

**Proceedings of SPIE—The International Society for Optical Engineering**

**Volume 398**

# **Industrial Applications of Laser Technology**

**William F. Fagan**  
*Chairman/Editor*

**April 19–22, 1983**  
**Geneva, Switzerland**

*Published by*  
**SPIE—The International Society for Optical Engineering**  
**P.O. Box 10, Bellingham, Washington 98227-0010 USA**  
**Telephone 206/676-3290 (Pacific Time) • Telex 46-7053**

**SPIE (The Society of Photo-Optical Instrumentation Engineers) is a nonprofit society dedicated to advancing engineering and scientific applications of optical, electro-optical, and photo-electronic instrumentation, systems, and technology.**

# Application of Denisyuk pulsed holography to material testing

R.L. van Renesse and J.W. Burgmeijer

Institute of Applied Physics TNO, P.O. Box 155, 2600 AD Delft,  
the Netherlands

## Abstract

When holography is applied outside the laboratory, some well known problems are experienced: vibrations, rigid body motion, stray daylight. Pulsed holography can overcome the difficulties with vibrations but the other problems are less easily solved. When the object area to be holographically tested is small, a very simple and convenient method may be employed, which was earlier described by Neumann and Penn<sup>6</sup>; they fixed the hologram holder rigidly on the object under test, thus avoiding rigid body motion of the object with respect to the hologram. In a similar configuration Denisyuk reflection holograms are made without any necessity of darkening the environment. The authors believe that the main reason that this technique is not widely used, is due to difficulties generally encountered in processing the Denisyuk hologram to good quality. A simple processing technique is described resulting in high quality reflection holograms which may be analysed by microscope up to interference fringe densities of about 30 fringes/mm. As examples the results of a projectile impact study and the study of early fatigue crack detection in a critical aeroplane structure will be presented.

## 1. Introduction

Surprisingly the history of wavefront reconstruction by recording standing waves in a volume-medium, appears to date back as far as 1810, when Goethe's extensive work on chromatics is published.<sup>1</sup> In this work we read the findings of T.J. Seebeck that silver-chloride under circumstances tends to adopt the color of the light it is exposed to. Along this principle the first color photographs were inadvertently made by E. Beckerel about 1848, who was unaware yet of the underlying principle.

After W. Zenker's explanation of the phenomena through the theory of standing light waves in 1868 it was the celebrated M.G. Lippmann who in 1891 succeeded in making brilliant color-photographs of excellent keeping quality that have stood the tooth of time and can still be found reconstructed in some musea.<sup>2</sup> It was soon recognized that color photography was to be developed along more practical lines and thus the fascinating technique of Lippmann photogfaphy became of only academic interest. It must be emphasized however that at the turn of the century the fabrication of simple volume reflection holograms was practically possible and was in fact accomplished by H.E. Ives in 1908<sup>3</sup> who actually made holographic optical elements using the green mercury line while applying a photographic bleach technique. In 1892 Lippmann had already demonstrated dichromated gelatin as an excellent medium for volume color photography.<sup>4</sup> The real potential of the technique, apart from color photography, was not realized however and practical applications had to wait until concrete form was given to Gabor's ideas (1948) on wavefront reconstruction. It was Yu.N. Denisyuk<sup>5</sup> who in 1962 revived the old Lippmann-process and demonstrated its suitability to wavefront reconstruction. While the simplicity of the optical principle has promoted its application to holographic interferometry, contrary to this the need for extremely fine-grained emulsions and the difficulties connected with their processing have restricted the general application of the technique in this field.

It is the purpose of this paper to discuss these difficulties and their convenient solution as well as the practical applicability of the technique to holographic interferometry.

## 2. An NDI-application

### 2.1 Crack-detection

An investigation presently carried out by our institute will serve as an introduction to the principles of the technique. We are developing a non-destructive inspection (NDI) method that will detect the initiation and growth of fatigue cracks at fastener holes in the wing of an aircraft. Using a periodical inspection procedure, fatigue crack growth has to be detected before the crack size has reached its critical value. The demand for reliable inspection methods comes from the need to fly some aircraft well beyond their initial design lifetime.

For the most current inspection methods, the detection of cracks under installed fasteners forms an only partly solved problem. The inspection procedure must be appropriate for use in the aircraft hangar without the need to dismantle the wing or dismantle fasteners. This last restriction is understood by taking into consideration that badly installing of fasteners is one of the principal causes of crack initiation in fastener holes.

One is most interested in the inspection of three fastener holes in a highly stressed area, the so called 'critical area', where a higher probability of fatigue crack initiation exists. This critical area has a flat, diffuse metal surface and is approximately circular with a 60 mm diameter. It is situated in the lower wing skin on a place not easily accessible for current NDI-apparature. Holographic techniques are known for the detection of fatigue crack growth<sup>6,7</sup>, but many of them have serious drawbacks for our application. For the detection of these flaws not only a suitable loading mechanism has to be found, but also some extra conditions inherent to the application outside the optical laboratory must be satisfied. We will now consider six principal items connected with the technique.

1. A loading method has to be found causing a detectable anomaly in the fringe pattern, which is a map of surface deformation, in the neighbourhood of holes where cracks have reached a length much smaller than the critical length. At the tip of the flaw a plastic deformation zone will be formed, which will reach the surface far earlier than the crack opening. Most NDI-methods are based on crack detection and not on detection of this plastic deformation zone. It is assumed that this zone will induce an anomaly in the deformation during loading. Several loading methods can be considered, for instance direct mechanical stressing, vibrational excitation, and impulse loading. For the latter two methods a double pulse laser would be necessary. In a preliminary experiment in a test facility for aircraft model constructions, we have demonstrated that a mechanical stressing method is successful. For an inspection procedure in a hangar however, this may be unpractical and therefore other loading methods are still searched for.

