NATIONAL BUREAU OF STANDARDS REPORT

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by

Sidney H. Greenfeld

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U. S. DEPARTMENT OF COMMERCE
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Jointly Sponsored by

Asphalt Roofing Industry Bureau

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1. INTRODUCTION

The ability to see within an asphalt or an asphalt product has always been hampered by the general opacity of the materials involved. The optical microscope has been used on very thin sections of asphalts, but its contributions have been limited primarily to surface configurations [1, 2][1/] and phase separations [3]. The election microscope has been applied to asphalt problems, but it has been shown that the observations reported were largely of artifacts introduced by specimen preparation [4, 5, 6].

X-rays have been used on asphalts in a number of types of analyses. While normal radiology has produced little useful information because of the transparency of asphalts to the wavelengths of x-rays normally used, x-ray diffraction and x-ray scattering studies have produced some useful information [7, 8]. However, the patterns have been quite diffuse and the interpretations have been questioned.

In recent years a point-projection microscope has been developed, which can be used with soft x-rays to "see" the interior of asphalt specimens as thick as 50 mils. Preliminary results of the application of this technique to asphalt problems will be covered in this report. This study was made in close collaboration with Dr. S. B. Newman, who is in charge of the microscopy laboratory at the National Bureau of Standards.

2. APPARATUS

The point-projection x-ray microscope, shown schematically in Figure 1, was of the Cosslett and Nixon type [9], manufactured by Micro X-Ray Laboratories, Ltd., of Surrey, London, England. It consisted of a Wehnelt type election gun with grid bias, electromagnetic condenser and objective lenses and a series of interchangable targets. The spot size on the targets was about one micron; however, slight scattering of the x-rays resulted in an effective x-ray source slightly larger than this. All x-ray photographs were made with an aluminum target, yielding x-rays of 8.34 Å wavelength.

The election gun was evacuated by mechanical and oil diffusion pumps to a pressure of 10^{-4} mm. of mercury. A complete description of this apparatus may be found in reference [9].

[1/] Numbers in brackets refer to literature references at the end of this report.

