annual review

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Crystallization

Part I. Transport Phenomena of Nucleation and Crystal Growth

Papers dealing with the transport phenomena involved in crystal growth and nucleation are reviewed

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This review is actually a continuation of the 1969 crystallization review not only in a chronological sense but also in another aspect. The general introductory comments on the field of crystallization, as well as the introductory statements inserted into the various sections and subsections for the purpose of tying current developments to previous works and accepted ideas in the field, were not repeated in this year's review. The reader is urged, if he wants to view the current papers in perspective, to read also the corresponding sections in last year's review (*2A*-*4A*).

This year the review has been subdivided into two parts. In Part I papers dealing with the *transport phenomena* involved in crystal growth and nucleation are considered. Papers dealing with the *process* and the various techniques of crystallization will be reviewed in Part II.

The review covers the one-year period from the Spring of 1969 to the Spring of the current year, 1970. Most, but not all, of the papers published during that period are included. Certain papers touched several aspects of crystallization, and they are presented and discussed in more than one place in this review. Papers on the crystallization of polymers and on the recrystallization in a solid phase were intentionally omitted.

A section on a process related to crystallization, *i.e.*, scale formation on a heat transfer surface, has been included in the crystallization review for the first time.

Books and Reviews

Many books and reviews dealing with crystallization have appeared since the last review (*2A*, *3A*, *4A*).

The book "Crystallization from Solutions and Melts" edited by Palermo and Larson (*22A*), brings together a collection of papers originally presented at an AIChE Symposium. One group of papers deals with recent developments in industrial crystallizers. Such subjects as population balances, mixing effects in continuous crystallizers, continuous sugar and urea crystallization, and the dynamic behavior of isothermal crystallizers are covered. Another group of papers discusses the growth and purification of crystals from the melt.

A book that delves into many areas of nucleation was edited by Zettlemoyer (*31A*). Special emphasis was given to the theories of homogeneous and heterogeneous nucleation. Nucleation theories were also discussed in a paper by Lothe and Pound (*15A*).

Henisch (*10A*) in his book "Crystal Growth in Gels," describes the nucleation and growth of crystals in gels. Henisch also traces the history and nature of this technique of growing crystals, and discusses the role that the structure and properties of the gel play.

In their book "Physics of Ice," Riehl, Bullemer, and Engelhardt (*25A*) consider the growth and properties of different forms of ice crystals.

Several books were translated from Russian. These include Volumes 7 and 8 of the series "Growth of Crystals" edited by Sheftal (*26A*, *27A*). Books on crystal growth from solution by Khamskii (*12A*) and Petrov, Treivus, and Kasatkin (*23A*) were also translated from Russian. Another translated book (*9A*) deals with the growth and properties of semiconductors. A book on solid state problems edited by Madelung (*16A*) has some chapters in English and some in German.

Matz (*18A*) edited a book on crystallization principles and techniques, while Suchet (*28A*) edited a text on the growth of monocrystalline inorganic compounds.

Excellent review papers have also been published recently. A review of the articles on crystallization published during 1966 and 1967 was edited by Matz (*17A*). A paper by Mullin (*19A*) outlined the principles used in the design and evaluation of industrial crystallizers. Tiller (*29A*) reviewed the technology used in the growth and purification of single crystals.

The growth of single crystals and epitaxial films from the vapor phase was discussed by Nitsche (*21A*), while Mutafchiev (*20A*) considered the thermodynamics and kinetics of crystallization. The growth of single crystals from liquid and vapor phases, zone melting and zone refining, and the Bridgeman–Stockbarger and Verneuil growth techniques were described by Hurle (*11A*).

The basic principles and methods of crystal growth were covered in a USSR Symposium edited by Chang (*5A*). Methods of growing and etching semiconductors and single crystals were discussed.

The equipment, phase rules, and conditions involved in the growth of inorganic compounds at high temperatures and pressures were reviewed by Kleber and Wilke (*13A*). The kinetics and methods used to produce organic substances were treated by Anikin, Dugacheva, and Merzhanov (*1A*). Elliot (*8A*) discussed the preparation and structure of apatite crystals.

Chinmulgund (*6A*) reviewed the growth of pure and substituted single crystals of garnet from the melt and from solution, and described and compared the methods. Recker (*24A*) discussed the growth of laser crystals by Verneuil and Czochralski methods. Wakatsuki (*30A*) reviewed the synthesis of diamond crystals and described the apparatus, methods, catalysts, and conditions of growth that are used. Dipietro (*7A*) reviewed the growth of alum single crystals, while the preparation of gallium arsenide crystals by the Czochralski, floating zone, and Bridgeman techniques was described by Klohn and Wandinger (*14A*).