2. During hologram exposure vibrations and motions must not cause translations between object and holographic plate more than a fraction of the wavelength. The method applied by most holographers is to shorten the exposure time, such that translations during this time-interval are restricted. To get sufficient exposure energy however a high power pulsed laser is needed. We have used a ruby laser having a pulse width of 25 ns and a pulse energy of 30 mJ.

3. Rigid body motions of the object relative to the holographic plate between the two exposures of the hologram will give an unwanted extra number of fringes during reconstruction. There are many solutions for this problem, which all have their own drawbacks. These fall into 3 categories:

- a. Using a double pulse laser, the pulse interval can be as short as 1  $\mu$ s. The object translation in this interval will be small enough to prevent most of the formation of unwanted fringes. A drawback then is the restriction to dynamical loading techniques, for instance vibration excitation and impulse loading.
- b. A compensation after the exposure of the interferogram is possible, e.g. using fringe control, sandwich-holography, holographic-moiré. At the present time these techniques are still too sophisticated for a routine-based inspection by untrained personnel.
- c. During the exposure of the hologram, rigid body motions can be compensated for by a technique introduced by Neumann and Penn.<sup>8</sup> They clamped the holographic plate holder to the object with the holographic plate as near to the object as possible, and used a collimated laserbeam as to optimally compensate for rigid body motion. (Figure 1). The holographic technique applied is Denisjuk reflection holography.<sup>5</sup> Although Denisjuk reflection holography has the advantage of simplicity, the impossibility to adjust the intensity-ratio between object and reference beam limits its applications. Generally objects holographed this way must possess a high reflectance or otherwise be coated with a suitable metallic paint, to render a sufficient intensity-ratio.<sup>8</sup>

We have used the Denisjuk technique without any surface preparation of the object. Therefore it is demanded that the holographic processing performs optimally with respect to diffraction efficiency and signal-to-noise ratio. This subject will be discussed in section 3. A drawback of the technique may be that the size of the inspected area is restricted to the size of the holographic plate. For our application however the size of the area to be inspected was small enough to be covered by a 4x5" plate. The technique offers a very efficient compensation for machine vibrations and motions. Some small vibrations between object and plate cannot be compensated for, so that the use of a pulsed laser is still needed for their complete elimination.

4. A surplus of daylight exposure on the holographic plate will seriously decrease the contrast of the interference fringes, and along with it the brightness of the reconstructed image. The technique of Denisjuk pulsed holography however offers a good opportunity to expose holograms in a daylight environment.

A plateholder was constructed which is shielded on one side by the object and on the other side by a mechanical shutter with a large aperture. During the opening of the shutter (1 s) the pulsed laser is fired and the exposure to daylight will generally be negligible.

5. Simplicity of the set-up and its operation is demanded in case of operation by unskilled personnel. The need for a simple set-up is the more urgent since the location of the area to be inspected is not easily accessible. As is illustrated by figure 1 the Denisyuk holographic set-up is very simple and easy to adjust. The plateholder can be manufactured compact so that it can be conveniently installed at all kinds of locations.

6. Interpretation of the fringe pattern will be most easy if it can remain qualitative. In that case, the abrupt changes in the curvature of the fringes that appear near the plastic deformation zone, allow the qualitative discovery of a crack before it can be detected by the usual means. If the anomaly in the fringe pattern is only slight, or if the crack-size has to be evaluated quantitatively, automatic fringe readout and processing becomes necessary.

## 2.2 The effect of shrinkage

Due to the processing of the hologram the emulsion will generally shrink, which affects reconstruction angle and wavelength. This section evaluates these reconstruction parameters while the causes of this shrinkage will be discussed in section 3.

The holographic plate is exposed by a coherent plane wave of wavelength  $\lambda_e$  under an angle  $\alpha_e$  with the normal (Figure 2). Part of this wave is transmitted through the emulsion and subsequently diffusely reflected by the object. Each object point will radiate a nearly spherical wave towards the plate, which will interfere with the original plane wave. The period of the resulting interference pattern is nearly  $\lambda_e/2n$  in a medium of refractive index  $n$ . If the angle  $\alpha_e$  is negligible, the interference pattern consists of planes practically parallel to the surface of the holographic plate. In practice a small angle  $\alpha_e$  is adjusted as to separate the zero-order from the reconstructed image. After processing the fringe period is given by:

$$d = s \frac{\lambda_e}{2n}, \quad (1)$$

where  $s$  denotes the shrinkage factor.

For the reconstruction of the hologram a wavelength  $\lambda_r$  is used, which is not necessarily equal to  $\lambda_e$ . According to Bragg's law constructive interference of the light reflected by successive planes is obtained by:

$$2nd \cos \alpha_r = \lambda_r \quad (2)$$

Equations (1) and (2) may be considered in two different ways:

1. If the reconstruction is made under the same angle as the exposure ( $\alpha_r = \alpha_e \approx 0$ ), an optimally bright reconstruction is obtained by selecting the proper wavelength  $\lambda_r$  of the reconstruction light source.

From eq.(1) and (2) then follows:

$$\lambda_r = s \lambda_e \quad (3)$$

Due to the shrinkage of the emulsion the reconstructed wavelength is shifted by a factor  $s$  with respect to the original wavelength  $\lambda_e$ . Thus the shrinkage factor  $s$  may be experimentally determined by measuring  $\lambda_r$ . The results of these experiments for different processing parameters will be discussed in section 3.