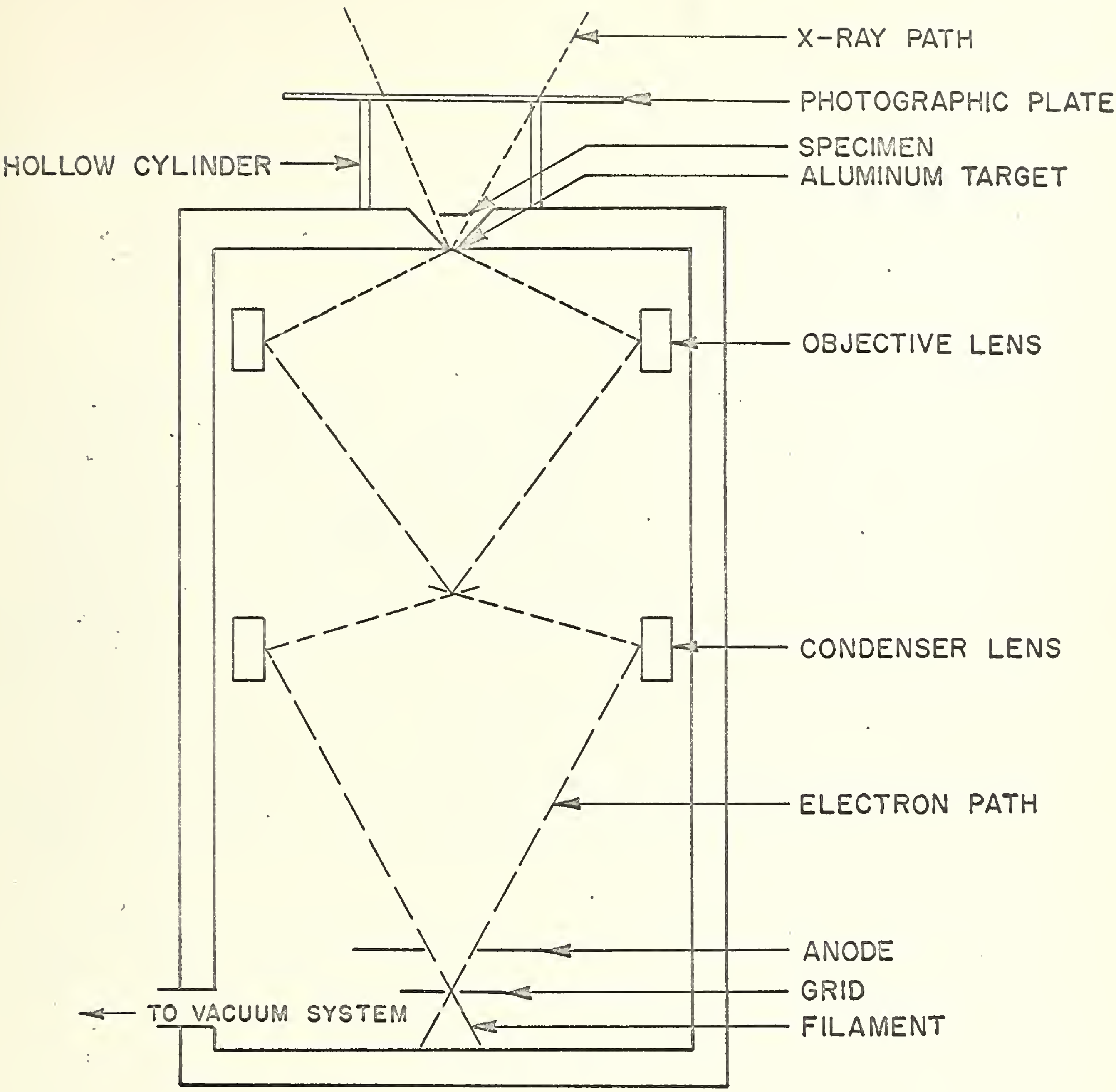

FIGURE 1. SCHEMATIC OF X-RAY PROJECTION MICROSCOPE

3. PROCEDURE

At the beginning of each day the election gun of the x-ray projection microscope must be evacuated and focused. The focusing is accomplished by placing a small piece of screen grid on the aluminum target and projecting its image on to a phosphor-coated sheet of lead glass. Adjustments are made by changing the magnetic fields in the lenses. When the microscope is ready the specimen is placed in position, a hollow cylinder of appropriate height to produce the desired magnification placed concentric with the specimen, a photographic plate placed on the cylinder and the exposure made. The plate is developed and viewed immediately. The gun was operated at 12.5 KV and 30 μA for all of the photographs in this report.

As seen in Figure 1, the focusing is entirely within the gun, all points in the x-ray path are always in focus when the gun is properly focused. Thus, the magnification is controlled by regulating the relative distances of the specimen and the plate from the x-ray source, the target of the election gun.. All points in the specimen are in focus, but points in different planes within the specimen are magnified to different degrees.

The photographs in this report are at various magnifications. No effort was made to position the specimen or control its thickness precisely. Thus, the results must be interpreted on a qualitative basis only.

4. RESULTS AND DISCUSSION

Figure 2 - Unexposed Asphalt

This photomicrograph is typical of many taken of commercial samples of coating-grade asphalt. There are indications of materials of different densities dispersed in the asphalt. These range from air bubbles, the white, round area, to various types of debris. Some of this litter may be introduced during specimen preparation, for dust and lint tend to collect on the freshly cut asphalt surfaces, but the bulk of the foreign material is in the asphalt.

FIGURE 2. A COMMERCIAL COATING-GRADE ASPHALT

Figure 3 - Cross Section of Asphalt Film Weathered for 88 Months on the Roof of the Industrial Building - Magnification 120X

In all weathered films, a more-or-less stratified appearance has been found. There is a surface layer, which is obviously less dense than the rest of the coating. This layer is the "velvet" film seen on asphalts, which consists of partially oxidized organic material. The more-completely oxidized material has been leached from this layer, leaving a film of low density. Below this surface layer is a denser layer; this second layer possibly consists of an accummulation of non-diffusing materials (primarily asphaltenes) which have partially collapsed on themselves because of a deficiency of oils. The third layer is again less dense, possibly because the oils have diffused out, leaving an incompletely filled, but not collapsed, asphaltene structure. Finally, the bulk of the asphalt remains below.