Nucleation of Crystals

Nucleation in general involves a phase change that results in the appearance of new material in a system. We are interested here in the particular case when this new phase is a solid. The nucleation of crystals from vapor, liquid, and solid phases are all

PART I.
Transport Phenomena of Nucleation and Crystal Growth

- B. Nucleation of Crystals
- C. Secondary Nucleation
- D. Studies in General Crystal Growth Theory
- E. Experimental and Theoretical Studies on Solution Growth
- F. Experimental and Theoretical Studies of Crystallization from Melt
- G. Experimental and Theoretical Studies of Crystallization from Vapor

PART II.
Crystallization Processes

- H. Crystallization Processes involving Suspensions of Crystals—Theory and Experimental Studies
- J. Co-precipitation and Purification in Crystallization Processes from Solution
- K. Development and Studies on Industrial Crystallizer Practice
- L. Production of Single Crystals from Melt
- M. Purification by Melt Crystallization
- N. Miscellaneous Crystallization Systems, Processes and Techniques
  - (Na) Vapor-Liquid-Solid Growth
  - (Nb) Gel Growth
  - (Nc) Whisker Growth
  - (Nd) Hydrothermal Growth
  - (Ne) Processes Related to Crystallization: Scaling
  - (Nf) Ice

relatively well studied but not necessarily well understood phenomena.

Generally a distinction is made between homogeneous nucleation (nucleation that occurs in the absence of a second phase), and heterogeneous nucleation (nucleation that occurs in the presence of a second, foreign phase). A further distinction is made if nucleation occurs in the presence of a second phase and if the second phase is the same material that is being nucleated. This is usually called secondary nucleation and will be discussed in the next section of the review.

The papers in this section of the review have been classified along the above lines and presented in Tables I-B, II-B, and III-B.

Books and reviews on nucleation. A recent book edited by Zettlemoyer (*88B*) discusses many of the basic concepts of nucleation. In its 11 chapters, the 12 contributors cover a wide range of nucleation processes. Included are a general and theoretical introduction to nucleation processes (*25B*), a discussion of nucleation in liquids and solutions (*85B*), a description of nucleation in solids (*41B*), and a review of graining in sugar boiling (*84B*).

Hirth and Moazed (*39B*) surveyed the present state of heterogeneous nucleation theory and presented a comprehensive 40-page report. Pound and Karge (*68B*) reveiwed and compared the theories that describe the process of nucleation on heterogeneous surfaces, and the theories that attempt to explain the type of nucleation that results in the oriented overgrowth (epitaxy) of thin films.

Homogeneous nucleation theories. The classical theory of homogeneous nucleation first put forward by Volmer, Becker, Doring, and others about 40 years ago continued to be tested, examined, and expanded upon. This theory postulated that clusters of molecules (embryos) formed in a supersaturated environment by a process that began with the combination of two molecules to form a dimer. Successive additions of one molecule at a time to this dimer yielded in turn a trimer, tetramer, etc. Since this clustering process was considered to be a reversible one, some embryos grew at the same time others were dissolving. However, once an embryo attained a certain critical size, it could decrease its total free energy by growing, and therefore became stable. The problem of predicting the nucleation rate thus became one of calculating the rate at which embryos attained this critical size. To simplify the mathematics, the early investigators derived the steady-state nucleation rate. This was the nucleation rate that would exist after a steady-state distribution of different sized embryos was attained, if the concentration of unclustered molecules (monomers) remained constant, and if clusters larger than the critical size no longer entered into the nucleation process.

Gerlach (*32B*) re-examined the original theory and predicted that we should observe two different kinds of nucleation behavior that can be distinguished by the ease with which nuclei near the critical size grow. Kaishev and Stoyanov (*45B*) looked at the case where the phase being nucleated was not the equilibrium form of the crystals. They report that this nonequilibrium crystal form should be nucleated along with the equilibrium form at all supersaturations.

Kashchiev and Kaishev (*47B*) calculated the nucleation rate that should result if the initial concentration and distribution of embryos were not the steady-state values. Many others also studied the process of unsteady-state nucleation (*2B*, *34B*, *35B*, *46B*, *48B*, *72B*).

Sometimes sophisticated techniques are used to describe the process of homogeneous nucleation. White (*87B*), for example, used a steady-state random walk technique to model homogeneous nucleation. Wette, Allen, *et al.* (*86B*), in a computer experiment, simulated the crystallization of a system of particles interacting through a Lennard–Jones potential.

Heterogeneous nucleation theories. As a practical matter, homogeneous nucleation is difficult to attain even in the laboratory. Trace amounts of insoluble matter are always present, even in the purest materials. Even if this insoluble matter could be eliminated, the walls of the containing vessel would still present a second phase that the supersaturated phase would be exposed to. Therefore, there is great practical motivation for studying heterogeneous nucleation. Normally, two types of heterogeneous nucleation are observed. The first and more general case is when nucleation occurs in a random fashion at various sites on a substrate surface. The second is a special case of the first and occurs when the nuclei form on the substrate in such a way that the nuclei all have the same lattice orientation. The orientation of the nuclei is usually determined by the orientation of the substrate lattice. This second case of oriented overgrowth is called epitaxy. Much interest has been shown in trying to explain why one type of heterogeneous nucleation occurs in a random fashion while another leads to ordered nucleation and growth.

A model proposed by Kenty and Hirth (*50B*) predicts that epitaxy will take place in systems where the nucleus–substrate interfacial free energy is low, while random nucleation will occur for higher interfacial energies. Crosley, Douglas, and Mondolfo (*18B*) have examined the factors that determine the interfacial energies between the nucleus, substrate, and supersaturated phase. Tiller and Takahashi (*82B*) determined the electrostatic contribution to the total interfacial free energy between nucleus and substrate. A material with the proper electrostatic properties will have a low nucleus–substrate interfacial energy and be an effective substrate.