2. For optimal resolution of the image details in the reconstruction a monochromatic light source is needed. The application of a He-Ne laser for this purpose is most convenient. The reconstructed wavelength  $\lambda_r$  is then fixed so that we have to optimize the angle  $\alpha_r$ . It follows again from eq.(1) and (2) that:

$$\cos \alpha_r = \frac{\lambda_r}{s \lambda_e} \quad (4)$$

Next to the ruby laser ( $\lambda_e = 694 \text{ nm}$ ), we use a He-Ne laser ( $\lambda_r = 633 \text{ nm}$ ) for reconstruction of the holograms, so that according to eq. (3) we find  $\lambda_r/\lambda_e = 0.91$ . Therefore, if the shrinkage is more than 9% ( $s < 0.91$ ) there is no solution for eq. (4) and reconstruction becomes impossible unless a laser of shorter wavelength is applied. Without shrinkage of the emulsion ( $s = 1$ ) the angle of reconstruction  $\alpha_r = 24^\circ$ . Due to refraction this angle will become approximately  $37^\circ$  outside the hologram.

Generally, in a conventional set-up for transmission holograms the object can only be observed under limited angles, practically normal to the object surface. This is due to the rather large object to plate distance with respect to plate size. As a result the sensitivity of a suchlike set-up for object deformations is practically normal to the object surface so that mainly out-of-plane deformations are detected. Contrary to this the Denisyuk holographic set-up allows minimal object to plate distances, so that the object may be observed under rather large angles with the normal. Therefore sensitivity to in-plane deformations is obtained as well.

Considering the foregoing, it appears necessary to find a holographic process performing well enough to allow the making of Denisyuk holograms without surface preparation of the object. Moreover, this process should allow sufficient adjustment of the shrinkage factor  $s$ . In the next section the relevant processing parameters will be discussed.

### 3. Direct bleach

#### 3.1 Efficiency

The conversion of the developed image silver of an amplitude transmission hologram into a transparent silver compound image, after removal of the residual halide by the fixer, creates a phase hologram which diffraction efficiency is considerably increased. A suchlike conversion, which is generally referred to as "direct bleach", results in a silver halide, a silver ferrocyanide, or a silver mercuriochloride image. For a long time there has been a deeply rooted belief of many holographers that the conversion of the image silver into a compound with a refractive index substantially greater than that of the gelatin medium was demanded in order to prepare efficient holograms. The cause of the original misunderstanding that the refractive index modulation should increase along with increasing refractive index of the silver compound grains, is probably attributable to the implicit supposition that we deal with grains with large respect to the wavelength of the light in the medium. In that case the geometrical-optics formulation gives the resulting refractive index as the mean, weighted by volume, of the refractive indices of the respective components. However this supposition is obviously invalid in the case of the extremely fine-grained holographic emulsions where the grain diameter amounts a few tens of nanometers only. Such grains may be regarded as Rayleigh scatterers and the refractive index of the composition is given by the Lorentz-Lorenz equation, which predicts an approximate linear relation between refractive index of the composition and the polarizability of the composite substances. Efficient phase transmission holograms therefore may be obtained independent of the bleaching agent applied, as long as differences in concentrations of polarizable silver compound molecules are optimized by adjusting the exposure energy. This explains why in the past decades innumerable chemical formulations have been proposed, a research-effort reminiscent of the alchemic quest of the philosophers' stone, not resulting however in anything like a "final" formulation.

In earlier work<sup>9</sup> it was set forth that not the refractive index, but the molecular polarizability and the molecular volume of the silver compound concerned are the crucial parameters governing the phase variations resulting from the variations in prebleach optical density. The -at the first sight paradoxal- conclusion that follows from this proposition is, that efficient phase transmission holograms may be obtained by conversion of the image silver into a silver compound of refractive index equal to that of the gelatin. This was experimentally confirmed by conversion of the image silver into silver ferrocyanide which refractive index virtually equals that of the gelatin.

In view of photographic sensitivity the only and minor consideration that follows from the foregoing is that we might wish to apply the bleaching agent that converts the least possible density variations into sufficient phase variations. Evidently this is accomplished by the conversion of the image silver into the compound with the highest possible polarizability, silver mercuriochloride.

It must be emphasized that the foregoing specifically applies to transmission holograms where, beyond spatial frequencies of some hundreds of lines/mm, the gelatin surface will no longer show thickness variations in accord with variations in concentration of the silver or its compounds (ref. 9 section 4). The contrary is true for reflection type volume holograms, where Bragg-planes are oriented perpendicularly with respect to transmission type holograms.

In this latter case, even at very high spatial frequencies the gelatin is able to follow such variations in concentration to a large extent, therefore the foregoing considerations only apply to transmission holography.

#### 3.2 Scatter

The one-sidedness of the previous considerations becomes evident once the scattering

of the silver compound grains is taken into account. A bleaching agent that, for the smallest possible optical density modulation, renders optimal diffraction efficiency, may well result in an objectionably high noise-level. As it appears, the intensity of the light scattered by a particle, small with respect to the wavelength, is proportional to the square of its total polarizability.<sup>10</sup> This observation shows already that diffraction efficiency is indissolubly linked to light scatter. The situation becomes more complex however because the grain may in general not be regarded as a scatterer in a homogeneous gelatin medium. On the one hand the gelatin medium may be tanned through cross-linking of the gelatin molecules in the vicinity of the grain by the oxidation products of the developer, on the other hand the gelatin may be locally compressed through the forces exerted by the grain which increases in size when being bleached. These processes may change the mass density of the gelatin near the grain, thereby causing a change in the refractive index of the gelatin enveloping the grain. These considerations led to the proposition of the model of a scatterer as depicted schematically in figure 3.

