\sigma}{\eta} = 0. \]

The solution of this equation of motion is

\[ \frac{\sigma}{\sigma_0} = e^{-Et/\eta} = e^{-t/\tau}, \]

where

\[ \tau = \frac{\eta}{E} \]

is called the relaxation time.

Initially, the deformation in the Maxwell unit takes place in the spring, later, the dashpot starts to relax and allows the spring to contract. Most of the relaxation takes place within one decade of time on both sides of the relaxation time. This is shown clearly when the stress ratio is plotted on a logarithmic time scale (see Figure 5.5). The curve has a maximum slope at the time \( t = \tau \).

The stress-relaxation of the Maxwell unit may also be given in terms of a stress-relaxation modulus \( E_r(t) \):

\[ E_r(t) = \frac{\sigma(t)}{\varepsilon} = \frac{\sigma_0}{\varepsilon} e^{-t/\tau}. \]

Stress relaxation of the paint film is one of the reasons that paintings do not completely deteriorate, even when the temperature and the relative humidity change.

5.5 Conclusion

Oil paint is, in general, a mixture of oil and paint. During ageing the pigment molecules will form ionic bonds with the cross-linked network of the medium. As a result of the cross-linking and the ionic bonds, the mechanical properties of the paint will change.
5.5. CONCLUSION

Figure 5.4: Maxwell unit

Figure 5.5: Stress-relaxation of a Maxwell model on a logarithmic time scale, $\tau = 1$
First of all, the elastic modulus will increase, that is, the paint will become stiffer, and so will the strength. The thermal expansion coefficient on the other hand will decrease. However, it seems that the relation between the elastic modulus and the thermal expansion coefficient that holds for most polymers, does not hold for ionic polymers.
Chapter 6

Finite Element Analysis

6.1 Introduction

Finite Element Analysis is a numerical procedure for solving a continuum mechanics problem. It was applied by Mecklenburg and Tumosa (see [24, 25]) on a model of a painting to compute the stresses that occurred on cooling and drying. This chapter gives an introduction into the theory of Finite Element Analysis.

The idea of Finite Element Analysis (FEA) is to divide the structure into small regions, called elements, and to appoint a finite number of points, called nodes, where the elements are connected to one another (see Figure 6.1) [10, 12]. The elements and the nodes together are called the mesh.

6.2 Theory

FEA works by assuming a given distribution of the unknown variable(s) through each element. The equations defining the approximation distribution are known as interpolation functions, and can take any mathematical form, although in practice they are usually polynomials. The number of nodes in an element increases with an increase in the order of the polynomial, or with the number of unknowns in the polynomial.

Figure 6.1: Discretization of a region into finite elements
Example 6.1 For the rectangular element in Figure 6.2 with four nodes we can use the polynomial

\[ T(x, y) = \alpha_1 + \alpha_2 x + \alpha_3 y + \alpha_4 xy \]  

(6.1)
as interpolation function. This function then defines the variation of \( T \) over the element. If we know the value of \( T \) in the four nodes, i.e., \( t_i = T(x_i, y_i) \), then we can solve Equation 6.1 for \( \alpha_1, \alpha_2, \alpha_3 \) and \( \alpha_4 \):

\[
\begin{align*}
\alpha_1 &= \frac{1}{A} (x_2 y_3 t_1 - x_1 y_3 t_2 + x_1 y_1 t_3 - x_2 y_1 t_4), \\
\alpha_2 &= \frac{1}{A} (-y_3 t_1 + y_3 t_2 - y_1 t_3 + y_1 t_4), \\
\alpha_3 &= \frac{1}{A} (-x_2 t_1 + x_1 t_2 - x_1 t_3 + x_2 t_4), \\
\alpha_4 &= \frac{1}{A} (t_1 - t_2 + t_3 - t_4),
\end{align*}
\]

where we assume that the edges of the element are parallel to the coordinate axes (i.e., \( x_1 = x_4, x_2 = x_3, y_1 = y_2 \) and \( y_3 = y_4 \)) and \( A = (x_2 - x_1)(y_3 - y_1) \) is the area of the element.

Substituting these back into 6.1 we can write

\[ T = n_1 t_1 + n_2 t_2 + n_3 t_3 + n_4 t_4 = n^T \cdot t, \]

with

\[
\begin{align*}
n_1 &= \frac{1}{A} (x_2 y_3 - y_3 x - x_2 y + xy), \\
n_2 &= \frac{1}{A} (-x_1 y_3 + y_3 x + x_1 y - xy), \\
n_3 &= \frac{1}{A} (x_1 y_1 - y_1 x - x_1 y + xy), \\
n_4 &= \frac{1}{A} (-x_2 y_1 + y_1 x + x_2 y - xy).
\end{align*}
\]
6.2. THEORY

\( n(x, y) \) is called the shape function vector.

Assume that the rectangular element as given in Figure 6.2 has two degrees of freedom in every node. The variations of the displacements in the \( x \) and \( y \) direction, \( v_x \) and \( v_y \), can then be approximated in terms of the nodal displacements, \( u_{i,x} \) and \( u_{i,y} \), \( i = 1, \ldots, 4 \).

\[
\begin{pmatrix}
v_x \\
v_y
\end{pmatrix} =
\begin{pmatrix}
n_1 & 0 & n_2 & 0 & n_3 & 0 & n_4 & 0 \\
0 & n_1 & 0 & n_2 & 0 & n_3 & 0 & n_4
\end{pmatrix}
\begin{pmatrix}
u_{1,x} \\
u_{1,y} \\
u_{2,x} \\
u_{2,y} \\
u_{3,x} \\
u_{3,y} \\
u_{4,x} \\
u_{4,y}
\end{pmatrix}
= \mathbf{N} \mathbf{u}.
\]

The number of degrees of freedom is the dimension of the space of possible movements—displacements or rotations—of all nodes. For example, the element in Figure 6.2 has 8 degrees of freedom, that is, every node can move in the \( x \)- or in the \( y \)-direction. Then, for an element with \( n \) d.o.f. we can write the equation

\[
\mathbf{K}^{(e)} \mathbf{u}^{(e)} = \mathbf{F}^{(e)},
\]

where

- \( \mathbf{K}^{(e)} \) is an \((n \times n)\)-matrix, called the element stiffness matrix,
- \( \mathbf{u}^{(e)} \) is the element nodal displacement vector, and
- \( \mathbf{F}^{(e)} \) is the vector of element nodal loads.

The \( j \)th column of \( \mathbf{K}^{(e)} \) is the vector of forces (and perhaps moments) that must be applied to the element to make \( u_j = 1 \) and preserve static equilibrium while \( u_i = 0 \) for all \( i \neq j \).

For the whole structure, the equations have the same form:

\[
\mathbf{K} \mathbf{u} = \mathbf{F},
\]

where \( \mathbf{K} \) is the structure stiffness matrix. The \( j \)th column of \( \mathbf{K} \) is the vector of nodal forces that must be applied to the nodes to maintain static equilibrium when the \( j \)th d.o.f has unit displacement and all other d.o.f. have zero displacement.

When these equations are solved for \( \mathbf{u} \), the deformations of all elements are known and from the deformations we can compute the stresses.