Fletcher (*29B*) investigated the possibility that nucleation occurs at active sites on the nucleating particles and suggested that these sites are probably small, re-entrant corners or jogs in growth steps on the surface of the nucleating particle. Ball and Venables (*6B*),

TABLE I-B. REFERENCES ON BOOKS, REVIEWS, AND THEORIES OF NUCLEATION

Books and Reviews

(*12B, 13B, 16B, 25B, 26B, 39B, 41B, 68B, 84B, 85B, 88B*)

Homogeneous Nucleation Theories

(*1B, 2B, 11B, 23B, 27B, 31B, 32B, 34B, 35B, 44B, 45B, 46B, 47B, 48B, 52B, 56B, 67B, 71B, 72B, 73B, 76B, 78B, 86B, 87B*)

Heterogeneous Nucleation Theories

(*3B, 6B, 7B, 9B, 13B, 14B, 18B, 19B, 21B, 29B, 38B, 50B, 53B, 59B, 60B, 62B, 64B, 75B, 77B, 82B*)

TABLE II-B. STUDIES OF HOMOGENEOUS NUCLEATION

Pure Systems

Crystal	*Type of nucleation*	*Reference*
25 inorganic materials	solution	(*63B*)
AgBr	solution	(*69B*)
$BaSO_4$	solution	(*28B*)
$BaTiO_3$	melt	(*11B*)
Bi	melt	(*80B*)
citric acid	solution	(*61B*)
copper(II) 8-hydroxyquinolinate	solution	(*70B*)
H_2O	vapor	(*4B*)
In antimonide	melt	(*52B*)
KBr, KCl, KI, LiF, NaCl, NaF	solution	(*34B*)
Pb	melt	(*80B*)
Sn	melt	(*80B*)
Th(IV) oxyperchlorate	solution	(*5B*)

Impure Systems

Crystal	*Impurity*	*Type of nucleation*	*Reference*
$BaSO_4$	iron, magnesium, Teflon, thorium	solution	(*10B*)
$BaSO_4$	AgCl, Al_2O_3, $BaCrO_4$, NaCl	solution	(*28B*)
Bi	Cd, In, Pb, Sn, Te, Tl, Zn	melt	(*81B*)
$FeSO_4$	$MnSO_4$	solution	(*58B*)
KCl	$CaCl_2$, KNO_3, $MgCl_2$	solution	(*79B*)
Pb	Bi, Sn, Te, Tl	melt	(*81B*)
Sn	Bi, Cd, Cu, In, Pb, Te, Tl, Zn	melt	(*81B*)

Other References

Subject	*Reference*
nucleation of crystals in a tension field	(*8B*)
nucleation of $Al(OH)_3$ in an electron beam	(*15B*)
nucleation of anthracene under ultraviolet radiation	(*30B*)
technique for nucleating crystals in a centrifuge	(*74B*)

in experiments designed to test current nucleation theory, nucleated rare gas crystals on cooled graphite substrates under controlled conditions.

Experimental studies of homogeneous nucleation. Studies observing the nucleation of a particular material in the absence of a deliberately added foreign phase are presented in Table II-B. Experiments carried out in both pure and impure systems are listed, as well as a few references describing nucleation that occurred under special conditions.

Typical of the first class is a very extensive study by Nyvlt *et al.* (*63B*). Here the supercoolings at which 25 different inorganic solutions nucleated were determined at different temperatures, for different cooling rates, and in the presence and absence of seed crystals. The results were correlated by a nucleation model that could be used in the design of industrial crystallizers.

In some investigations, the effects of added impurities on nucelation were determined. Typical of this are studies by Svoronos (*79B*), who investigated the influence of $CaCl_2$, KNO_3, and $MgCl_2$ on the nucleation of KCl solutions, and Ferdousee and Nawab (*28B*), who nucleated $BaSO_4$ in the presence of both soluble and insoluble foreign particles.

The classical theory of homogeneous nucleation predicts that a whole range of different size embryos should exist in supersaturated solutions. Garten and Head (*31B*) argued that the phenomenon of crystalloluminescence is caused by the reflection of light by embryos that form in solutions undergoing nucleation.

Many years ago, Tammann observed that at very high supersaturations the rate at which organic melts nucleated began to decrease with increasing supersaturation. This was eventually attributed to the existence of a resistance to the diffusion of solute molecules across the melt–nucleus interface, something not generally considered in classical homogeneous nucleation theories. This phenomenon has been observed many times in melts, but Mullin and Leci (*61B*) only recently became the first to observe it for nucleation from solution.

Experimental studies of heterogeneous nucleation. Studies of heterogeneous nucleation are listed in Table III-B. The majority of these involve the vapor phase deposition of thin films on various substrates. Corbett and Boswell (*17B*), for example, studied the nucleation of Ag on single crystal MoS_2 substrates at different temperatures and deposition rates. Kosevich *et al.* (*55B*) created defects (color centers and impurity atoms) in KBr, KCl, and NaCl substrates, and then observed the deposition of thin films on these materials. They reported that nucleation occurred preferentially at the defects. Distler and Vlasov (*20B*), and Duemler and Kittl (*24B*) also studied the role that the structure and properties of the substrate play in heterogeneous nucleation. Hudson and Sandejas (*42B*) noted the type of nucleation that occurred on substrates whose surfaces had been contaminated by impurity, while Layton and Steger (*57B*) studied the nucleation of ice and liquid water on silver iodide substrates.