The spherical core of radius  $f \times r$  consists of silver compound of refractive index  $n_1$  and is enveloped by a spherical gelatin shell of refractive index  $n_2$ ; thus composing a scatterer of radius  $r$ , immersed in the regular gelatin medium of refractive index  $n_3$ .<sup>11</sup> The total polarizability  $\alpha$  of such a composite particle, small with respect to the wavelength, is given by Van de Hulst.<sup>12</sup>

The validity of the shell-model was confirmed experimentally by scattering measurements as well as phase measurements, the results of both approaches being in close agreement.<sup>11</sup> It is a remarkable coincidence that the proposition of this shell-model was accompanied by complementary experimental evidence collected by Joly e.a.<sup>13</sup>, who applied developers of different tanning activity and showed by means of electron photographs that the average size of the bleached grains is independent of the type of developer. Joly concluded therefore that the observed differences in noise-level must be attributed to gelatin distortions within as well as at the surface of the emulsion. As mentioned before the growth of the grains may be expected to exert radial forces on the adjacent gelatin, forces that may compress the shell of tanned gelatin, thus increasing its molecular density and with it the total polarizability of the scatterer. Alternatively however these forces may indeed further enhance the amplitude of the surface irregularities coincident with the surface grains. A simple comparative experiment with a developer of low tanning activity reveals indeed that it is not the collapse of the gelatin layer that accompanies the fixing step, but the bleaching that causes excess "reticulation" of the gelatin surface. Joly has shown that surface irregularities decrease with increasing tanning activity of the developer. From the foregoing it follows that a suchlike suppression of the surface scatter goes at the expense of an increased intra-emulsion scatter, since the decrease in surface irregularities is compensated by increased molecular density in the shells. Therefore, depending on the nature of processing the balance between intra-emulsion scatter and surface scatter may deflect between extremes. These observations sustain our conclusion that the intensity of scattered radiation may vary widely with the type of processing applied and that it is therefore not adequate to relate results of various bleaching agents without investigating the influence of processing parameters on these separate contributions to the total emulsion scatter.

This may be accomplished by utilizing an index-matching technique. Evidently, application of the direct bleach process will only yield optimal results if the total polarizability of the scatterers is minimized through a subtle processing that prohibits shell forming by allowing the emulsion to swell completely, and if the surface is index-matched as to eliminate surface scatter.

### 3.3 Gelatin collaps

The intricacy of the direct bleach process is further increased by the fact that the removal of the bulk of residual silver halide by the fixer causes the gelatin layer to collapse. For volume reflection holograms this involves a decrease in distance between Bragg planes, resulting in a generally unacceptable shift to shorter reconstructed wavelengths, an inconvenience that demands the application of a swelling agent like triethanolamine.<sup>14</sup> This is illustrated by a series of experiments with holographic volume gratings exposed on Agfa Gevaert 8E75 HD emulsions at 633 nm wavelength. Two plane waves of about equal intensity impinge perpendicularly on either side of the plate thus forming an interference pattern of period  $\lambda_c/2n$  parallel to the surface. The holograms were developed in pyrogallol developer of the following composition:

A pyrogallol	20 g	B sodium carbonate sicc.	60 g
sodium sulfite sicc.	nihil	distilled water to	1 l
distilled water to	1 l		

The solutions A and B are mixed in equal proportions immediately prior to development. As the mixture is liable to fast oxidation it should only be used once. Without sodium sulfite solution A has a limited shelf life.



Holograms are developed for 4 min. (20°C-cont.agit.), washed for 2 min. in running water, subsequently fixed in a 200 g/l sodium thiosulfate neutral fixer (10 min.- interm.agit), washed 10 min. in running water and finally bleached in formulations 2 or 4 as to convert the image silver into AgBr or Ag<sub>4</sub>Fe(CN)<sub>6</sub> respectively.<sup>9</sup> By means of a spectrometer the reconstructed wavelength  $\lambda_r$  was measured at each stage of the processing, as a function of optical density. The results of these experiments are given in the figures 4 and 5. Immediately after development a decrease in reconstructed wavelength nearly proportional to optical density is established. This is due to the first gelatin collapse after removal of bromine through reduction of AgBr by the action of the developer. As will be shown in the next section, the hardening activity of the pyrogallol developer has a limiting effect on the gelatin collapse. Subsequent removal of the residual AgBr by the neutral fixer drastically converses this picture. As the most of the bulk halide is removed at low optical density, the gelatin collapse is the most severe in this region as is indicated by the reverse in slope. A total shift in reconstructed wavelength of about 100 nm is encountered for optical densities between 2 and 3. The final conversion of the image silver into a transparent silver compound results in a considerable growth of the grains, in accordance with the increase in molecular volume, and consequently causes the gelatin layer to swell again. As the molecular volume of Ag<sub>4</sub>Fe(CN)<sub>6</sub> exceeds that of AgBr by a factor of 2.4, the slope of the graph is expectedly higher in the former case.<sup>9</sup>

The large amount of alternate shrinking and swelling of the gelatin appears to be disadvantageous since it is only at the bleaching stage of the processing that serious surface scatter is induced. A considerable band-broadening is observed as well (up to 0 nm), indicating that with these large variations in gelatin thickness and grain size the smoothness and coherence of the Bragg-planes diminishes at the expense of diffraction efficiency (max. observed 10%).

Considering the difficulties apparently inherent to the rather circumstantial developer-solvent-rehalogenate procedure, an alternative processing that avoids these problems should be very useful. A more direct approach to be considered therefore is the developer-silver solvent process that modulates the concentration of the residual silver bromide.

