DENSITY STRATIFICATION IN A LIQUID DURING TEMPERATURE CHANGES

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The formation of density strata has been observed in a broad class of liquids. This phenomenon has generally been thought of as being of a physical (mechanical) nature, rather than an effect of the nature of the solutions. It has been suggested that this effect may play a role in crystallization.

The effect we describe below is of some interest in connection with stratification in liquids and in solids crystallizing in them.

A concentration gradient of a solute is set up along the axis of test tube holding a room-temperature solution, and then the test tube is immersed in a vessel containing water at 60-90°C. After 5-10 sec, the illumination with a parallel light beam reveals strata with various impurity densities; there is an alternation of larger and smaller refractive indices of the adjacent layers (Fig. 1). An analogous effect occurs during the rapid cooling of a previously heated test tube.

After 5-10 min, diffusion erases the sharp boundaries between strata. The strata formed in the region with the least concentration gradient remain distinguishable for a long time. This effect is apparently of a general physical nature, since it is observed in aqueous solutions of sugar, potassium ferricyanide, $NiSO_4$, NaCl, KCl, $ZnCl_2$, $CdCl_2$, and NH_4Cl.

We note certain aspects of this effect. It is observed in either a vertical or horizontal test tube. In both cases, the strata are normal to the concentration gradient. When there is no initial concentration

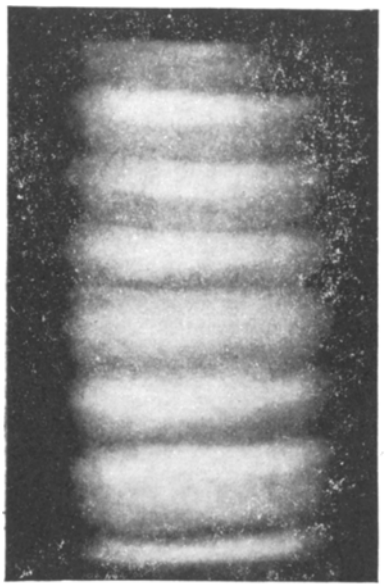

Fig. 1

Fig. 2

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gradient of the impurity, the strata do not arise during the subsequent temperature change. The strata do not arise during a slow heating or cooling, even when there is an initial concentration gradient. The appearance of the strata does not depend on the magnitude of the concentration, and it occurs at concentrations very far from saturation.

Investigators working with crystallization are quite familiar with the stratification of impurities in crystallized samples. Figure 2 shows impurity stratification in a polycrystalline aluminum rod obtained by the Stepanov method [1]; this phenomenon has not been completely explained. We suggest the melt stratification at a crystallization front may cause the impurity strata in the crystallized sample. When the effective impurity-distribution coefficient ahead of the crystallization front is significantly different from unity, there is always an impurity concentration gradient. The melting point changes as a result of the relative motion of the liquid and the phase boundary. The oscillations of the crystallization front which are observed experimentally [2] may also be explained on this basis. We note that a similar situation may occur during the stratification of geological sediments [3, 4].

Unfortunately, there is no explanation for this effect, nor is there a mathematical model for it. Before carrying out these experiments, we were aware that Schaaffs [5] had attributed the stratification observed in an aqueous solution of copper sulfate upon rapid cooling to a colloidal structure. However, our experiments rule out this explanation and show that the effect is more general, appearing under quite varied conditions.

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