Secondary Nucleation

Nucleation that occurs in the presence of the same kind of crystals that are being formed is usually called secondary nucleation. Other names such as crystal breeding and false grain have also been used. Strictly speaking, the nuclei do not have to come from the seed crystals, but the presence of the seeds is essential to the nucleation process. This kind of nucleation has just begun to receive interest because it is now relatively clear that the nucleations that occur in the suspensions of crystals encountered in industrial crystallizers are predominantly secondary nucleations. In contrast with homogeneous nucleation, secondary nucleation exhibits relatively low order dependence on supersaturation—that is, the rate at which secondary nucleation occurs does not change as drastically with changes in supersaturation as the rate of homogeneous nucleation does. In addition, secondary nucleation occurs at considerably lower supersaturations than homogeneous nucleation. Although the existence of secondary nucleation has been recognized for some 60 years now, the mechanism or mechanisms by which it occurs are not completely understood and much remains to be done.

TABLE III-B. STUDIES OF HETEROGENEOUS NUCLEATION

Crystal	*Substrate*	*Type of nucleation*	*Reference*
Ag	MoS_2	vapor	(*17B*)
Ag	NaCl	vapor	(*38B, 51B*)
Ag	W	vapor	(*65B, 66B*)
AgCl	triglycine sulfate	vapor	(*20B*)
Ar	graphite	vapor	(*6B*)
Au	Ag	vapor	(*24B, 54B*)
Au	C film	vapor	(*24B*)
Au	Cu	melt	(*22B*)
Au	KBr	vapor	(*55B*)
Au	KCl	vapor	(*33B, 37B, 55B, 83B*)
Au	Mg	melt	(*22B*)
Au	MgO	vapor	(*49B*)
Au	NaCl	vapor	(*21B, 33B, 37B, 55B*)
$BaSO_4$	Al_2O_3, $BaCrO_4$, AgCl	solution	(*28B*)
Bi	KBr, KCl, NaCl	vapor	(*55B*)
Cd	W	vapor	(*42B*)
CdS	NaCl	vapor	(*21B*)
CdSe	CdSe	vapor	(*36B*)
CdTe	CdTe	vapor	(*36B*)
GaAs	GaAs	vapor	(*3B*)
Ge	Ge	vapor	(*3B*)
Kr	graphite	vapor	(*6B*)
Si	Si	vapor	(*43B*)
H_2O	AgI	vapor, liquid	(*57B*)
H_2O	He	vapor	(*4B*)
Mo oxide	CO	vapor	(*40B*)
W oxide	CH_4, CO, CO_2, N_2	vapor	(*40B*)
Xe	graphite	vapor	(*6B*)

A paper that has evoked considerable interest in crystallization circles was presented by Clontz and McCabe (*1C*) at a recent meeting of the American Institute of Chemical Engineers. They conducted experiments with magnesium sulfate heptahydrate, a material that had been used by other investigators in previous secondary nucleation studies. Working with a system in which solution continuously flowed past a seed crystal mounted on a special holder, Clontz and McCabe did not observe secondary nucleation until they lightly tapped (contacted) the seed crystal with a glass or stainless steel rod. This tapping resulted in considerable nucleation. Clontz and McCabe found that the number of crystals produced by these contacts increased more or less linearly with supersaturation and with the contact energy, but did not seem to be affected by the liquid velocity.

It might appear at first that Clontz and McCabe were merely observing a microattrition process in which small pieces were being chipped from the parent seed during the contacts. Clontz and McCabe did report that seed crystals which had been tapped did not seem to be visibly damaged when they were examined under a 40× microscope, but it could be argued that the defects likely to be caused by these contacts either could be too small to be seen at this magnification or could have been healed over quickly as the seed crystal grew. However, it might also be argued that since contact nucleation was affected by the supersaturation, the only way that contact nucleation could be a microattrition process and still be dependent on the supersaturation would be for the fragments produced by the contacts to be in the same size range as the critical nucleus. Whether this is very likely is somewhat debatable. Obviously, there can be much speculation as to the exact nature of contact nucleation, but it is agreed that this is a most important finding, one which will undoubtedly receive further study.

The experiments conducted by Nyvlt and his coworkers (*2C*) were noted in the preceding section of this review. As mentioned there, Nyvlt *et al.* determined the metastable zone width of 25 different inorganic solutions both with and without seed crystals present. In all 25 cases, the solutions nucleated much more readily with the seed crystals present.

According to the classical nucleation theory discussed earlier, embryos smaller than the critical nucleus should exist in supersaturated phases. Leichkis and Mikhailov (*3C*), in a study where tin was solidified from the melt, proposed that secondary nucleation could occur by a mechanism in which these embryos became activated at the surface of a seed crystal.

Studies in General Crystal Growth Theory

Most theoretical papers dealing with the growth rate of crystals can be classified into rather specific subgroups depending upon the assumed rate-limiting step. These subgroups are: nucleation (with subsequent growth), surface kinetics, mass transfer, and compound. The last would account for those models where the various other subgroups are considered to act in either series or parallel to control growth.