#### 4. Reversal bleach

##### 4.1 General

If after development of an amplitude hologram, omitting the fixing step, a suitable silver solvent is applied that does not significantly remove any of the residual silver-halide, a phase hologram is obtained as well. In this case, which is generally referred to as "reversal bleach", the concentration of the silver bromide that was originally deposited in the emulsion, is modulated. This reversal process owns a few advantages over the direct process:

- The omission of the fixing-step reduces the process to essentially two steps, which considerably curtails processing time.
  - The residual AgBr grains have not suffered any changes in size, neither has the gelatin adjacent to the grains undergone any differential tanning. Therefore these grains are not enveloped by any shell-structure, neither have they been the cause of any surface irregularities, and thus may be expected to render the least scattering.
- Conversely it has been argued by Phillips<sup>15</sup>, that removing irregular chunks of silver will leave an irregular vacancy in the gelatin host, and the scatter may thus be as severe as if the image were fixed and rehalogenated. For the same reason however one might argue that the fixing step, which removes the AgBr grains, must be expected to bring about a suchlike effect, which as experiments show, does not appear. If indeed the silver grain removal would leave marked voids in the bulk gelatin, a considerable background scatter should be observed at higher prebleach densities. Our experiments however have shown invariably that the reverse is the case: scatter decreases with prebleach density and becomes negligible at densities beyond 2 or 3, in the case of reversal bleach. Evidently, the bulk of residual halide, which is inversely proportional with optical density, is the main source of scatter. As will be shown in section 3.2 the application of a strong hardening developer prohibits further shrinking of the gelatin layer on removal of the silver grains. The creation of vacuoles in this case seems inevitable therefore, so that on second thoughts the observed decrease in scatter with optical density is somewhat surprising. Evidently the concept of bubble-like voids remaining in the gelatin after removal of the silver grain seems erroneous. It is difficult however to imagine what alternatively may take place in the emulsion, as collapse of the voids seems excluded by the absence of any wavelength shift.

##### 4.2 Experimental

We have conducted a series of experiments, similar to those described in section 2.3, with the reversal bleach technique to the purpose of disclosing the influence of processing parameters on the wavelength shift.

The straight pyrogallol developer was selected again because of its strong hardening activity which was expected to minimize gelatin collaps and thus to minimize wavelength shift. This is illustrated by a series of comparative experiments with a metol developer, the formulations of which is obtained by substituting metol for pyrogallol in solution A (section 2.3) and adding 100 g/l sodium sulfite.

Metol as a developing agent has a low tanning activity, while the high sulfite content further eliminates any remaining hardening activity by reducing the primary oxidation products of the developing agent so that these can no longer crosslink the gelatin molecules. The result of these comparative experiments is illustrated in figure 6. After development in the pyrogallol developer a shift in reconstructed wavelength is observed, virtually equal to that of the previous experiments, due to the liberation of bromine. Subsequent dissolution of the image silver by a 4 g/l potassium dichromate bleach with low pH (4 cc  $H_2SO_4$ /l) surprisingly does not induce any further collaps of the gelatin, as was already mentioned in section 3.1. This is probably due to the fact that the hardening activity of the developers' oxidation products has a limited diffusion lifetime so that the gelatin is cross-linked in the direct vicinity of the developed grains. Gelatin collaps by removal of these grains by the silver solvent is therefore prohibited by the rigidity of the locally hardened gelatin. Completely contrary to this, the removal of the undeveloped residual AgBr, consequently contained by unhardened gelatin, will cause a strong collaps of the gelatin layer as was demonstrated in the figures 4 and 5. The results with the non-hardening metol-sulfite developer shown in figure 6, sustain this explanation. Not only does the virtual absence of any hardening effect allow a considerably larger gelatin collaps during development, subsequent dissolution of the silver by the dichromatic bleach now brings about an expected further gelatin collaps. It may be noted that the largest shrinkage is rendered by removal of the bromine that has an atomic volume almost twice as large as that of silver.

These experiments show that phase holograms may be obtained by means of a simple reversal bleach procedure without the large and disastrous shrinking and swelling movements of the gelatin that occur during the cumbersome direct-bleach procedure. Shrinking occurs though, but is limited to a few tens of nanometers by the hardening activity of the pyrogallol. These experiments further suggest that the amount of the wavelength shift may be manipulated to a large extent by adjusting the hardening activity of the developer formulations by the sulfite content.

Figure 7 shows the results of a series of experiments demonstrating this possibility of reconstructed wavelength manipulation by increasing the sulfite content from nihil to 10% by weight. The wavelength shift increases linearly with about 10 nm/% sulfite up to a 3% concentration, beyond which the effect gradually saturates. At a concentration of 10% the shrinkage appears to have reached maximum values as no more difference between a pyrogallol-sulfite and the metol-sulfite developer can be observed. For prebleach optical density between 2 and 3.5 the color of He-Ne exposed holograms may thus be varied between a deep orange-red and a neutral-green.

Apart from the apparent advantages of the reversal bleach procedure combined with a tanning developer over the direct bleach procedure, the resulting diffraction efficiency of the holographic Bragg-gratings thus obtained, is of great interest. Diffraction efficiencies were measured from the spectrographs and are presented in figure 8. Optimal efficiencies typically amount 20% and are obtained for prebleach optical densities between 2.5 and 3.5. Bandwidths typically amount 20 nm.

Because the hardening pyrogallol development is accompanied by the formation of a considerable brownish stain of the gelatin, the transparency of the hologram suffers a decrease proportional to prebleach density. Evidently the resulting absorption will cause a decrease in diffraction efficiency; in view of the simplicity of the related processing however, this disadvantage seems of little interest for NDI-holography. It may be mentioned however that this gelatin stain can be removed for instance by a 1-1½ min. bath in a 0.05% potassium permanganate/0.4% sulfonic acid solution in water. As it appears that gelatin stain and gelatin hardening are independent to a high degree<sup>16</sup>, the resulting gelatin collaps is only slight (10-15 nm). This stain removal however is accompanied by an increased background scatter, which is to be expected because this stain effectively absorbs multiple scattering by the halide grains. A further processing step, which consists of dyeing the gelatin with a suitable dye, is therefore demanded to suppress this scatter while preserving the gain in diffraction efficiency, which is increased to typically 30%.

