On Nucleation and Growth of Silver Halide Tabular Grains

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Introduction

AgHal grains is a multistage process and final grain size, thickness, size distribution as well as grain shape depend on each stage conditions. A scrutiny of two main Precipitation of uniform tabular precipitation stages - nucleation and growth shows that they can proceed at the expense of coalescence or diffusion. Conditions of the nucleation might govern nearly all grain dispersion characteristics. It is also reasonable that conditions of the nucleation and the growth can cause prevailing of one of these two mechanisms. The aim of this paper is to prove this supposition and to find correlation between the prevailing mechanism and the final tabular grain size, thickness and their distribution.

Tabular grain kinetics simulation

It seems to be generally accepted that at the outset of tabular grains precipitation regular nuclei form. These nuclei converse into tabular nuclei either by coalescence or by diffusion. Each ripening of the fine AgBr also causes tabular mechanism must have its own peculiar kinetics. Since grain formation its kinetics can be used as a model of tabular nuclei growth kinetics.

The diffusion growth kinetics. In case of diffusion growth the moving force of the process is Ostwalt ripening. Since both diffusion and material deposition are not the slowest stages of the process we choose grain dissolution as the limiting stage. Assuming that dissolu-tion rate of AgBr grain varies directly with its solubility we have calculated a dissolution kinetics of the fine AgBr emulsion of d=0,07 mcm, Cv=15% and conventional size distribution. The calculated kinetics is thus a tabular nuclei growth kinetics and is shown on fig 1.

The coalescent growth kinetics. In case of coalescent tabular nuclei formation its kinetics can be calculated by the Smoluchowski-Muller equation for rapid agglomeration of the biphase system. The first phase are fine regular grains and the second one are constantly growing tabular grain. The coalescent growth kinetics is calculated with respect to the model of the anisotropic coalescent growth.[1] The calculated kinetics of the agglomeration and thus tabular nuclei growth kinetics is shown on fig 1.

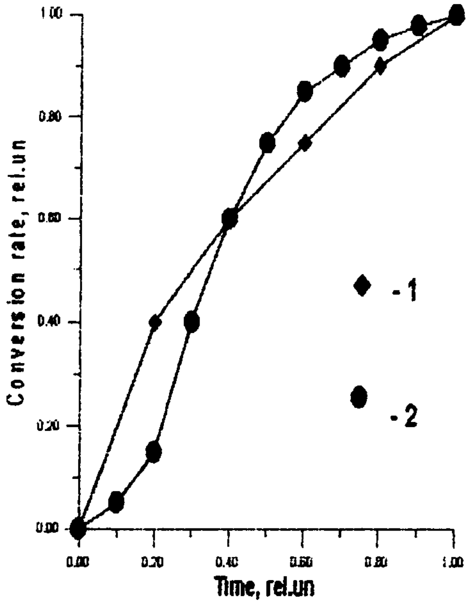

Figure 1. Tabular grain nuclei growth kinetics In case of (1) diffusion or (2) coalescent growth

Results and Discussion

A procedure of tabular grain growth kinetics investigation during fine AgBr ripening was reported recently.[2] Conditions variety of the fine AgBr ripening are depicted in table I.

All tabular grain growth kinetics in the range of ripening conditions from table I have a specific S - shape and thus they are very close to the simulated kinetics of the coalescent growth. Varying of the ripening conditions shifts a kinetics curve to more rapid or slow one but it doesn't change its shape. This result undoubtedly proves that the growth mechanism in a wide range of the ripening conditions is coalescent.

Table I Conditions of fine AgBr ripening

pBr value	1,0 - 1,5
temperature	40 - 60 Celsius
gelatine concentration	2 - 8 %
potassium nitrate concentration	0 - 2 mol/L

Some previous papers[3,4] dedicated to the study of AgHal ripening in presence of effective silver halide solvent can be interpreted as an evidence of the prevailing diffusion mechanism. Kinetics of the fine AgBr ripening in presence of ammonia prove this supposition. Fig.2 shows tabular grain growth kinetics in presence of different amounts of the ammonia. All kinetics have been obtained with regard to the fine AgBr enlargement due to the Ostwald ripening. It is seen that adding of ammonia drastically changes the kinetics shape proving an overwhelming diffusion.