It is always difficult to prove any mathematical model to be the correct one for a particular system since, usually, experimental data are never extensive enough to rule out other alternatives. Also, the rate-controlling mechanism certainly can shift drastically with a change in experimental conditions such as supersaturation, agitation, impurity level, and temperature. No single system appears to have received sufficient detailed study so that definitive conclusions can be drawn.

Nucleation theories. To place the nucleation models in perspective, there are two limiting cases. First, there is the simple model that assumes the entire growth process is controlled by the rate of nucleation of a critical size nucleus, subsequent layer growth being infinitely rapid. At the other extreme, the simple polynuclear growth model assumes that each layer is composed of nonoverlapping critical nuclei with essentially negligible step velocities. Obviously, there are all varieties of intermediate models.

One of the more difficult problems with all nucleation theories is the estimation of the actual rate of formation of critical sized nuclei. A distribution of embryos of various sizes exists on the surface with continuous growth and dissolution; a birth of a critical sized nucleus is, then, a relatively rare event. The estimation of embryo distribution functions has been the subject of several recent papers (*2D*, *9D*, *14D*, *15D*).

Also, the interaction of critical sized nuclei with growing steps is of continued interest and has been simulated on a computer by Bertocci (*1D*), and with empirical relations by Matz (*17D*). This is a very difficult area to study without invoking rather stringent (and sometimes unrealistic) assumptions.

Other nucleation studies have been published by Chernov and Trusov (*5D*) who concentrated on electrostatic effects, and by Hirth and Kenty (*12D*) who failed to observe the predicted classical effect of supersaturation when silver was deposited on rock salt. Finally, Voronkov (*31D*) discusses the rate of step motion as affected by one-dimensional nucleation. Presumably Voronkov is referring to the formation of kinks when he speaks of one-dimensional nuclei though, in his paper, he indicates that the density of one-dimensional nuclei increases with supersaturation and, in general, kink density (or spacing) is not ordinarily considered to be a function of this variable.

Mass transfer theories. No significant papers were published in the past year considering mass transfer as the rate limiting step (*25D*, *28D*, *32D*). This is reasonable since we can now predict rather accurately the rate of solute transfer from the bulk to a surface if the fluid dynamics are well characterized and the physical properties of the solution known. Exceptions might be taken in cases of well-agitated crystallizers with low particle-to-solution relative velocities—and also, in some cases, dissolution data do not agree with those predicted by mass transfer theories (*8D*).

Kinetic theories. Most kinetic theories are an offshoot of the dislocation (or spiral) growth theories so admirably developed by Burton, Cabrera, and Frank over 20 years ago. In fact, the so-called BCF theory has come to signify to many the simple equation

$$R = AS^2 \tanh(B/S) \qquad \text{(1-D)}$$

where R is the rate of growth normal to the surface, A and B are temperature dependent constants, and S is the supersaturation. Equation (1-D) fits many diverse systems surprisingly well.

The conceptual visualization of the dynamic processes of adsorption, surface diffusion, and kink incorporation is fascinating and studies in this general area are still of interest. Schwoebel (*23D*) considers the capture of atoms at the kinks, Treivus (*29D*) emphasizes the effect of low supersaturation, Chernov and Dukova (*4D*) examine the effect of morphological factors, and Temkin discusses step movement (*26D*).

Forces between molecules on a surface during growth have been considered by De Wette *et al.* (*7D*) and between steps by Schwoebel (*22D*). In the latter paper, the conclusions indicate a possible mechanism to explain multiple step heights—*i.e.*, adjacent steps behave as though they mutually attract or repel one another with the sign of the step-forces depending upon the capture probability for atoms approaching from either direction.

In other articles, the Kossel model for growth and evaporation was analyzed in detail for a [100] face (*27D*), and equilibrium crystal shapes were related to free energies (*19D*, *21D*).

Impurity effects are always difficult to treat analytically. Englert and Tompa (*10D*) have delineated the effect of impurities near an edge dislocation for metal growth, while others have been concerned with impurity-induced step oscillations (*24D*) and impurity transients (*11D*).

A number of papers have been concerned with melt growth theory and growth from nonaqueous mixtures (*3D*, *6D*, *13D*, *16D*, *18D*, *30D*).

A doctoral thesis by Ohara (*20D*) contains a summary of the available growth theories and a critical evaluation of both the mathematics involved and the assumptions utilized in the various methods.

Experimental and Theoretical Studies on Solution Growth

Kinetic studies for growth from pure solutions. A number of attempts for the derivation of a kinetic expression (growth rate as a function of supersaturation) for the growth of crystals from solution appeared also in this year's literature (*6E*, *8E*, *14E*, *16E*, *17E*, *31E*, *32E*, *47E*, *48E*). It seems however that the most important contributions of these papers lie in some general conclusions that can be drawn from them.

