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Abstract

Full Text

PHYSICAL CHEMISTRY

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EMISSION OF CHEMICALLY ACTIVE PARTICLES DURING THE OXIDATION OF ALUMINUM

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We have found that a freshly prepared aluminum surface in vacuum produces a contact impression on X-ray film when kept under atmospheric conditions. The results of a study of this phenomenon are presented below. The specimens were prepared by vacuum deposition of aluminum onto glass plates (4.5×4.5 cm^2), mounted along the walls of a Pyrex cylinder (diameter 39 cm, height 32 cm), at the center of which, on several turns of tungsten wire, weighed portions with a total weight of 0.5 g were fastened.

After the apparatus had been evacuated to a pressure of $\sim 5 \cdot 10^{-5}$ mm Hg and prolonged degassing had been carried out, the weighed portions were evaporated; air was then admitted into the apparatus, and the specimens were rapidly (25-30 sec) transferred to the X-ray film. Mirror deposits gave a reproducible, although very weak, blackening of the film. To intensify the effect, instead of a mirror deposit a finely dispersed aluminum aerosol was deposited on the plates, obtained by evaporating the weighed portions in an argon atmosphere at a pressure of 0.1-1.0 mm Hg. (^{1})

Fig. 1. Kinetics of oxidation of an aluminum aerosol with an average particle size
$d \sim 10^{-5}$ cm (740 mm Hg, 23°C)

Various commercial light-sensitive photographic plates and films were tested as detectors.

However, in all cases no blackening was detected. Only X-ray films of types X and XX (from different factories) proved suitable. It was noticed that the sensitivity of these films to the given effect increases with aging, giving clear results approximately one year after manufacture. Attempts to use a photomultiplier

Fig. 2

Figure 2: Fig. 2

with a uviolet window proved unsuccessful both in current and in pulse modes. It was established that thin (1 mm) luminescent crystals of CsJ, NaJ, KJ, anthracene, and naphthalene not only do not amplify, but completely eliminate the effect. No sensitizing action of an oil film was detected either.

To clarify the role of oxygen in the observed effect, a special apparatus was constructed in which the specimen could be moved and pressed against the X-ray film in successive positions without breaking the vacuum. After deposition of a mirror coating or aerosol on a glass plate, the apparatus was evacuated to a pressure of $\sim 10^{-4}$ mm. Experiments showed that, for equal exposures (10 hours each), in vacuum and atmospheric

under these conditions blackening of the film occurred only in the presence of air. In addition, these experiments established the absence of radiation that might have been expected as a result of crystallization of the initially amorphous particles of aluminum aerosol (^{2}).

Figure 1 gives a histogram of the dependence of the blackening density of type X film over 1 hour on the time after preparation of samples with an aerosol having an average particle size of $\sim 10^{-5}$ cm (measured with an electron microscope). The width of the steps corresponds to the time during which each of the samples was kept in contact with the given piece of film. The height of the steps is equal to the integral blackening density divided by the exposure time. The blackening density was measured with a DFE-10 densitometer and averaged over the area of the sample. The shaded rectangles show the root-mean-square scatter of the data for six samples. The solid curve drawn through the average points gives an idea of the kinetics of the process caused by oxidation of aluminum.

Fig. 2. Burning-out of a finely dispersed aerosol (average particle size $d \sim 10^{-6}$ cm) on a heated glass surface.

It is interesting to note that finely dispersed aerosols with particle size $d \sim 10^{-6}$ cm gave impressions in the form of the outline of the plate, as shown in Fig. 2. This is explained by the difference both in the heating temperatures and in the dispersity of the aerosols on the plane and on the edges of the plate. Indeed, since air was admitted into the instrument immediately after evaporation of the deposition, owing to heating of the plane of the plate the aerosol was oxidized before the sample fell onto the film. The particles settling on the edges, however, traveled a longer path in argon and, consequently, had time to grow to a larger size (^{1}).

The lowering of the temperature at the edges of the plate and the presence of larger particles considerably slow the oxidation process. For comparison, we note that aerosols with particle size $< 10^{-6}$ cm sometimes ignite spontaneously

Fig. 3

Figure 3: Fig. 3

Fig. 4

Figure 4: Fig. 4

upon first contact with air. To determine the nature of the active agent, various substances were placed between the sample and the film. The experiments showed that the effect is completely eliminated by the following materials used: black paper in one layer, a mica plate (10 μ), a cover microscope glass (0.1 mm), and a disk of optical quartz (2 mm).

Figure 3 shows typical impressions of samples with a shadow from a cover glass and a quartz disk (40 mm in diameter) for a two-day exposure (in the latter case the shadow of a brass ring 2 mm thick is also shown). Measurements with a densitometer established that the blackening density of the film under the parts of the cover glass and quartz covered by the samples is indistinguishable from the background. The most demonstrative measurements are of the effect as a function of the thickness of the air interlayer between the sample and the film. To avoid possible shedding of aluminum powder from the glass plate (although this was unlikely), in all experiments the film was placed above the sample.

Impressions of brass rings (exposure–2 days) showed that the blackening density of the film inside the rings, to within the scatter of the experimental data ($\sim$ 20%), is constant over distances of $0 \div 3.5$ mm (the thickness of the rings was varied in steps of 0.5 mm each). At the same time, the edges of the sample beyond

beyond the limits of the ring gave, with increasing thickness of the air gap, an increasingly diffuse shadow (Fig. 3). In subsequent experiments, tubes of various lengths and diameters made of aluminum, copper, and brass were used. The tubes were thoroughly washed in benzene and alcohol. The dependence of the density of blackening of the film on the length of the air column inside the tubes is shown in Fig. 4 (exposure–2 days). It is easy to see that the data for different materials practically fall on one and the same smooth curve.

Convincing evidence is provided by the experiment in which the gap (1 mm) between the specimen and the film was blown through with a strong jet of compressed air. In this case the effect is completely absent, although a control specimen, with the same gap width and the same exposure (8 hours), gave a clear picture.

Fig. 3. Impressions on cover glass and on a quartz disk, demonstrating the absence of light radiation during oxidation of aluminum aerosol ($d \sim 10^{-5}$ cm, exposure–2 days)

Fig. 4. Absorption of the active agent by an air column inside tubes of various

materials: *a*—brass ($d = 16$ mm); *b*—copper ($d = 24$ mm); *v*—aluminum ($d = 30$ mm)

Summarizing the foregoing, the following conclusions may be drawn. The effect of blackening of the film is due to the chemical action of a very active agent released during the oxidation of aluminum. It is possible that this agent is ozone. The action of this agent on old X-ray film is detected at a considerable distance in air (up to 3 cm inside the tubes). Emission of light, both in the visible and in the ultraviolet regions of the spectrum, during the oxidation of aluminum has not been detected.

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Note: Figure translations are in progress. See original paper for figures.