This particular film is 19.5 mils thick; it had lost 6.5 mils in thickness and 24% of its initial weight.

Figure 4 - Cross-Section of Asphalt with 50% Dolomite Weathered 75 Months on the Roof of the Industrial Building - Magnification 240X

Several strata of varying density are quite apparent. The surface layer seems to be almost entirely mineral matter with little asphalt to hold it together. The specimen is unsupported; hence, the apparently detached mineral matter is actually held in position by some material transparent to x-rays of this wavelength.

Below the low-density surface layer is a high-density layer. This area seems to be unusually rich in material between the dolomite particles.

The lowest layer, almost 50% of the film, is of an intermediate density.

A possible interpretation of this picture may be that the weathering at the surface has depleted the surface of asphalt binder through oxidation followed by solution of water-soluble end products. The second layer is of high density because the least dense material, the oils, have diffused to the surface and have left the more dense resins and asphaltenes behind. The rest of the film is changed to a lesser degree. It is more akin to the original coating.

FIGURE 3. AN ASPHALT FILM WEATHERED 88 MONTHS OUT OF DOORS - 120X

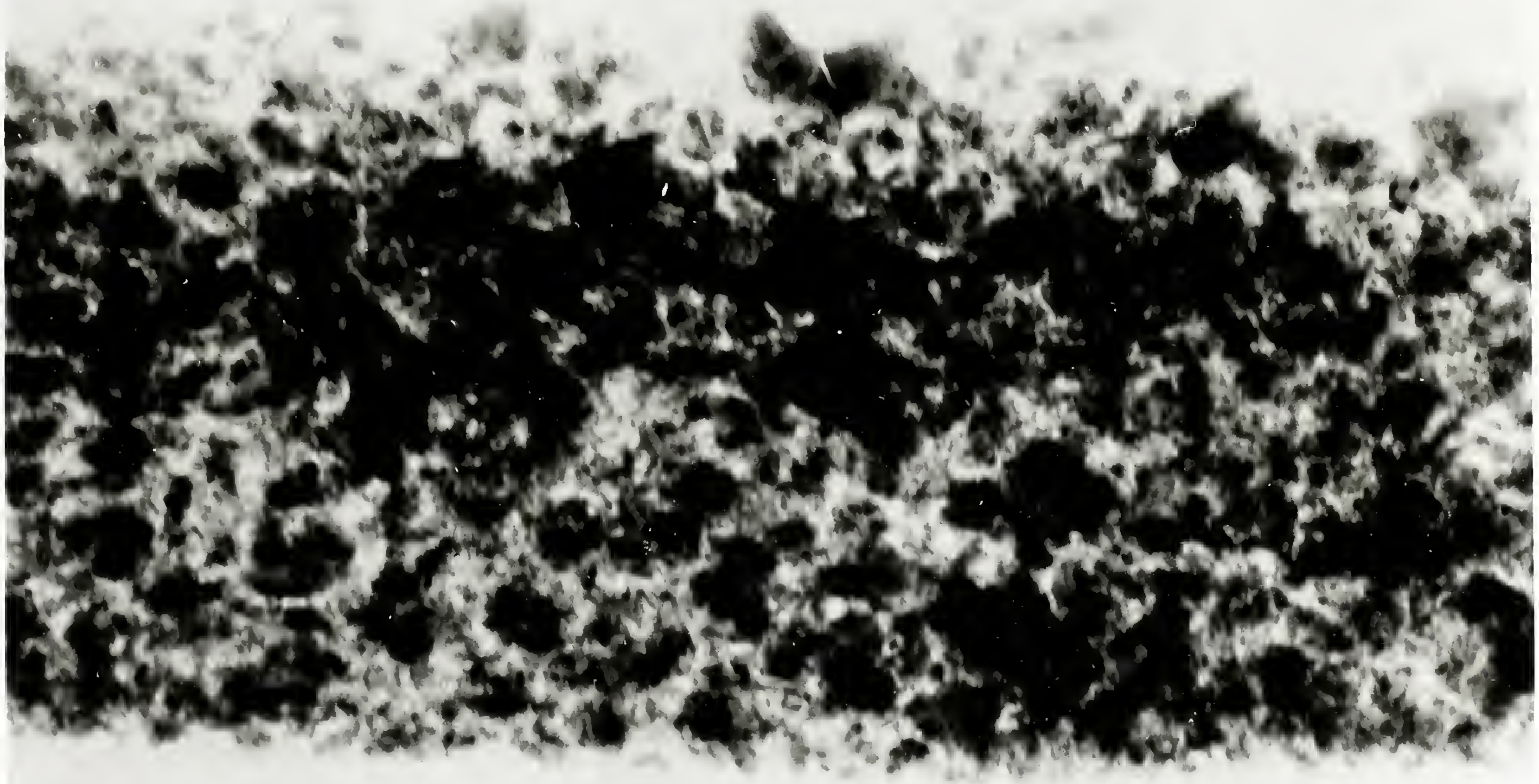

FIGURE 4. AN ASPHALT FILM CONTAINING 50% DOLOMITE, WEATHERED 75 MONTHS OUT OF DOORS - 240X

Figure 5 - Random Sample of 215-pound Uniform-Thickness Asphalt Shingle - Magnification 70X

This sample of a commercial asphalt strip shingle may not be typical, but it demonstrates vividly the value of the x-ray microscope for observing latent failures in roofing products. The mechanical embedment of the granules is quite evident. There is no contact (let alone adhesion) of coating and granules in large areas of their interface.