One such conclusion concerns the sudden and sometimes dramatic increase of the dependence of the growth rate on supersaturation, at high supersaturation values. Levina and Belyustin (*47E*) working with Seignette salt crystals obtained kinetic data, which at low supersaturation could be correlated by a linear equation of the form $R = aS - b$ (it is interesting to note that another work with the same salt crystals (*31E*) resulted in a different nonlinear equation), where R is the grow rate (length/time) and S the supersaturation. Above a certain S, the data started suddenly to deviate; the authors, however, found that the data

could still be correlated by a straight line of higher slope. Botsaris and Denk (*6E, 14E*) working with potassium alum crystals also pointed out that the growth rate data show a linear dependence on S at low S, but deviate drastically from the linear law at higher values of S.

There is evidently an agreement among various investigators for the linear region at low supersaturations. They attribute it to a dislocation growth mechanism. It appears, however, that in addition to this dislocation growth, another mechanism exists that may be operating at all supersaturations, but becomes important at high supersaturations. The above mentioned works with Seignette salt and alum crystals suggested two-dimensional nucleation as the additional mechanism. The former (*47E*) assumes that the two-dimensional nuclei are formed at the base of macroscopic steps; this leads to the disintegration of the macroscopic steps into elementary steps and somehow to higher growth rates. The latter study (*6E*) follows a phenomenological approach, pointing out that an equation based on a compound growth mechanism (dislocation growth plus mononuclear two-dimensional nucleation) can correlate both the linear data at low S and the nonlinear at high S.

The hypothesized above disintegration of the macroscopic steps into smaller steps was actually observed in a recent Soviet study. Dukova and Gavrilenko (*17E*) measured growth rates of β-methylnaphthalene crystallized from an alcoholic solution; at the same time they observed the morphology of the growing faces—*i.e.* the heights of the steps. They found that at low supersaturations, there is a tendency for fairly thick steps to predominate. These steps break up into thinner steps as S increases. At higher supersaturation the number of thick steps increases again. The growth rates, on the other hand, changed accordingly. They were linear with S, then they became nonlinear, and finally at higher supersaturations changed back to linear.

Nancollas and his group continuing their work on the kinetics of crystallization of 2-2 electrolytes, reported also a "growth surge" at high supersaturations. The growth of $SrSO_4$ (*8E*) and also $CaSO_4$ (*48E*) crystals followed an equation in which the rate is proportional to the square of supersaturation S. However, at high S the exponent was much higher, sometimes as high as 13. It is not clear, however, because of the technique used, whether this "growth surge" is indeed a growth phenomenon or is due to incipient secondary nucleation at higher supersaturations.

Doremus (*16E*) studied the crystallization of another slightly soluble salt, namely $BaSO_4$. He worked at higher supersaturations than previous investigators, and as a result he found an exponent of S (in the kinetic expression) that was much higher than those reported previously.

The fact that the growth rate varies much stronger with S at high S appeared also in a study of the dendritic growth of ammonium chloride crystals (*32E*).

A conclusion of practical importance from all the above works is that when in practice a power form kinetic expression is employed over a wide range of supersaturation, the exponent of supersaturation may not be the same for the whole range.

The effect of variables other than supersaturation on the growth rate of crystals was presented in a few papers. Kahlweit (*32E*) and Treivus (*83E*) studied the effect of temperature. Belyustin and Pavlova (*4E*) reported the effect of liquid velocity on the dissolution and growth of $Zn(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$ crystals. The dependence of the growth rate on the liquid velocity was on the 0.68 power. No attempt was made by the authors to compare the experimental mass transfer coefficient to those estimated by empirical correlations (for example, Froessling equation).

The role of diffusion in the growth of crystals from solution is discussed in two other papers (*24E, 39E*). The former presents also experimental data on the concentration distribution in the liquid surrounding the crystal, and points out a quantitative disagreement between these data and theoretical data calculated from Fick's law.

Crystals for which kinetic data for their growth from aqueous solutions were reported included: sodium chloride (*91E*), potassium chloride (*78E*), aluminum fluoride (*77E*), barium nitrate (*34E*), dicalcium phosphate dihydrate (*51E*), and sucrose (*90E*).

Nielsen summarized a number of his ideas and data on crystal growth kinetics in a review article (*64E*).

Effect of impurities or additives on growth rate. As in the past, very few studies were reported last year where the emphasis was on the quantitative effect of impurities on the absolute growth rate of specific crystal faces. The most important probably is that of Michaels, Brian, and Beck (*56E*) on the effect of surface active agents on adipic acid crystals. Although the results in general were comparable to those found by Michaels and his coworkers in previous studies (see 1969 Review), the continuous observation technique employed in this recent study allowed certain previously unobserved growth characteristics to be distinguished. For instance, they report what probably constitutes the first well-documented case of growth enhancement by an additive. They also observed a growth instability at certain impurity concentrations and supersaturations; that is, the growth rate of crystals growing under these conditions would become unstable, dropping discontinuously to a rate lower by an order of magnitude. This type of instability was invoked in another study to explain data on the growth of alum crystals (*6E*).

Quantitative data ($NaClO_3$ crystals–SO_4— impurity) were also reported by Bliznakov and Kirkova (*5E*). Again the data were correlated by the Bliznakov equation discussed in the 1969 Review. This equation is based on the assumption of a Langmuir-type adsorption of impurities on the growing crystal face. Marshall and Nancollas (*51E*) also analyzed their results on the effect of sodium pyrophosphate on the growth of dicalcium phosphate dihydrate crystals rather successfully in terms of a Langmuir-type adsorption isotherm. The significance of their work, however, is diminished because their technique records mass deposition rates rather than the more meaningful individual face growth rates.