**Example 6.2** Sometimes it is possible to derive the stiffness matrix directly. The structure in Figure 6.3 consists of three springs of stiffness \( k_1 \), \( k_2 \) and \( k_3 \). We can see each string as a one-dimensional finite element. By assigning a unit displacement to every node in turn, each time
writing the necessary forces as a column in a \( (4 \times 4) \)-matrix, we find

\[
\begin{pmatrix}
-k_1 & (0) & (0) & (0) \\
0 & k_1 & (0) & (0) \\
0 & (0) & k_2 & (0) \\
0 & 0 & -k_2 & k_3
\end{pmatrix}
\begin{pmatrix}
u_1 \\
u_2 \\
u_3 \\
u_4
\end{pmatrix}
= \begin{pmatrix}
-P \\
0 \\
0 \\
0
\end{pmatrix}.
\]

The boxed parts give the element stiffness matrices for the three elements. Because node 4 is restrained from motion, i.e. \( u_4 = 0 \), the actual number of degrees of freedom is 3 and the equation becomes

\[
\begin{pmatrix}
-k_1 & 0 & 0 \\
0 & k_1 + k_2 & -k_2 \\
0 & -k_2 & k_2 + k_3
\end{pmatrix}
\begin{pmatrix}
u_1 \\
u_2 \\
u_3
\end{pmatrix}
= \begin{pmatrix}
-P \\
0 \\
0
\end{pmatrix}.
\]

For elasticity problems, we can use the variational formulation to derive the finite element equations. This procedure consists of minimizing the potential energy \( \Pi \) of the system:

\[
\Pi = U_e - W,
\]

where \( U_e \) is the strain energy in the body and \( W \) is the work done by the external loads acting on the body.

For a differential element of volume \( dV \), the strain energy is given by

\[
dU_e = \frac{1}{2} (e - e_0)^T \cdot \sigma,
\]

where \( e \) is the total strain vector, \( e_0 \) is the initial strain vector and \( \sigma \) is the vector of stress components. Hence the total strain energy for a finite volume can be calculated by

\[
U_e = \int_V \frac{1}{2} (e - e_0)^T \cdot \sigma \, dV.
\]  
\[
(6.7)
\]

For convenience, we write the stress and the strain as vectors, instead of tensors, i.e.,

\[
\sigma = (\sigma_x, \sigma_y, \sigma_z, \sigma_{xy}, \sigma_{xz}, \sigma_{yz})^T \quad \text{and} \quad e = (e_x, e_y, e_z, e_{xy}, e_{xz}, e_{yz})^T.
\]

Then we can write Equation 4.2 as

\[
(e - e_0) = A\sigma,
\]
where we also account for the initial strain $\varepsilon_0$, or as

$$\sigma = D(\varepsilon - \varepsilon_0),$$

where the matrix $D = A^{-1}$ depends on the elastic modulus $E$ and the Poisson ratio $\nu$ of the material.

A two-dimensional elasticity problem is either a plane stress or a plane strain approximation of a real three-dimensional situation. Plane stress means that the stress normal to the plane is zero, whereas plane strain means that the strain normal to the plane is zero. In the case of a plane stress problem, the strain $\varepsilon$ has three components, the two tensile strain components $\varepsilon_x = \frac{\partial u}{\partial x}$ and $\varepsilon_y = \frac{\partial v}{\partial y}$, and the shear strain component $\varepsilon_{xy} = \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}$. Hence,

$$\varepsilon = \begin{pmatrix} \varepsilon_x \\ \varepsilon_y \\ \varepsilon_{xy} \end{pmatrix} = \begin{pmatrix} \frac{\partial}{\partial x} & 0 & 0 \\ 0 & \frac{\partial}{\partial y} & 0 \\ \frac{\partial}{\partial y} & \frac{\partial}{\partial x} & 0 \end{pmatrix} \mathbf{v} = \mathbf{B} \mathbf{u},$$

where $\mathbf{u}$ is the nodal displacement vector and

$$\mathbf{B} = \begin{pmatrix} \frac{\partial}{\partial x} & 0 & 0 \\ 0 & \frac{\partial}{\partial y} & 0 \\ \frac{\partial}{\partial y} & \frac{\partial}{\partial x} & 0 \end{pmatrix} \mathbf{N}.$$

And $D$ for a plane stress problem is given by

$$D = \frac{E}{1 + \nu^2} \begin{pmatrix} 1 & \nu & 0 \\ \nu & 1 & 0 \\ 0 & 0 & \frac{1-\nu}{2} \end{pmatrix}. \quad (6.8)$$

When we substitute these into (6.7), we get

$$U^e = \frac{1}{2} \int_V \mathbf{u}^T \mathbf{B}^T \mathbf{D} \mathbf{B} \mathbf{u} - 2\mathbf{u}^T \mathbf{B}^T \mathbf{D} \varepsilon_0 + \varepsilon_0^T \mathbf{D} \varepsilon_0 \ dV.$$

For the body to be in equilibrium, its potential energy must be a minimum, hence

$$\frac{\partial \Pi}{\partial \mathbf{u}} = \frac{\partial U^e}{\partial \mathbf{u}} - \frac{\partial W}{\partial \mathbf{u}}
= \left( \int_V \mathbf{B}^T \mathbf{D} \mathbf{B} \ dV \right) \mathbf{u} + \int_V \mathbf{B}^T \mathbf{D} \varepsilon_0 \ dV - \frac{\partial W}{\partial \mathbf{u}} = 0,$$

which can be written as

$$K\mathbf{u} = \mathbf{F},$$
which is exactly Equation 6.6.

Now, we have the equilibrium equation, which can be solved by taking the inverse of the stiffness matrix

\[ u = K^{-1}F. \]

However, determining \( K^{-1} \) requires a large amount of CPU time. Moreover, \( K \) is usually narrowly banded, that is, besides a narrow band around the diagonal, all matrix elements are zero. But \( K^{-1} \) is, in general, no longer banded. Therefore an alternative method is used, namely Cholesky decomposition of \( K \). First step is to obtain the lower triangular matrix \( L \) such that

\[ LL^T = K, \quad (6.9) \]

and the next step is to solve the equations with respect to the unknowns \( x \) and \( u \)

\[ Lx = F \quad (6.10) \]
\[ L^Tu = x. \quad (6.11) \]

The lower triangular matrix \( L \) has the same bandwidth as \( K \).

### 6.3 Stochastic Finite Element Analysis

One of the major drawbacks of finite element analysis is due to the fact that the materials are modelled to be homogeneous. In reality, however, most materials are not. Especially paint, being a mixture of medium and pigments in very inhomogeneous. A way to overcome this problem is using stochastic finite element analysis, in which the spatial variability of the material properties can be modelled [16, 23].

Although these techniques were not used in this research, a short introduction is given here, because this might be a good starting point for future research. In the following section two methods will be described, the von Neumann expansion method and the perturbation method.

#### 6.3.1 Monte Carlo simulation and the Neumann expansion method

Under the assumption that \( K \) contains parameters which are subjected to spatial variabilities, the solution method in which Equations 6.9 and 6.10 are applied sample by sample is called the direct Monte Carlo method. Because, in this case the Cholesky decomposition has to be done for each sample, which requires a large amount of computer time, it is, in general better to use the Neumann expansion method. The stiffness matrix \( K \) is then decomposed into two matrices

\[ K = K_0 + \Delta K, \]
6.3. STOCHASTIC FINITE ELEMENT ANALYSIS

where $K_0$ represents the stiffness matrix in which the spatially variable parameters are replaced by their representative values (e.g. mean values or medians) and $\Delta K = K - K_0$. The Neumann expansion of $K^{-1}$ takes the following form

$$K^{-1} = (K_0 + \Delta K)^{-1} = (I - P + P^2 - P^3 + \ldots)K_0^{-1},$$

with

$$P = K_0^{-1}\Delta K.$$

Then the vector $u$ can be written as the following series, where $u_0 = K_0^{-1}F$,

$$u = u_0 - Pu_0 + P^2u_0 - P^3u_0 + \ldots = u_0 - u_1 + u_2 - u_3 + \ldots$$

This expansion series may be terminated after a few terms, provided that it converges. A convergence criterion that can be used is

$$\frac{||u_i||_2}{\sum_{k=0}^{i} -1^k u_k}_2 \leq \delta_{err},$$

where $\delta_{err}$ is the allowable error.