Some coating has been brought on to the exposed surface of the granule in the center of the photograph, possibly by rotation of the granule during embedment. A thin spot in the coating exists just to the right of this granule.

Regions in the saturated felt show significantly lower density than others. If these areas are less dense because of lower saturant, it would appear that the interfaces between coating and felt have become depleted in saturant. The saturant could have fluxed with the coating or could have been driven back into the felt. Small isolated areas in the body of the felt seem to be lacking saturant as well.

The variation in thickness of back-coating and back surfacing is quite apparent.

Figure 6 - Shingle That Has Clawed - Magnification 70X

This section taken from the center of a tab of a clawed, thick-butt shingle shows the condition existing when extreme clawing has occurred. The double overlay construction is apparent and still in good condition. A thin strata of saturated felt, just below the coating, also seems to be in good condition. (The low-saturation interface area seen in Figure 5 is not apparent here.) However, just below this layer is the bulk of the saturated felt, largely deteriorated. The back surfacing is full of holes and some of it has worked up into the felt. It appears that the original shingle may have had a light back-coating.

Figure 7 - Peace River Glass Mat with Kansas Asphalt. Thickness - 42 mils. Exposed 600 hrs to 51-9C cycle. Magnification - 110X

The glass mat occupies the lower two-thirds of the total thickness of this film. The mat is quite open and the individual fibers are surrounded by asphalt. The asphalt surface shows some signs of weathering; it is less dense than the interior and seems to have a fuzzy texture. The mottled appearance of the asphalt is difficult to explain, but is not untypical. Many asphalt specimens, before exposure, appear like this one.