Kern in a very interesting article (*33E*) summarizing his ideas on the relation between growth kinetics and adsorption of impurities, pointed out that an adsorbed impurity may not always exist in the form of separate molecules on the crystal surface. In certain systems the adsorbed species may interact significantly and form an adsorbed two-dimensional aggregate. In that case the adsorption isotherm used predicts the existence of a critical temperature below which the formation of these aggregates takes place.

The existence of such a critical temperature was demonstrated by Portnov *et al.* (*71E*) in their study of the crystallization of Rochelle salt in the presence of Direct Blue Dye. A plot of the growth rate in the presence of the dye *vs.* temperature shows a sharp drop at 17°C, while no such anomaly is observed in the absence of the dye. The amount of dye incorporated into the crystal also increased sharply at 17°C.

The question of the selectivity of the impurities or additives was considered by Panov *et al.* (*66E*). In an unsuccessful attempt to establish a correlation between the chemical composition and the effect of an additive, they studied the effect of a large number of surface active agents on 18 crystallizing systems.

Other papers dealt with the mechanism of the impurity action. MacGillavry (*50E*) explained it in terms of a complex formation of the impurity with the crystal and the water on specific crystal planes. Glasner (*21E*) speculated that impurities affect the formation of the subcritical nuclei, which according to his hypothesis (see 1969 Review) are depositing on the crystal surface and make the crystal grow. An apparently similar model for growth and impurity (more specifically surface active agents) effect was also advanced in a recent Soviet paper (*67E*).

A novel way for studying the effect of additives was employed by Hartman (*27E*). It is known that NaCl whiskers can be formed in supersaturated NaCl solutions to which either polyvinyl alcohol or cellophane has been added. Hartman studied the effect of a series of metal-ion impurities on NaCl crystallization by observing their effect on the formation of whiskers in the above solution. He explained the results in terms of the Sears mechanism (see 1969 Review).

The crystallization of calcium sulfate is of practical importance since it is involved in the scale formation in boilers and also in the efficiency of phosphoric acid production by the action of sulfuric acid on phosphates. It is not surprising, therefore, that a large number of additives have been tested for their effect on $CaSO_4$ crystallization (*35E, 41E*). In the latter paper an increase in $CaSO_4$ growth rate is claimed with increasing concentration of calcium acetate and formate, and pentaerythritol. It appears, however, that this is not a true growth acceleration case. Since a rather high concentration of additives was used, it is possible that the additives affected the solubility and therefore the supersaturation of the $CaSO_4$ solutions.

Other practical systems for which the effect of additives was investigated include medicines (sulfanilamide, phenacetin) (*70E*) and lactose (*85E*).

In crystallization from solutions, the solvent is viewed by certain authors as a type of impurity. Two recent papers dealt with solvent effects on certain systems (*44E, 75E*).

TABLE I-E. STUDIES ON THE COCRYSTALLIZATION OF ISOMORPHOUS SALTS

Crystallizing System	*Reference*
$Ba(NO_3)_2$–$Pb(NO_3)_2$	(*62E, 63E*)
$Ni(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$–$Mg(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$	(*62E, 63E*)
$AlK(SO_4)_2 \cdot 12\ H_2O$—$CrK(SO_4)_2 \cdot 12\ H_2O$	(*93E*)
$Al(NH_4)(SO_4)_2 \cdot 12\ H_2O$–$AlK(SO_4)_2 \cdot 12\ H_2O$	(*93E*)
$Zn(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$–$Ni(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$	(*93E*)
$Zn(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$–$Co(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$	(*93E*)
$ZnK_2(SO_4)_2 \cdot 6\ H_2O$–$NiK_2(SO_4)_2 \cdot 6\ H_2O$	(*92E, 93E*)
K_2SO_4–K_2CrO_4	(*93E*)
$MgSO_4 \cdot 7\ H_2O$–$NiSO_4 \cdot 7\ H_2O$	(*42E, 46E*)
$CoSO_4$–$NiSO_4$–H_2O	(*46E*)
$MgSO_4$–$CoSO_4$–H_2O	(*46E*)
$MgSO_4$–$FeSO_4$–H_2O	(*46E*)

A serious problem in crystallization experiments is the unintentional introduction of impurities either by the solvent or the dissolved material used for the preparation of the supersaturated solution, or even from the walls of the container. Campbell and Nancollas (*8E*) offered some striking examples of the effect of trace impurities (introduced in the deionized water by the ion-exchange resin matrix) on the growth rate of the system they were studying. Punin and Petrov (*72E*) on the other hand, traced the nonreproducibility of the results they obtained in crystallization experiments with various batches of "reagent grade" KCl, to a strongly surface-active compound, which was always present in the KCl reagent. This compound, a primary aliphatic amine, is contained in the commercial grade KCl. When, however, a reagent of any grade is produced from this commercial grade no special measures are usually taken to remove this amine. Raksany and Voszka (*74E*) compared various methods for the preparation of extremely pure NaCl by recrystallization from solution and concluded that the most effective method involved crystallization from solution in a Teflon vessel.