These series of solution is equivalent to the following recursive equation

$$K_0u_i = \Delta Ku_{i-1} \quad (i = 1, 2, \ldots).$$

The most outstanding feature of this approach in the case of Monte Carlo simulation for the spatial variation of material properties is that matrix factorization is required only once for all samples.

6.3.2 Perturbation method

Another way to deal with spatial variation of the material properties is the **perturbation method**. Let $\alpha_i (i = 1, 2, \ldots, k)$ be the random variables that represent the spatial variation of the material properties. Then, $K$ can be expanded in the following form with the assumption that each $\alpha_i$ is small ($\alpha_i \ll 1$) and has zero mean:

$$K = K^0 + \sum_{i=1}^{k} K^i_i \alpha_i + \frac{1}{2} \sum_{i=1}^{k} \sum_{j=1}^{k} K_{ij}^{ij} \alpha_i \alpha_j + \cdots,$$

in which $K^0$ is the stiffness matrix evaluated at $\alpha = (\alpha_1, \ldots, \alpha_k) = 0$, and

$$K^i_i = \frac{\partial K}{\partial \alpha_i} \bigg|_{\alpha=0}, \quad K_{ij}^{ij} = \frac{\partial^2 K}{\partial \alpha_i \partial \alpha_j} \bigg|_{\alpha=0}.$$
The external force vector $F$ may also involve the random variables $\alpha_i$, and then $F$ can be expanded in the same way. And, even so, the unknown displacement vector $u$:

$$
\mathbf{u} = \mathbf{u}^0 + \sum_{i=1}^{k} \mathbf{u}_i^i \alpha_i + \frac{1}{2} \sum_{i=1}^{k} \sum_{j=1}^{k} \mathbf{u}^{ij} \alpha_i \alpha_j + \cdots,
$$

in which the coefficient vector can be represented by the following set of recursive equations:

$$
\begin{align*}
\mathbf{u}^0 &= (K^0)^{-1} F^0 \\
\mathbf{u}^1 &= (K^0)^{-1} (F^1 - K^1 \mathbf{u}^0) \\
\mathbf{u}^{II} &= (K^0)^{-1} (F^{II} - K^2 \mathbf{u}^1 - K^1 \mathbf{u}^1 - K^{II} \mathbf{u}^0)
\end{align*}
$$

### 6.4 Conclusion

Mecklenburg and Tumosa showed that finite element analysis can be used to compute the stress development in a painting when subjected to environmental changes. In the following two chapters their models and extensions of these will be used to give an idea of how this can be done.

FEA is used extensively in the industry to compute stress development in complex structures. Therefore, a lot of standard programs are available. However, one of the major drawbacks of this method is the fact that throughout each element the materials are modelled to be homogeneous. Hopefully, in the future this problem can be overcome by using stochastic finite element analysis.
Chapter 7

The MARC system

7.1 Introduction

To apply the FEA as explained in Chapter 6 to actual models of paintings we used an existing standard program called the MARC system which is developed by MARC Analysis Research Corporation. The MARC system was chosen because it has a wide variety of features. Furthermore, it contains an easy to use graphical user interface, and last but not least, the Centre for High Performance Applied Computing in Delft has a licence for the use of the program MARC, and therefore, employees of the Delft University of Technology can use it on one of the Cray computers.

The MARC system contains two integrated programs MARC and Mentat that facilitate analysis of engineering problems in the field of structural mechanics, heat transfer and electromechanics. Mentat has a graphical user interface and can be used for pre- and postprocessing, whereas MARC does the actual analysis (Figure 7.1). This chapter describes the MARC system and gives an overview of the possibilities of the program that will be of interest in our research.

7.2 Mentat

Mentat is an interactive computer program that prepares and processes data for use with the finite element method. The program verifies keyboard input and returns recommendations or warnings when it detects questionable input. The Mentat program can process both two- and three-dimensional meshes to do the following:

- generate and display a mesh,
- generate and display boundary conditions and loadings, and
- perform postprocessing to generate contour, deformed shape and time history plots.

The data that is processed includes

- nodal coordinates,
- element connectivity,
- nodal boundary conditions,
- nodal coordinate systems,

53
• element material properties,
• element geometric properties,
• element loads,
• nodal loads/non-zero boundary conditions, and
• element and nodal sets.

When the model is set up with Mentat it can be saved as a .mud (binary) or .mfd (ASCII) file for further use in Mentat or as a data file with a .dat extension for analysis with MARC.

7.3 MARC

MARC has four comprehensive libraries, making the program applicable to a wide range of uses:

The Procedure Library contains procedures such as static, dynamic and creep analysis.

The Material Library includes many material models that represent most engineering materials. For example, linear elasticity, plasticity and viscoelasticity. All properties may depend on temperature.

The Element Library contains over 140 elements.

The Program Function Library contains subroutines in FORTRAN designed to speed up and simplify analysis work.
A MARC data file contains three logically distinct sections: the parameters which describe the problem size and type, the model definition options which give a detailed description of the problem and the history definition options which describe the load history.

During the analysis the program produces a log file and an output file and a file that can be used for postprocessing with Mentat. This last file will have a .t16 (binary) or .t19 (ASCII) extension.

### 7.4 An example

Two test runs were done to get to know the program. The material parameters given by Mecklenburg and Tumosa [25] were used to verify the models by comparing them with their results. Both tests modelled a painting consisting of three layers, canvas, glue and Naples yellow oil paint. In the first test the painting was subjected to a temperature drop from 23°C to -3°C at constant relative humidity of 5%. In the second test the painting was desiccated from 70% RH to 10% RH at a constant temperature of 23°C. The exact model parameters can be found in Appendix A.

For the FEA we used 8-noded elements with a trilinear interpolation function. Here follows an example of a data file produced with Mentat for the first test. However, the dimensions of the problem were adjusted, particularly, the thickness of the layers, to clarify the structure of the mesh as given in Figure 7.2. Furthermore, the number of elements given in this example is less than the number actually used in the test runs because of space considerations.

The first part of the data file gives the problem type and size:

```plaintext
title test1
$ .... MARC input file produced by Mentat 3.2.0
sizing 100000 27 64 192
elements 7
processor 1 1 1
$no list
all points
setname 3
end
$..................
```

Then, after two lines concerning the solution algorithm and optimization, follows the information about the mesh. The connectivity is defined by giving the type number and the nodes belonging to each element. The definition of the mesh is then completed by giving the coordinates of each node.

```plaintext
solver
  0 0 0
optimize 9
connectivity
  1 7 1 21 23 17 5 22 24 18
  2 7 17 23 25 19 18 24 26 20
```
Figure 7.2: Mesh of a simple model of a three-layered structure with 27 elements. The arrows represent the boundary conditions.
7.4. AN EXAMPLE

coordinates

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</tbody>
</table>
CHAPTER 7. THE MARC SYSTEM

The next part defines the material properties. All three layers are modelled as isotropic materials, and the elastic modulus, the Poisson ratio and the thermal expansion coefficient are given, and, of course, the labels of the elements that model the material. For the glue and the paint layer the effect of the temperature on the elastic modulus is also given. This effect is assumed to be linear, therefore, it is sufficient to give the elastic modulus at the initial and at the final temperature.