## 5. Results

The excellent performance of the reversal bleach processing described, rendered the possibility to make Denisjuk holograms of low reflectance diffuse objects. Although the diffraction efficiency considerably decreases with the reflectance of the object, reconstructions remained sufficiently bright to allow for their convenient evaluation. As an example a few results are presented of our investigation of the fatigue crack growth



discussed in section 2.1. The area of interest has a diffuse reflectance of only 10% for the ruby wavelength of 694 nm. The holograms were made in an environment with machine vibrations and stray daylight. Figure 9 shows the double-exposure interferograms of the critical area for an increasing number of stress cycles. The interference pattern given in figure 9b significantly indicates the development of a crack well before it could be visually detected.

An important advantage of the Denisyuk technique moreover is that the image is reconstructed in the immediate vicinity of the plate, so that it is generally accessible for low magnification microscopes. Extremely fine details of the interference pattern may therefore microscopically be evaluated without difficulty. This advantage was utilized in a recent study of impact phenomena.<sup>17</sup> Figure 10a gives an example of an interferogram of a ceramic target 1  $\mu$ s after impact. Figure 10b gives an enlarged portion of this interferogram showing the development of cracks after impact. Microscopic observation of such interferograms showed fringe densities up to 30 lines per mm. Fringe visibility in these areas was still sufficient to allow evaluation of the interferograms.

## 6. Discussion

A simple processing has been developed that allows the making of low-noise, high-efficiency Denisyuk phase reflection holograms. Even of objects of relatively low diffuse reflectance, holograms are obtained of a quality sufficient for their convenient interferometric evaluation. Further advantages of the processing described are the low emulsion shrinkage as well as the possibility of shrinkage manipulation, and so, reconstructed-color manipulation, through the sulfite content of the developer. Application of this processing allows the practical and easy utilization of Denisyuk holography outside the laboratory. The latter technique readily solves the problems connected with rigid body motion if the plate holder is fixed to the object.<sup>8</sup> Moreover the reconstruction of the object in the immediate vicinity of the plate allows microscopic evaluation of the fringe pattern up to high spatial frequencies. Finally the object may be observed under widely different angles so that sensitivity to in-plane as well as out-of-plane deformation is obtained.

## 7. Acknowledgement

The authors wish to thank the National Aerospace Laboratory in Emmeloord for their collaboration in the experiments on aircraft wing specimen. The research work, described in this paper, has been supported by the National Defence Research Organization in the Netherlands.

## 8. References

1. Goethe, J.W., *Naturwissenschaftliche Schriften*, Bnd. V, 156-159, Ausg. R. Steiner Verlag, Dornach (1982).
2. Lippmann, M.G., *La photographie des couleurs*, *Compt.Rend.* 112, 274-277 (1891).
3. Ives, H.E., An experimental study of the Lippmann color photograph, *Astrophys. J.* 27, 325-352 (1908).
4. Valenta, E., *Die Photographie in natürlichen Farben*, *Encyclopädie der Photographie*(2), W. Knapp (ed.), Halle a. S. (1894).
5. Denisyuk, Yu.N., On the reproduction of the optical properties of an object by the wavefield of its scattered radiation, *Opt. and Spectr.* 15, 279-284 (1963).
6. Erf, R.K. (ed.), *Holographic Nondestructive Testing*, Ac.Press, New York (1974).
7. Vest, C.M., *Holographic Interferometry*, John Wiley & Sons, New York (1979).
8. Neumann, D.B. and Penn, R.C., Off-table Holography, *Exp. Mech.* 15, 241-244 (1975).
9. Renesse, R.L. van, and Bouts, F.A.J., Efficiency of Bleaching Agents for Holography, *Optik* 38, 156-168 (1973).
10. Kerker, M., *The scattering of light*, p. 39f., Academic Press, New York/London (1969).
11. Renesse, R.L. van, Scattering properties of fine-grained bleached emulsions, *Phot.Sci. Eng.*, 24, 2, 114-119 (1980).
12. Hulst, H.C. van den (ed.), *Light scattering by small particles*, p73f., John Wiley & Sons, New York (1957).
13. Joly, L. Vanhorebeek, R., Development effects in white-light holography, *Phot.Sci.Eng.* 24, 2, 108-113 (1980).
14. Rowley, D.M. A holographic interference camera, *J.Phys.E: Sci.Instrum.* 12, 971-975 (1979).
15. Phillips, N.J., Ward, A.A., Cullen, R., Porter D., *Advances in holographic bleaches*, *Phot.Sc. and Eng.*, 24, 2, 120-124 (1980).
16. Tull, A.G. Tanning development and its application to dye transfer images, *J.Photogr. Sci.*, 11, 1-26 (1963).

17. Kolkert, W.J., Amelsfort, R.J.M., Renesse, R.L. van, On the early response of a sintered  $n\text{-Al}_2\text{O}_3$  ceramic target plate impacted by a low velocity lead projectile, 6th int. symp. on ballistics, 27-29 Oct. 1981, Orlando, Florida.