FIGURE 5. A COMMERCIAL, UNEXPOSED UNIFORM-THICKNESS ASPHALT SHINGLE - 70X

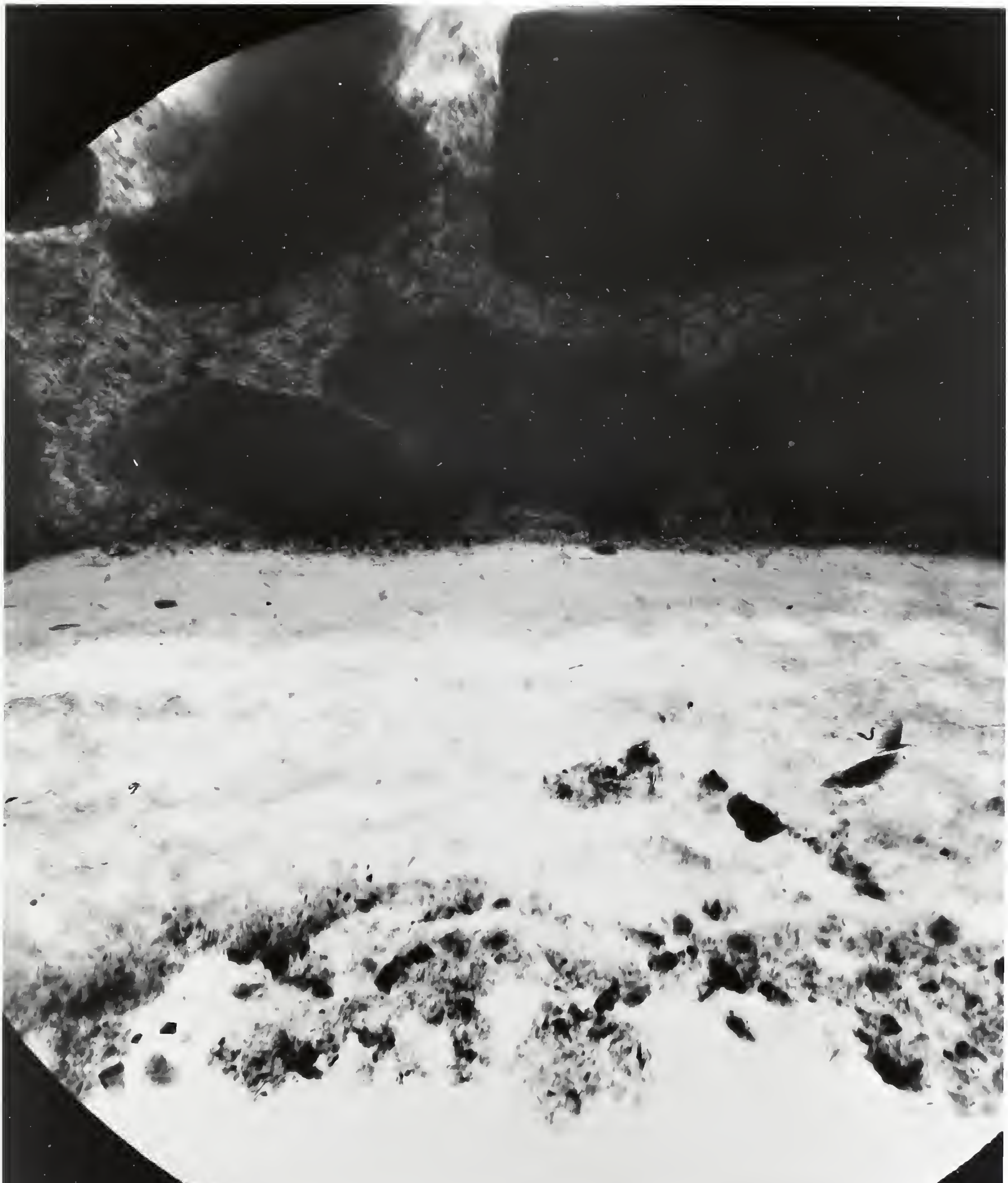

FIGURE 6. A THICK BUTT SHINGLE THAT HAS DEVELOPED SEVERE CLAWING - 70X

FIGURE 7. AN ASPHALT FILM CONTAINING A PEACE RIVER GLASS MAT AFTER 600 HOURS IN THE WEATHER-OMETER - 110X

Figure 8 - Johns-Manville Glass Mat with Kansas Asphalt. Thickness - 30 mils. Exposed 600 hrs to 51-9C cycle. Magnification 120 X.

The stratification occurring during weathering is quite apparent in this picture. The glass mat, which is of the same weight as the one in Figure 7, tends to be concentrated in the lowest part of the asphalt film. Some debris is clearly seen in the asphalt.

Figure 9 - Monoform Roofing - Clay Emulsion Reinforced with Chopped Glass Fibers on a Coated Sheet - Magnification Not Determined.

The mineral matter in the coating on the saturated felt is visible at the bottom of the picture. The random and irregular distribution of the glass fibers, as individuals and in clusters, throughout the clay emulsion residue is quite apparent. Clay particles are also quite apparent.

5. SUMMARY

The application of the x-ray point projection microscope to asphalt problems has been demonstrated through a number of photographs. The use of soft x-rays permits observence of variations in the asphalts as well as differences in absorbences among asphalts and additives.