isotropic

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isotropic

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isotropic

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temperature effects data

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</table>

The fourth part defines the initial loads. That is, the initial temperature and the boundary condition that models the frame of the painting. The assumption is made that only the nodes directly at
7.5. THE RESULTS OF THE TEST RUNS

The results of the test runs can again be viewed with the Mentat program. An example of the possible output produced by Mentat is given in Figure 7.3, where we can see the direction of the maximum principal stress in each of the elements in the paint layer for the first test run.
CHAPTER 7. THE MARC SYSTEM

Table 7.1: Maximum principal stresses for painting subjected to a temperature drop from 23°C to -3°C.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Maximum Principal Stress</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paint</td>
<td>1.995 – 1.998 MPa</td>
</tr>
<tr>
<td>Glue Size</td>
<td>3.250 – 3.260 MPa</td>
</tr>
<tr>
<td>Canvas</td>
<td>2.260 – 2.270 MPa</td>
</tr>
</tbody>
</table>

Note that, although the maximum principal stress is represented by an arrow, it is not a vector, but a tensor component in the principal coordinate system of that particular element. The arrow just gives the direction of one of the principal coordinate axes. The range of the values of the maximum principal stress for the first test are given in Table 7.1. The quantities given are the values of the stress at the upper surface of each layer. Due to continuity the stresses at the upper surface of the glue size and the canvas are equal to the stresses at the lower surface of the paint and the glue size, respectively.

Table 7.2: Maximum principal stresses for painting subjected to desiccation from 70% RH to 30% RH.

<table>
<thead>
<tr>
<th>Layer</th>
<th>Maximum Principal Stress</th>
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</thead>
<tbody>
<tr>
<td>Paint</td>
<td>0.912 – 0.915 MPa</td>
</tr>
<tr>
<td>Glue Size</td>
<td>18.62 – 18.64 MPa</td>
</tr>
<tr>
<td>Canvas</td>
<td>18.32 – 18.33 MPa</td>
</tr>
</tbody>
</table>

The maximum principal stresses in the paint layer as computed by Mecklenburg and Tumosa ranged from 2.09 MPa to 2.11 MPa [25]. It is hard to say how this difference with our results is caused, because they give no information on the type of elements and the boundary conditions used. The directions of the maximum principal stresses is similar to our results. Their conclusion is that if the paint film cracks, the crack pattern will be perpendicular to the direction of the maximum principal stresses. However, this conclusion seems a bit hasty, because the values of the second principal stresses are only 0.1% less than the maximum principal stresses.
Figure 7.3: The directions of maximum principal stresses for a painting subjected to a temperature drop from 23°C to −3°C.
8.1 Introduction

The material parameters of paint as used by Mecklenburg and Tumosa in their simulations, were obtained from experiments on 13 year old paint [25]. One of the main objectives of the MOLART project is to study the effects of ageing of paint on the present condition of a painting. However, experimental data on the mechanical properties of aged paint is hardly available. Nevertheless, by using the material properties of polyacrylate and zinc polyacrylate as given by Holliday [18], simulations were done to show the possible effects of the ageing of paint on the stress development in a painting. The results of these simulations are given in Section 8.2 of this chapter.

To give an even better idea of the possibilities of FEA, the second section of this chapter describes the results of simulations inspired by the fact that a few months ago the painting 'Cathedra' by Barnett Newman was cut with a knife in 'Het Stedelijk Museum' in Amsterdam. With the use of FEA we tried to establish the stress development in a painting on canvas when it is stretched on a frame, the results of which can be found in Section 8.3, and in a painting with a tear in the canvas as well as when the tear is sewed with a material stiffer or less stiff than the original painting. The results of these last simulations are given in Section 8.4.

8.2 The effects of ionic bonds

With the model as used in the test runs as a basis (see Chapter 7), some calculations were done to obtain an idea on the influence of ageing, or more specific, of ionic bonds on the stress development in a painting subjected to a temperature drop. The dimensions of the model used were the same as in the test runs, as were the material properties for the two lower layers (the canvas and the glue layer). The paint layer was modelled as a polyacrylic acid and as a zinc salt of polyacrylic acid. The mechanical behaviour of these materials were described in Chapter 5.

The resulting maximum principal stress in the paint layer is given in Table 8.1. Because the variation in the maximum principal stress in the paint layer is relatively small, only the average values are given. More exact values, as well as the stresses in the other two layers can be found in Appendix B.

As we can see, the stress developed in the paint layer in case of the zinc salt is less than half of the stress developed in the paint layer in case of the polyacrylic acid without the ionic bonds.
CHAPTER 8. MODELLING STRESS DISTRIBUTIONS IN PAINTINGS

<table>
<thead>
<tr>
<th>Material</th>
<th>Elastic Modulus (Pa)</th>
<th>Thermal Coefficient (°C⁻¹)</th>
<th>Maximum Principal Stress (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polyacrylic acid</td>
<td>13 · 10^9</td>
<td>5.5 · 10⁻⁵</td>
<td>2.6 · 10^7</td>
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<tr>
<td>Zinc salt of polyacrylic acid</td>
<td>19.5 · 10^9</td>
<td>1.4 · 10⁻⁵</td>
<td>1.0 · 10^7</td>
</tr>
<tr>
<td>Using Barker’s relation</td>
<td>19.5 · 10^9</td>
<td>4.49 · 10⁻⁵</td>
<td>3.2 · 10^7</td>
</tr>
</tbody>
</table>

Table 8.1: Maximum Principal Stresses

This is due to the fact that the thermal expansion coefficient of the zinc salt is much lower than expected. This is even more obvious if we compare this with the stress when we model the paint layer as a material having the elastic modulus of the zinc salt and satisfying Barker’s relation (see Equation 5.1) for thermal expansion coefficients.

8.3 Stretching a painting on a frame

Before modelling a painting with a tear in the fabric, we wanted to know the effects of stretching a painting on a frame on the stress levels in the canvas. To do so, we used a plane stress model of a painting with dimensions 2m x 4m. And the boundary conditions were chosen such that the painting was stretched 2cm in both directions, i.e., all nodes on the outside edge of the painting were modelled to have an initial strain of 1cm.

The model is orthotropic, i.e. there are three different elastic moduli, one in the warp direction of the fabric, one in the weft direction of the fabric and one shear modulus. We used two different orientations. In the first (Orientation I) the warp of the fabric was directed horizontally, and in the second (Orientation II) the warp of the fabric was directed vertically.

The tensile elastic moduli used were provided by William Wei, and came from experiments done at the 'Instituut Collectie Nederland' on cotton, linen and a canvas made from cotton and linen with a glue sizing and a ground layer (see Table 8.2). The Poisson ratio was chosen to be 0.3 and the shear moduli were chosen to be equal to the tensile moduli in the warp direction.

The results are given in Table 8.2. The corner nodes were modelled to have an initial strain of 1cm in two directions, which caused the high stress levels in the corners of the painting.
8.4 A painting with a tear

To model a painting with a tear in the fabric we used a plane stress model, i.e., a two-dimensional model, with a mesh as shown in Figure 8.1. Around the tip of the tear the 6-noded triangular elements were used and the nodes on the midsides adjacent to the tip are moved to one quarter of the edge length (see Figure 8.2), throughout the rest of the painting, 8-noded rectangular elements were used.

The boundary conditions and the dimensions are chosen equal to those of the models in the previous section. The fabric was chosen to be the grounded cotton with linen in orientation I. The tear was modelled to be 1 cm wide. The calculations were done for the model with an open tear, a closed tear, that is, a model of the original painting, and for models with a tear filled with different types of material.