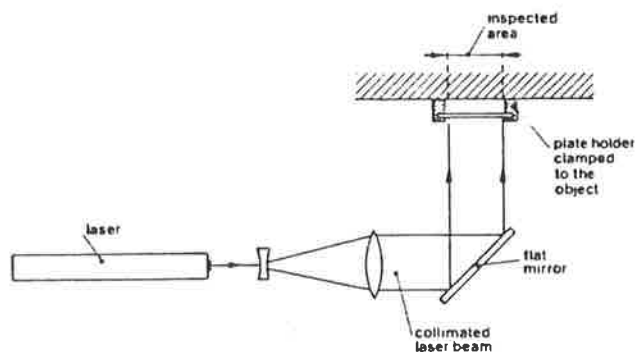


figure 1 arrangement for the exposure of reflection holograms

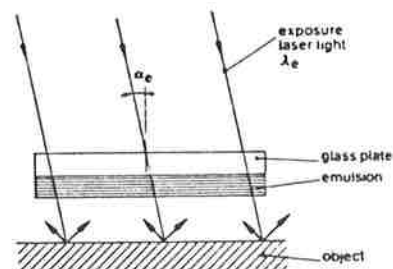


figure 2 formation of interference planes during exposure

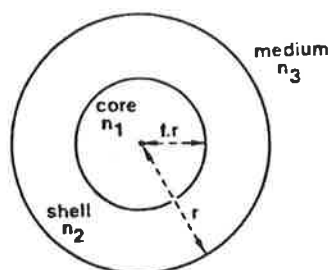


figure 3 model of a silver-compound scatterer enveloped by a tanned gelatin shell in a non-tanned gelatin medium

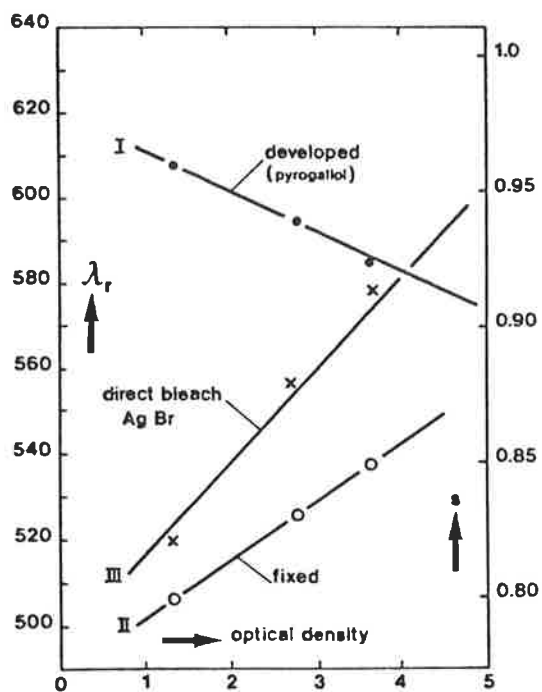


figure 4 direct bleach into AgBr

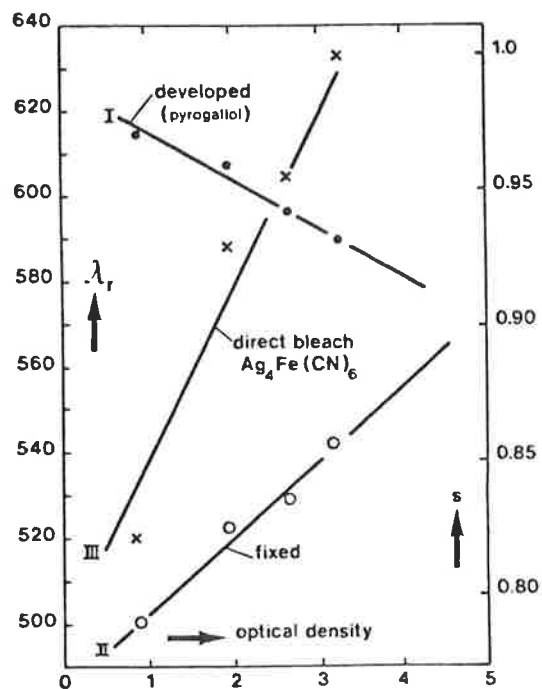


figure 5 direct bleach into  $\text{Ag}_4\text{Fe}(\text{CN})_6$

Reconstructed wavelength  $\lambda_r$  and shrinkage factor  $s$  as a function of optical density.

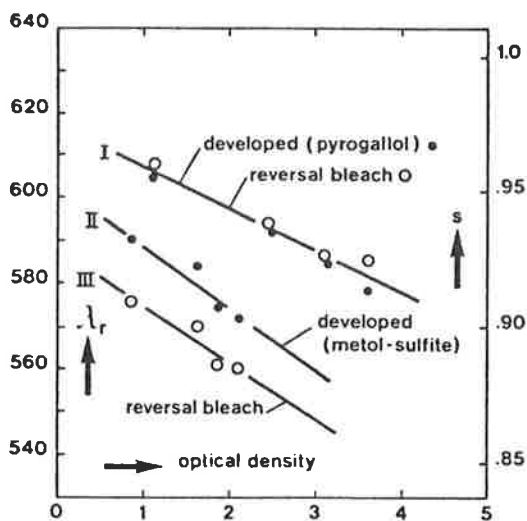


figure 6 comparison of a non-hardening metol developer and a hardening pyrogallol developer

reversal bleach: reconstruction wavelength  $\lambda_r$  and shrinkage factor  $s$  as a function of optical density.

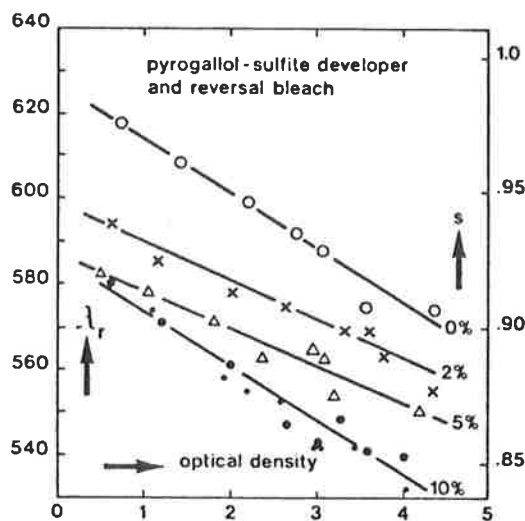


figure 7 influence of sulfite content  
 • pyrogallol - 10% sulfite  
 • metol - 10% sulfite