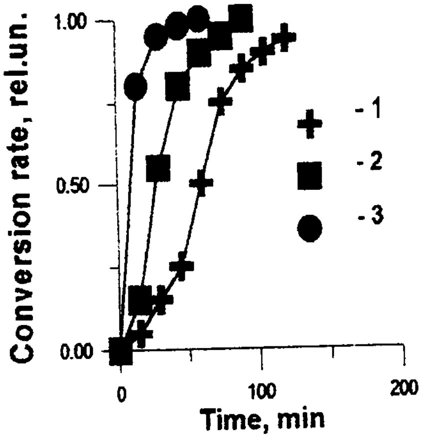

Figure 2 Tabular grain growth kinetics dependence on ammonia concentration. 1 - 0 M/M Ag; 2 - 0,01 M/M Ag; 3 - 0,05 M/M Ag.

The growth mechanism reveals a great determination of the final emulsion uniformity. Just small amounts of the ammonia greatly decrease emulsion uniformity causing appearance of rods, nonparallel twinned grains and an increase of the tabular grains size and thickness variety coefficient. The coalescent growth reveals much more perfect uniformity of the tabular grains

It is found that in case of the prevailed coalescent growth the growth kinetics influences tabular grain emulsion dispersion characteristics. Fig 3 shows dependence of an average tabular grain size and variety coefficient on pBr.

It is seen that size and Cv vary with pBr as well as with other kinetics factors which accelerate the growth (a decrease of pBr accelerates the ripening; in the same way it affects the growth rate, for example, unrelated salt concentration increase). A plausible explanation of dependence (see fig 3) can be derived from the model of anisotropic coalescent growth, which was published recently[2]. According to the model parallel twinned fine grains act as a coalescent nuclei, the lateral growth coinciding with twin planes direction. A moving force of the anisotropic coalescence is an electrostatic attraction between negatively charged surface of regular fine grain and a positively charged side face of the tabular grain. A rate of attraction depends on a double electric layer potential of the tabular grain and hence on its thickness and symmetry. A convenient fine pBr emulsion comprises parallel twinned grains of different thickness and symmetric perfection and thus having different ability to attract regular grains. This results in different growth rate of each specific tabular grain. An accelerating of the tabular growth (at lower pBr level for example) model parallel twinned grains with the lowest potential ineffective regarding to those with higher potential. The more effective nuclei consume the less effective among other regular grams and thus the number of coalescent growth centres decreases. Since total fine grain amount in our ripening experiments is always constant the size of the resulting tabular strains becomes greater with growth acceleration.

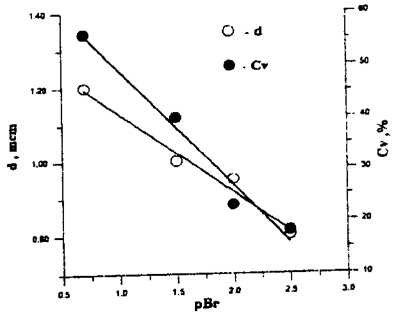

Figure 3. Average grain size and variety coefficient dependence on ripening pBr value. 1 - d, 2 - Cv

Conclusions

The growth of tabular grains can protect through two channels: diffusion and coalescence. AgHal solvents cause prevailing of the diffusion when in absence of solvents the diffusion is negligible. The decrease of pBr enhances diffusion though the coalescence sleeps prevailing. The increase of the diffusion component aggravates both size and crystallographic uniformity of tabular strains.

A comprehension of the role of the growth mech-anism is very important for a corroboration of highly uniform tabular grain emulsion since at the outset of tabular grains precipitation only regular nuclei are forming. Their conversion into the uniform tabular grain nuclei needs conditions which exclude diffusion. Double jet precipitation following the uniform tabular nuclei for-mation will finally result in uniform tabular emulsion formation.

References

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