The resulting maximum principal stresses at the tip of the tear are shown in Table 8.3. More detailed results can be found in Appendix B.3. When merely looking at the values as given in Table 8.3, the sewing of the tear does not seem to have a great influence on the stress levels, when compared to the stress levels in the model of the original painting. Contrary to the stress level at the tip of the tear in the model with the open tear, which is more than twice as high as the stress in the original painting. But when examining the stress distribution in the vicinity of the tip of the tear as shown in Figures 8.3, 8.4 and 8.5, one notices the considerable higher maximum principal stress within the tear when it is filled with a material with a high elastic modulus.
66

CHAPTER 8. MODELLING STRESS DISTRIBUTIONS IN PAINTINGS

Figure 8.2: The elements around the tip of the tear

<table>
<thead>
<tr>
<th>Material, Filling the Tear</th>
<th>Elastic Modulus of the Material (Pa)</th>
<th>Maximum Principal Stress at the Tip of the Tear (Pa)</th>
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<tr>
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<tr>
<td>Filled III</td>
<td>12.5 \cdot 10^9</td>
<td>3.02 \cdot 10^7</td>
</tr>
</tbody>
</table>

Table 8.3: Maximum principal stresses at the tip of the tear
Figure 8.3: Maximum principal stresses near the tip of the tear
Figure 8.4: Maximum principal stresses near the tip of the tear, where the tear is filled with a material with $E = 12.5 \cdot 10^9$. 
Figure 8.5: Maximum principal stresses near the tip of the tear, where the tear is filled with a material with $E = 12.5 \cdot 10^7$
Chapter 9

Stochastic Modelling of Fatigue

9.1 Introduction

Elastic deformation is a reversible phenomenon, that is, if the stress does not reach a critical level (and no fracture occurs), the material will deform back to its original shape. When we model a painting as behaving linear elastic, changes in temperature as well as relative humidity will do no damage whatsoever, when kept in a small enough range.

In reality, this is not always true. Especially on a microscopic level. When subjected to time-varying loadings a material's ability to carry the intended loading will deteriorate. This phenomenon is called fatigue.

Fatigue failure, generally, consists of two phases. The first one includes the micro-damage accumulation, cracking and deterioration on the micro-level. This phase terminates when a macro crack is born as a result of coalescence of micro cracks, micro voids and other micro defects. The second stage is in fact the macroscopic fatigue crack propagation up to the final failure. Fatigue crack growth is the result of interaction between the micro-damage near the crack tips and the stress field in the cracked bodies. In the last decade many stochastic models were developed for fatigue crack growth [29, 30].

All the phenomena related to fatigue, i.e. crack initiation and growth, as well as the final failure considered as a loss of integrity, instability, or both, are subjected to the influence of stochastic factors. These factors may be classified in several ways. For example, one may divide them into three groups: the randomness of material properties; random defects and imperfections of structural components; random loads, actions and environmental conditions.

This chapter describes the different models for representing fatigue damage, and it gives a stochastic model for fatigue life prediction when a material is subjected to a random load. The chapter ends with a short survey of other types of stochastic models, which deal with random initial defects, and random material properties.

9.2 Representations of fatigue damage

9.2.1 S-N data

The general approach in fatigue life prediction is to relate the fatigue life of a construction, subjected to a random load, to laboratory fatigue experiments of simple specimens subjected to a cyclic load with constant amplitude, so-called S-N data. The S-N curve displays the stress
amplitude, $S$, versus the number of cycles to failure, $N$. Analytical representation of the S-N curves is commonly assumed to have the form

$$N = kS^{-b},$$

where $k$ and $b$ are material parameters estimated from test data.

In situations with a variable amplitude loading we can estimate fatigue life using these S-N curves and the Palmgren-Miner rule. This rule asserts that the total damage $D$ is a linear accumulation of the partial damage done by each cycle. The damage done by a loading cycle with stress amplitude $S_i$ can be given as

$$\frac{1}{N_i},$$

where $N_i$ is the number of cycles to failure at this stress amplitude and can be found from the S-N curve. Let $n_i$ denote the number of actual loading cycles with stress amplitude $S_i$, then the Palmgren-Miner rule gives the total accumulated damage:

$$D = \sum_i \frac{n_i}{N_i}. \tag{9.1}$$

### 9.2.2 Continuum Damage Mechanics

For fatigue damage we can also use the continuum damage mechanics model. Continuum damage mechanics was developed mainly by Kachanov [19]. The damage variable is considered as a surface density of intersections of cracks and cavities. Let us consider a certain section $S$ of the body. Let $A_0$ be the initial area of the undamaged section. As a result of damage, a certain part of the section is fractured (lost). Denote the lost area by $A$.

In case of isotropic damage, the cracks and voids are equally distributed in all directions. The damage variable can be considered as a scalar function of the time $t$ and is defined by

$$D(t) = \frac{A(t)}{A_0}.$$

We have that $0 \leq D(t) \leq 1$ and $\omega$ is a positive monotonically increasing function. For undamaged material $D = 0$ and at fracture $D = 1$.

In case of uniaxial tension, we define the notion of actual stress as

$$\sigma_a = \frac{F}{A_0 - A} = \frac{F}{A_0(1 - \omega)} = \frac{\sigma}{\psi},$$

where $\sigma$ is the nominal stress and $\psi = 1 - D$ is called the 'continuity'.
9.2. REPRESENTATIONS OF FATIGUE DAMAGE

Now, the assumption is made that the strain response of the body is modified by damage only through the actual stress and that the rate of damage growth is determined primarily by the level of the actual stress. Hence, the elastic strain of a damaged material is given by

\[ e = \frac{\sigma_0}{E} = \frac{1}{E\psi} \sigma. \]

So, the Hooke's law here has its usual form with the elastic modulus \( E \) being replaced by \( E\psi \).

Assuming that the evolution of the function \( D(t) \) depends only on the values of \( D(t) \) at some initial time instant and on the external actions \( Q(t) \), one can postulate the following general differential equation model for damage accumulation \( D(t) \):

\[ \frac{dD(t)}{dt} = f(D(t), Q(t)). \quad (9.2) \]

Kachanov gave a simple form of this kinetic equation in the case of uniaxial tension [19]:

\[ \frac{dD(t)}{dt} = -A \left( \frac{\sigma}{\psi} \right)^n, \]

where \( A > 0 \) and \( n \geq 1 \) are material constants.

Continuum damage models can be used to describe the evolution of a crack pattern. Then the damage variable is a measure of the density of the cracks. Or, on a microscopic level, it can be used to describe the accumulation of micro cracks into a macro crack.

9.2.3 Crack growth models

In models for crack growth, a more specific way to represent damage is used. In most cases the crack length of just one crack is modelled and fatigue crack growth can be characterised by

\[ \frac{d\alpha}{dN} = F(\alpha, S, C, \theta, \zeta), \quad (9.3) \]

where
- \( \alpha \) denotes the length of the dominant crack,
- \( S \) denotes the state of stress,
- \( C \) symbolically denotes the material properties,
- \( \theta \) denotes the temperature,
- \( \zeta \) symbolically denotes all other parameters, and
- \( N \) denotes the number of cycles, as a continuous variable, (or continuous time) corresponding to the crack length \( \alpha \).

Unfortunately, there is not enough knowledge concerning the influence of each of the preceding variables on fatigue crack growth, and analytical forms of the general relations like Equation 9.3 are lacking even for traditional fatigue problems [30].
In Chapter 4 it was already shown that the stress intensity factor $K$ (see Equation 4.10) is a suitable quantity to characterise crack growth in (linear) elastic materials. And in models for fatigue crack growth the stress intensity factor – or more specifically – the stress intensity range $\Delta K = K_{\text{max}} - K_{\text{min}}$ is used as well.