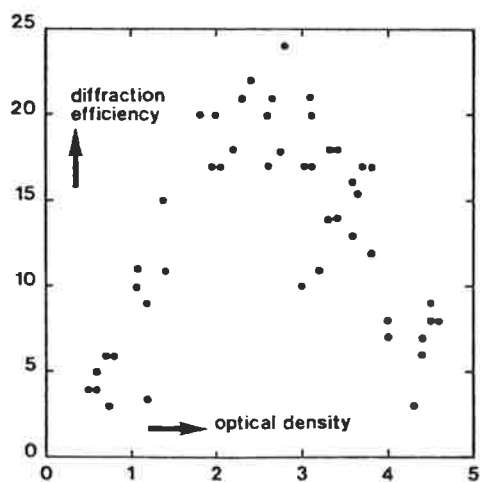
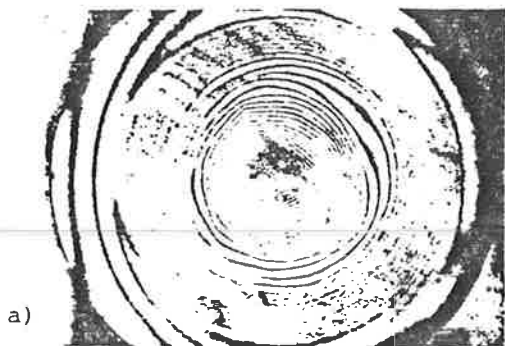


figure 8 reversal bleach: diffraction efficiency (%) as a function of optical density.



a)



b)

figure 10 a) interferogram of ceramic target 1  $\mu$ s after impact (1.5 x enlarged)  
 b) detail of a), showing crack development (13 x enlarged)

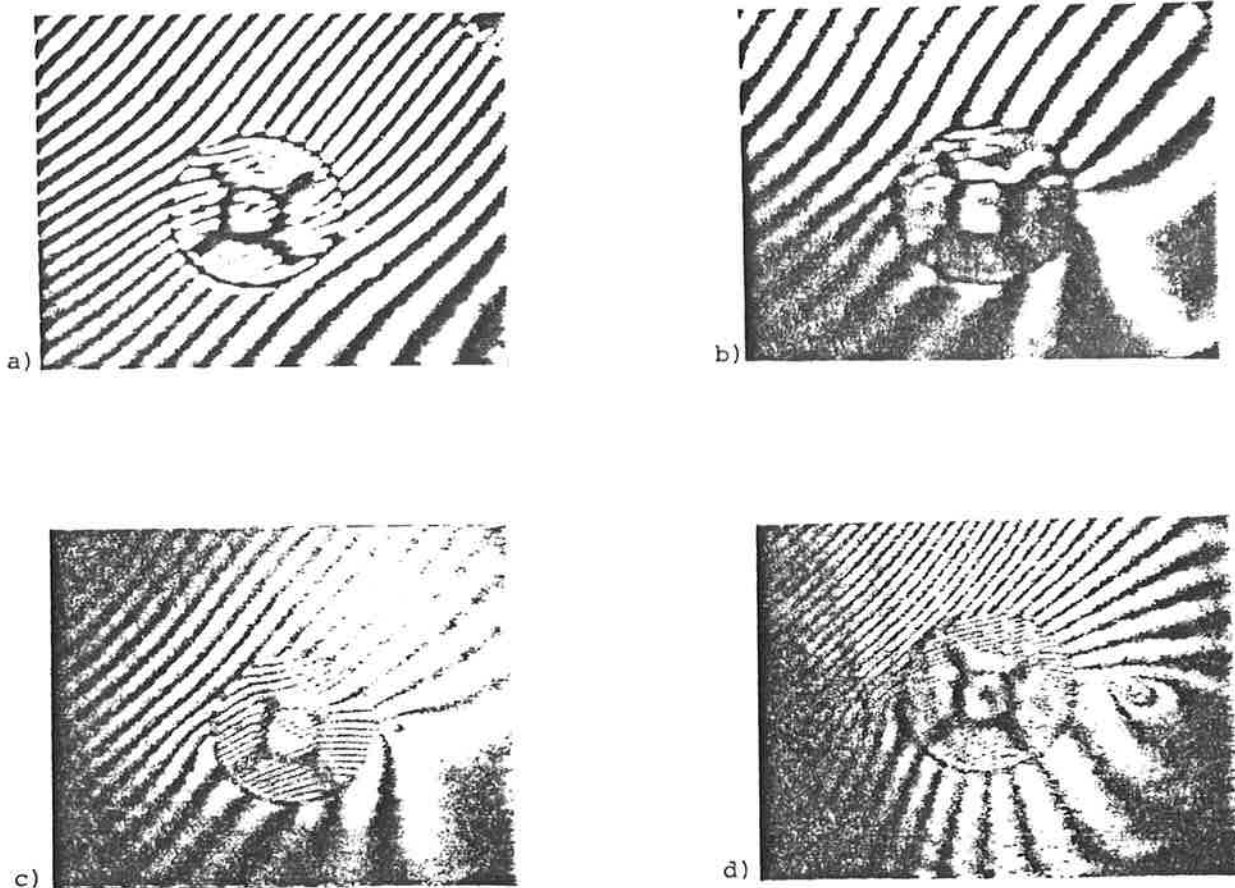


figure 9 Double exposure interferograms of the critical fastener in a specimen of the wing. The mechanical load on the specimen is increased between the two exposures.

The interferograms are made after an increasing number of stress cycles:

- a) no anomaly in the pattern,
- b) indication of the development of a crack on the right side of the fastener,
- c) the crack on the right side reaches the surface; crack development on the left side is also indicated in the pattern,
- d) a few cycles before complete fracture of the specimen.