6. REFERENCES

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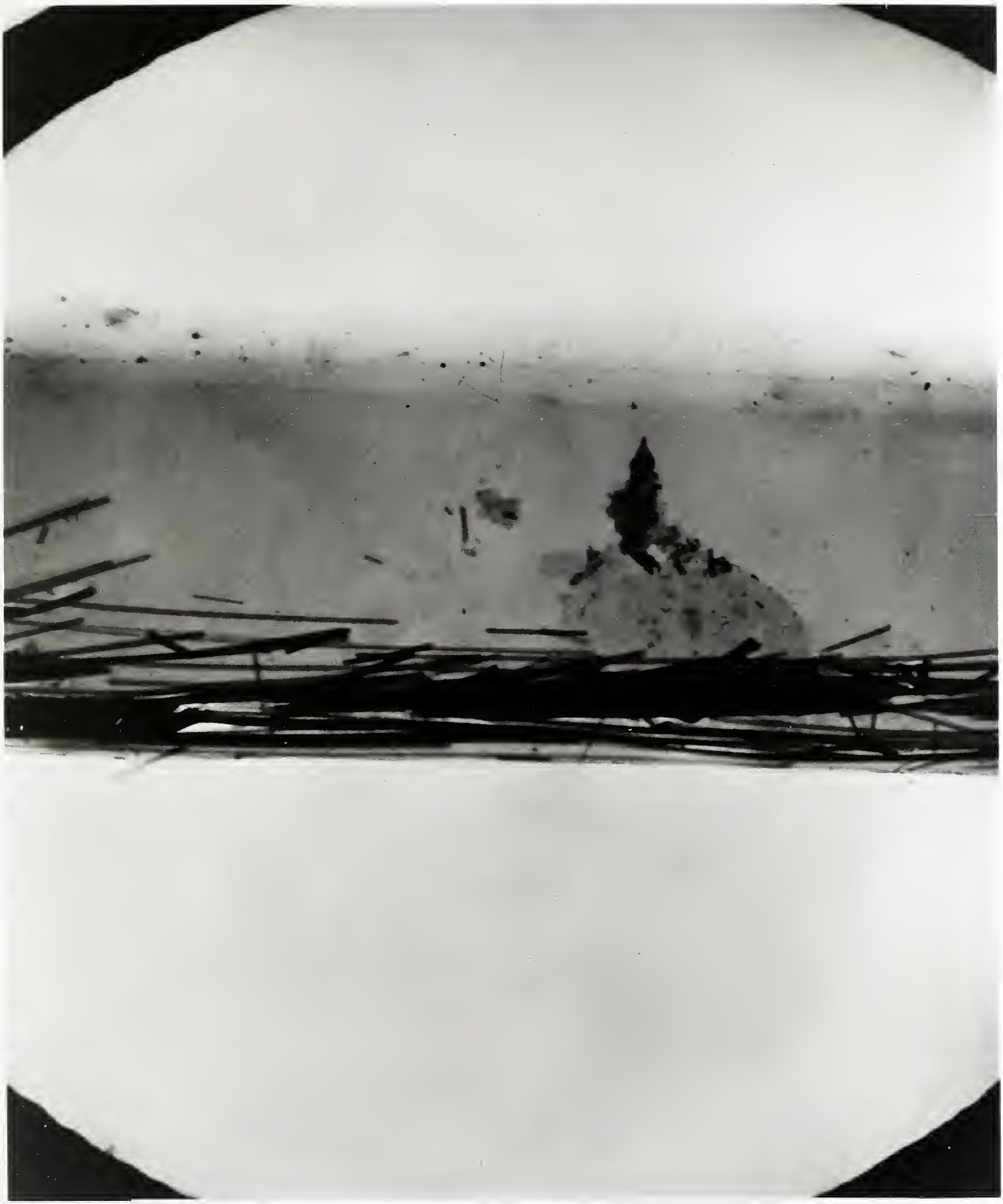

FIGURE 8. AN ASPHALT FILM CONTAINING JOHNS-MANVILLE GLASS MAT AFTER 600 HOURS IN THE WEATHER-OMETER - 120X

FIGURE 9. GLASS FIBER REINFORCED CLAY EMULSION ON A COATED BASE SHEET

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