**Example 9.1 (Paris-Ergodan equation)** A commonly used equation for the crack growth rate is the Paris-Ergodan equation, which is given by

$$\frac{da}{dN} = C(\Delta K)^m,$$

where $C$ and $m$ are regarded as material constants.

### 9.3 Random fatigue loads

To predict the life of a component subjected to complex loading histories, a loading is usually reduced to a sequence of events that can be regarded as compatible with constant amplitude fatigue data. The methods that make such reductions possible are known as cycle counting techniques.

Generally, a cycle is defined by specifying its highest and its lowest point, $M$ and $m$, $M \geq m$. A cycle count is a procedure transforming a function $s(t)$, $0 \leq t \leq T$ into a set of cycles $(M_i, m_i)_{i \in I}$, for a certain index set $I$. Usually, we have that the $M_i$'s are the local maxima of the function $s(t)$.

**Example 9.2 (Peak-valley cycle count)** Let $s(t)$, be a continuous function and let $(M_i)_{i \in I}$ be the set of all local maxima of $s(t)$, then the peak-valley cycles are defined by $(M_i, m_i)_{i \in I}$, where $m_i$ is the local minimum following $M_i$.

**Example 9.3 (Rainflow cycle counting algorithm)** Let $s(t)$, $0 \leq t \leq T$, be a continuous function. For any fixed point $t_0 \in [0, T]$, let $t^+$ be the time of the first up-crossing of the level $u$ after $t_0$, or $t^+ = T$ if no such up-crossing exists in $(t_0, T]$. And let $t^-$ be the time of the last down crossing before $t_0$, or $t^- = 0$ if no such down-crossing exists in $(0, t_0]$. Let us define

$$H^-(t_0) = \max_{t^- < t < t_0} \{s(t_0) - s(t)\},$$

$$H^+(t_0) = \max_{t_0 < t < t^+} \{s(t_0) - s(t)\}.$$

The rainflow cycle amplitude $H(t)$ at time $t$ is defined as (see figure 9.1)

$$H(t_0) = \min\{H^-(t_0), H^+(t_0)\}.$$
9.3. RANDOM FATIGUE LOADS

\[ H^+ = H(t_0) \]

\[ \Delta H \]

\[ s(t_0) = u \]

\[ t^- \]

\[ t_0 \]

\[ t^+ \]

Figure 9.1: Rainflow cycle amplitude

Hence, for every \( t \in [0, T] \), we can define a rainflow cycle as

\[(M_t, m_t) = (s(t), s(t) - H(t)).\]

For smooth loads (\( C^2 \)-functions) a rainflow cycle only has a non-zero amplitude when \( M_t \) is a local maximum [29].

When we reduce the function of the loading history \( s(t), t \in [0, T] \), into a set of cycles \( \{(M_t, m_t)\}_{t \in T} \), we can generalise the Palmgren-Miner rule (9.1). Denote by \( f(x, y) \) a damage caused by a cycle \((x, y)\). Assume \( f(x, y) \geq 0 \) is increasing in \( x \) and decreasing in \( y \). If the set of cycles with positive amplitudes is countable, the total damage \( D(T) \) is given by

\[ D(T) = \sum_{i \in I_T} f(M_i, m_i). \]

If the function \( f(x, y) \) is known explicitly, we can also derive a different formula for the total damage \( D(T) \), using the counting distribution function of the cycle count. When \( s(t) \) is a continuous function with a countable set of cycles \( \{(M_i, m_i)\}_{i \in I_T} \) on \([0, T]\), then the counting distribution function \( N_T(x, y) \) is defined by

\[ N_T(x, y) = \# \{ i \in I_T | M_i > x \geq y > m_i \}. \]

**Theorem 9.4** [29] Assume \( f(x, y) \) is twice continuously differentiable. If \( f(x, x) = 0 \) and \( N_T(u, u) \) is a bounded function of \( u \), the total damage is finite and given by

\[ D(T) = - \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} N_T(x, y) \frac{\partial^2 f(x, y)}{\partial x \partial y} dy dx - \int_{-\infty}^{+\infty} N_T(x, x) \left. \frac{\partial f(x, y)}{\partial y} \right|_{y=x} dx. \]
The fatigue failure time $T^f$ is defined by

$$D(T^f) = 1.$$ 

Hence, the failure probability is given by

$$\mathbb{P}\{T^f \leq T\} = \mathbb{P}\{D(T) \geq 1\}.$$ 

The expected damage can be written as

$$E(D(T)) = \sum_{i \in \mathcal{I}_T} E(f(M_i, m_i)),$$

or, using Theorem 9.4, as

$$E(D(T)) = \int_{-\infty}^{+\infty} \int_{-\infty}^{\infty} E(N_T(x, y)) \frac{\partial^2 f(x, y)}{\partial x \partial y} dy dx - \int_{-\infty}^{+\infty} E(N_T(x, x)) \frac{\partial f(x, y)}{\partial y} \bigg|_{y=x} dx. \tag{9.4}$$

Calculation of $E(f(M_i, m_i))$ is very difficult because even if $s(t)$ is a stationary process, the set of all local maxima $(M_i, m_i)_{i \in \mathcal{I}_T}$ could be non-stationary (i.e., $E(f(M_i, m_i)) \neq E(f(M_{i+1}, m_{i+1}))$). However, there are cases in which it is possible to calculate, or at least approximate, $E(N_T(x, y))$.

### 9.3.1 Rainflow Count on Smooth Ergodic Loads

In this section we assume that the loads are stationary and ergodic processes with finite intensity of local maxima, $c_M$. Let $(M_i, m_i)_{i \in \mathcal{I}_T}$ be the set of all local maxima, $M_i$, and the following minima, $m_i$, of $s(t)$ on $[0, T]$ (i.e., this is the peak-valley cycle count of $s(t)$), then

$$c_M = \lim_{T \to \infty} \frac{|(M_i, m_i)_{i \in \mathcal{I}_T}|}{T} < \infty.$$

And let $(M_i, m_i^{RFC})_{i \in \mathcal{I}_T}$ denote the rainflow cycle count. Denote by $N_T^{RFC}(u, v)$ and $N_T^{PV}(u, v)$ the counting distributions of the rainflow count and the peak-valley count respectively, and define the count intensities by

$$\mu^{RFC}(u, v) = \lim_{T \to \infty} \frac{N_T^{RFC}(u, v)}{T},$$

and

$$\mu^{PV}(u, v) = \lim_{T \to \infty} \frac{N_T^{PV}(u, v)}{T},$$

respectively.
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The counting intensity of the peak-valley count can simply be derived using the ergodicity of the load:

\[
\mu_{PV}(u, v) = \lim_{T \to \infty} \frac{\#(\{M_i > u, m_i < v\})_{i \in I_T}}{T}
\]

\[
= \lim_{T \to \infty} \frac{\#(\{M_i > u, m_i < v\})_{i \in I_T}}{\#(\{M_i\})_{i \in I_T}}
\]

\[
= c_M \text{Prob}(M_1 > u, m_1 < v),
\]

where Prob is the ergodic probability.

The rainflow count intensity also depends upon the sequence of local extremes, but now we need the following lemma about the counting distribution.

**Lemma 9.5** Let \( s(t), t \in [0, T], \) be a continuous function and \( \{M_i, m_i\}_{i \in I_T} \) the set of rainflow cycles, then

\[
N_T(u, v) = \#\{t \in [0, T] | t \text{ is a } u \text{-downcrossing and } s(t) - H^+(t) < v\}
\]

\[
= \#\{t \in [0, T] | t \text{ is a } u \text{-downcrossing and } s(t + \cdot) \text{ crosses } v \text{ before } u\}.
\]

Using this lemma, we get

\[
\mu_{RFC}(u, v) = \lim_{T \to \infty} \frac{\#(\{M_i > u, m_i^{RFC} < v\})_{i \in I_T}}{T}
\]

\[
= c_M \lim_{T \to \infty} \frac{1}{\#(\{M_i\})_{i \in I_T}} \#\{M_i > u, m_i < u \text{ and } \{m_{i+1}\}_{i \geq 1} \text{ crosses } v \}
\]

\[
\text{before } \{M_{i+1}\}_{i \geq 2} \text{ crosses } u
\]

\[
= c_M \text{Prob}\left\{M_1 > u, m_1 < u \text{ and } \{m_{i+1}\}_{i \geq 1} \text{ crosses } v \text{ before } \{M_{i+1}\}_{i \geq 2} \text{ crosses } u \right\}
\]

\[
= c_M \int_u^\infty \text{Prob}\left\{m_1 < u \text{ and } \{m_{i+1}\}_{i \geq 1} \text{ crosses } v \text{ before } \{M_{i+1}\}_{i \geq 2} \text{ crosses } u \right\} f_{M_1}(x) dx,
\]

(9.5)

where \( f_{M_1} \) is the ergodic density function.

### 9.3.2 Markov extremes

To give an approximation of \( \mu_{RFC}(u, v) \), we assume that the sequence of local extremes form a stationary Markov chain and that the extremes can take only a finite number of discrete values. Let \( \{M^D_i, m^D_i\}_{i \in I_T} \) denote the sequence of (discrete) local maxima and following local minima. Let \( u_1 > u_2 > \cdots > u_n \) be the discrete levels that \( M^D_i \) and \( m^D_i \) can take, and let \( P = (p_{ij}) \) and \( \bar{P} = (\bar{p}_{ij}) \) be the transition probability from maximum to the following minimum and from minimum to the following maximum, respectively, i.e.,

\[
p_{ij} = P\{m^D_k = u_j | M^D_k = u_i\},
\]

\[
\bar{p}_{ij} = P\{M^D_{k+1} = u_j | m^D_k = u_i\}.
\]
For fixed $i$ and $j$, $i \leq j$, the discrete version of Equation 9.5 is given by

$$
\mu^{\text{RFC}}(u_i, u_j) = c_M \sum_{k=1}^{i-1} \text{Prob} \left\{ m^D_1 < u_i \text{ and } \{ m^D_l \}_{l \geq 1} \text{ crosses } u_j \text{ before } \{ M^D_l \}_{l \geq 2} \text{ crosses } u_i \mid M^D_1 = u_k \right\} \mu_{\text{MD}}(u_k),
$$

where

$$
\mu_{\text{MD}}(u_k) = \lim_{T \to \infty} \frac{\# \{ i \in \mathcal{I}_T \mid m^D_i = u_k \}}{\# \{ M^D_i \}_{i \in \mathcal{I}_T}}.
$$

The sequence of maxima $\{M_i\}_{i \in \mathcal{I}_T}$ is also a Markov chain with transition matrix $P$. Assume that this chain of maxima is ergodic and let $\pi$ denote the stationary distribution.

The conditional probability in Equation 9.6 can be written as the sum

$$
\sum_{r=0}^{\infty} p_r(u_k),
$$

with

$$
p_0(u_k) = \mathbb{P} \{ m^D_1 < u_j \mid M^D_1 = u_k \},
$$

$$
p_r(u_k) = \mathbb{P} \{ m^D_{r+1} < u_j, u_j \leq m^D_{r-1} < M^D_i < u_i \text{ for all } l = 2, 3, \ldots, r+1 \mid M^D_i = u_k \},
$$

for fixed $i$ and $j$. Consequently the rainfall count intensity becomes,

$$
\mu^{\text{RFC}}(u_i, u_j) = c_M \sum_{k=1}^{i-1} \tau_k \left( \sum_{r=0}^{\infty} p_r(u_k) \right).
$$

The intensity of the maxima $c_M$ can be difficult to obtain, but when the counting intensity is computed for a discrete Markov chain, $\mu^{\text{RFC}}_{\text{Markov}}(u, v)$, assuming $c_M = 1$, it can be renormalized using the downcrossing intensity of the load $\mu(u) = \mu^{\text{RFC}}(u, u)$ in the following way:

$$
\mu^{\text{RFC}}_{\text{Markov}}(u, v) = \frac{\mu(0)}{\mu^{\text{RFC}}_{\text{Markov}}(0, 0)} \mu^{\text{RFC}}_{\text{Markov}}(u, v).
$$

Then, we can approximate the expected damage by replacing $\mathbb{E}(N_T(u, v))$ by $\mu^{\text{RFC}}_{\text{Markov}}(u, v)$ in Equation 9.4.
When this project started MOLART's main interest was to decide how the chemical and physical alteration of ageing paint would effect the stress distribution and the formation of craquelure on paintings. However, despite the numerous research done on the chemical ageing of paint, very little is known about changes in mechanical behaviour with age.

Chemical research done within the MOLART project indicated that during ageing the pigment molecules in oil paint will form ionic bonds with the crosslinked network of the medium. It was possible to do some simulations by modelling a fresh paint layer consisting of polyacrylic acid and an aged paint layer consisting of a zinc salt of polyacrylic acid. These simulations showed again the importance of understanding the material properties. The hypothesis that the stress levels developed upon cooling would be higher for the zinc salt, because of the fact that the ionic bonds would increase the elastic modulus of the material, turned out to be false, due to the fact that the thermal expansion coefficient of the zinc salt was much lower than expected. As a consequence the stress in this material remains low, despite an increase in stiffness.

To further explore the possibilities of using finite element analysis (FEA) to compute stress distributions in paintings, simulations were done on a model of a painting with a tear in the canvas. These simulations were inspired by the fact that the ‘Cathedra’, a painting by Barnett Newman, was cut several times. The simulations show that when such a tear is sown with a material with a elastic modulus 10 times that of the original canvas, the maximum principal stress around the tip of the tear decreases, whereas the maximum principal stress inside the tear, i.e. in the sewing material, becomes almost twice as high. For a sewing material with an elastic modulus 10 times less than the original canvas the opposite is true. In this approximation, a choice for a relatively stiff repair material creates the benefit of a lower chance of tearing of the primary cut.

During this research project it became clear that finite element analysis is a good method for understanding the effects of changing mechanical properties of ageing art materials on the stress development in paintings, provided that these mechanical properties are known. The material parameters needed to perform a finite element analysis are the elastic modulus and, when one is interested in the effects of environment, the thermal and the moisture expansion coefficient. In the future, it might also be interesting to enter a time factor into the models, but in that case the stress-relaxation behaviour of the material must be known, as well.

The drawbacks of FEA are first of all the fact that the materials are modelled to be homogeneous throughout each element, whereas paint is a mixture of pigments and medium, and by definition, non-homogeneous. Secondly, to decide whether fracture will occur, more material parameters
are needed, as for instance the strength. And although the directions of the maximum principal
stress as computed by FEA can give an indication of how the global crack pattern might develop,
it gives no clue about the more local features of the craquelure pattern.

The first problem could be tackled by using stochastic finite element analysis, in which it is
possible to model spatial variabilities in the material parameters and their effects on the stress
distribution in a painting. Another possibility, which deals with inhomogeneity on a micro­
scopic level might be to use a model described by Boriek et al [1], that computes the distribution
of voids in polymeric materials and their effect on the stresses in these materials using FEA.

Micro-voids can cluster and eventually grow into a macro crack when subjected to constant or
cyclic stress. Such damage accumulation from micro-voids to a macro crack can be described
by continuum damage models. It will be necessary to understand the mechanical behaviour
on a microscopic level if such models are to be used in the future. It would be interesting to
apply MOLART’s knowledge on the molecular structure of aged paints for development of such
a model.

Accumulation of damage can be the result of cyclic loading. Even if the stress does not reach the
critical value during each individual cycle, repeated stress peaks will deteriorate the material's
ability to carry the intended load. Hence, every loading cycle will increase the damage with a
certain amount. If we know the damage as a result of individual cycles of all possible stress
amplitudes, it will be possible to compute the expected damage in certain time intervals by
modelling the environmental changes, and hence the stress levels in a painting, by stochastic
processes.

Damage accumulation models can also be used to model macroscopic damage. The damage
variable could then represent the depth of a crack, the degree of decohesion between two layers,
or the density of a crack pattern.
Bibliography


Appendix A

Models used for the test runs

Model 1: Cooling down a canvas painting

- 3 layers:
  - canvas, 0.0152 cm
  - glue, 0.00508 cm
  - Naples yellow oil paint, 0.0152 cm

- 76 x 102 cm

- elements 1-100: canvas
  elastic modulus 38.6 MPa
  thermal coefficient 0.1 \cdot 10^{-4} per °C
  poisson ratio 0.3

- elements 101-200: glue
  elastic modulus
  
  - at 23°C 37.91 \cdot 10^{2} MPa
  - at -3°C 48.60 \cdot 10^{2} MPa
  thermal coefficient 0.25 \cdot 10^{-4} per °C
  poisson ratio 0.3

- elements 201-300: paint
  elastic modulus
  
  - at 23°C 3.27 \cdot 10^{2} MPa
  - at -3°C 10.34 \cdot 10^{2} MPa
  thermal coefficient 0.52 \cdot 10^{-4} per °C
  poisson ratio 0.3

- element type: 8-noded, three-dimensional brick element with trilinear interpolation function:
  \[ T = \alpha_1 + \alpha_2 x + \alpha_3 y + \alpha_4 z + \alpha_5 xy + \alpha_6 yz + \alpha_7 xz + \alpha_8 xyz \]

- boundary conditions: all nodes at the frame are fixed in the x, y and z direction

- initial temperature: 23°C
  nodal temperature: -3°C

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Model 2: Desiccating a canvas painting

- 3 layers:
  - canvas, 0.0152 cm
  - glue, 0.00508 cm
  - Naples yellow oil paint, 0.0152 cm

- 76 × 102 cm

- elements 1-100: canvas
  - elastic modulus 35. MPa
  - moisture coefficient 0.1 \cdot 10^{-3} per percent RH
  - poisson ratio 0.3

- elements 101-200: glue
  - at 70\% RH 25.1655 \cdot 10^{2} MPa
  - at 10\% RH 16.0635 \cdot 10^{2} MPa
  - moisture coefficient 0.264 \cdot 10^{-3} per percent RH
  - poisson ratio 0.3

- elements 201-300: paint
  - at 70\% RH 76.31 \cdot 10^{2} MPa
  - at 30\% RH 41.51 \cdot 10^{2} MPa
  - moisture coefficient 0.257 \cdot 10^{-4} per percent RH
  - poisson ratio 0.3

- element type: 8-noded, three-dimensional brick element with trilinear interpolation function:
  \[ T = \alpha_1 + \alpha_2 x + \alpha_3 y + \alpha_4 z + \alpha_5 xy + \alpha_6 yz + \alpha_7 xz + \alpha_8 xyz \]

- boundary conditions: all nodes at the frame are fixed in the x, y and z direction

- initial relative humidity: 70 RH
  nodal relative humidity: 30 RH
Appendix B

Results of the Finite Element Analysis

B.1 The effect of ionic bonds

The results given below are copied from the output files from MARC. Given are the stresses in the lower left corner (nodes 1, 5, 9 and 13, and elements 1, 101 and 201) and the stresses in the middle (nodes 133, 134, 309 and 426, and elements 45, 145 and 245).
### Polyacrylic Acid

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B.2 Stretching a canvas

The results given below are the stresses computed by MARC in one of the corner nodes (node 1) and in the node in the middle (node 63).
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stress

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B.3 A painting with a tear

The results given below are the stresses computed by MARC in the nodes near the tip of the tear. Node 15 is the node at the tip of the tear and the other 9 nodes are the ones that lay on a small circle around it (see Figure fig-newman-mesh2).
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MOLART – Molecular aspects of Ageing of painted Art – is a 5-year co-operative project between art historians, restorers, analytical chemists and technical physicists funded by the Netherlands Organisation for Scientific Research (NWO). The object of MOLART is the development of a scientific framework for the conservation of painted art on the molecular level.

Craquelure in paintings is a very complex phenomenon and its study brings together a large number of scientific disciplines, such as art history, conservation and restoration science, chemistry and physics. Within this report we gave an introduction into these different disciplines focussing on the aspects of these disciplines relevant to the research on craquelure and other mechanical damage in paintings.

Cracking or, more generally, fracture is induced by mechanical stresses in a material. Tensile stress causes deformation, provided that the material is not restrained. Environmental changes are an important cause of internal stress in the different paint layers. The mechanical properties of a material, e.g. the stiffness and the strength, change with temperature and relative humidity. Furthermore, the different layers will try to shrink upon desiccation and cooling. Because these layers are restrained, this produces a stress-rise in the painting.

The first stage of our project consisted of a literature study to obtain a more thorough understanding of craquelure in paintings and to explore the different types of research already done on this subject within the field of conservation science. During this study we found that research previously carried out on the effects of environmental changes covers a wide range of methods, which makes it difficult to compare the results.

A technical approach was used by Mecklenburg and Tumosa when trying to predict the stresses in a painting when subjected to changes in temperature and relative humidity using finite element analysis. Finite element analysis is a numerical computing technique for solving problems in the field of continuum mechanics. Their results served as a starting point for our research.

Chemical research done within the MOLART project indicated that during ageing the pigment molecules in oil paint will form ionic bonds with the crosslinked network of the medium. It was possible to do some simulations by modelling a fresh paint layer consisting of polyacrylic acid and an aged paint layer consisting of a zinc salt of polyacrylic acid. These simulations show that the stress in the zinc salt remains low, despite an increase in stiffness. This low stress is due to the fact that this material tends to expand much less under a temperature rise than was expected.

To further explore the possibilities of using finite element analysis (FEA) to compute stress distributions in paintings, simulations were done on a model of a painting with a tear in the canvas.
These simulations were inspired by the fact that the ‘Cathedra’, a painting by Barnett Newman, was cut several times. The simulations show a choice for a relatively stiff repair material creates the benefit of a lower chance of tearing of the primary cut.

During this research project it became clear that finite element analysis is a good method to understand the effects of changing mechanical properties of ageing artists materials on the stress development in paintings, provided that these mechanical properties are known.

Criteria from the field of fracture mechanics can be used to decide whether failure will occur when a structure is subjected to certain stress levels. However, even when the stress in a structure never reaches the critical level for fracture or plastic flow to occur, there still can be some resulting damage on a microscopic level. Especially, when subjected to a cyclic load, the microscopic damage resulting from each cycle can add up and finally result in fracture. This phenomenon is called fatigue and many of the factors related to fatigue are stochastic in nature, such initial defects and environmental conditions. The final stage of our project consisted of the study models that can compute the expected damage and the fatigue life prediction of a painting, when the environmental changes, or the stress levels, are modelled as stochastic processes.
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