Impurity incorporation. Many of the investigators in this area are distinguishing two different types of impurity incorporation. In the first type, the distribution of the impurity between the aqueous and the solid phases follows the relation (Berthelot–Nerst or *D*-law)

$$\frac{x}{y} = D\,\frac{a - x}{b - y}$$

while in the second type it follows the relation (Doerner and Hoskins or λ-law)

$$\ln \frac{a}{a - x} = \lambda \ln \frac{b}{b - y}$$

where a and b are the initial concentrations of the two salts (*i.e.*, solute and impurity) in the solution, x and y the amount of each salt crystallized, and D and λ the distribution coefficients.

Murthy and Mahadevappa (*62E, 63E*) worked with the problem of fractional crystallization of two isomorphous salts of different molecular weights. They concluded that both the *D*- and the λ-laws were not applicable to their system, probably because of nonequilibrium conditions. Their experimental results agreed, however, quantitatively with the predictions of the Abu Elamayem equation for the fractional crystallization of isomorphous salts; this equation applies to both equilibrium and nonequilibrium systems. On the other hand, in another paper (*93E*), it was reported that the *D*-law holds over a wide range of concentrations and supersaturations (even under nonequilibrium conditions) for the isomorphic systems studied. In the same paper, the relation between the growth rate and the chemical nature of a given crystal face, and the inclusion of the impurity along that face was also discussed. The above and other investigations on isomorphous salts (or impurities) coprecipitation are summarized in Table I-E.

Strictly speaking the above investigatons deal with the cocrystallization of a pair of isomorphous compounds. The distinction between crystallizing solute and impurity is rather artificial. In other systems, however, the distinction between the crystallizing species (macrocomponent or host lattice) and impurity (microcomponent) is real. A large number of studies dealt with this type of systems (see Table II-E) without succeeding, however, in illuminating the mechanism of this important type of impurity incorporation.

Techniques for study of crystal growth and for growth of single crystals from solution. Interferometric techniques for measuring solute concentrations and gradients in the neighborhood of a growing crystal face continued to be used by investigators (*24E, 80E*). A tracer technique was used by Bovington and Jones (*7E*) to study the kinetics of dissolution of barium sulfate, while a cinematographic technique for measuring the growth of single crystals of adipic acid was employed very successfully by Michaels *et al.* (*56E*) (see also section on Effect of Impurities on Growth Rate). An apparatus consisting of small cells for the observation of the crystallization of proteins is described by Dunnill (*18E*).

Techniques for the production of single crystals of desired size and form from solutions have been reported by various authors. A report from the Air Force Cambridge Research Labs presents a method for producing crystals of poorly soluble materials by diffusion growth (*45E*), while another report from the same laboratory describes evaporation and suspension growth methods and lists crystallizing systems that give beautiful colored crystals (*15E*). Fischinger (*20E*) suggests to students who would like to grow large single crystals that instead of tying a seed crystal to a string and suspending it in a saturated solution, they attach the seed to a flat polyethylene disk that can provide a convenient floating canopy that supports the immersed and downward-pointing crystal.

A method for growing plate-like crystals with a predetermined thickness was suggested by Collan (*12E*). Small seed crystals are inserted between two glass plates, the separation of which is fixed by spacers; the plates are then submerged into the supersaturated solution.

A recent patent (*10E*) describes the use of ion-exchange membranes for the growth of single crystals. The large-scale manufacture of single crystals from solution is the topic of three Czech publications (*3E, 43E, 52E*).

Studies dealing with the preparation of single crystals of particular compounds are listed in Table III-E.

Miscellaneous. A number of papers dealing with miscellaneous other aspects of growth from solution—like epitaxial growth (*9E, 28E, 30E*), effect of ultrasounds (*26E, 57E, 86E*), effect of gamma-irradiation (*61E*) and others—have been included in the list of references without being discussed here. These papers can be easily recognized since the title of each reference has been included in the list.

Experimental and Theoretical Studies of Crystallization from the Melt

An outline of our present understanding of the nature of crystallization from the melt was presented by Southin and Chadwick

TABLE II-E. STUDIES ON THE INCORPORATION OF IMPURITIES IN CRYSTALS GROWN FROM SOLUTION

Crystallizing Species (Macrocomponent)	*Impurities (Microcomponent)*	*Reference*
Metasilicate nonahydrate	Ca^{2+}	(*2E*)
Ammonium paratungstate	Cu^{2+}, Fe^{3+}, Co^{2+}, Mn^{2+}	(*25E*)
NH_4Cl	Fe^{3+}	(*40E*)
KCl	Rb^+	(*49E*)
KCl	Pb^{2+}	(*22E, 23E*)
KCl	Cd^{2+}	(*22E*)
NaCl	Pb^{2+}, Cd^{2+}	(*22E*)
NaCl	Ag^+	(*54E*)
$Pb(NO_3)_2$	Ba^{2+}	(*73E*)
$SrHPO_4$–$BaHPO_4$ (Solid Solution)	Ca^{2+}	(*58E*)
$Cs[I(IBr)]$	Rb^+	(*69E*)

TABLE III-E. PREPARATION OF SINGLE CRYSTALS FROM SOLUTION

System	*Reference*
AgI	(*59E*)
Alums	(*15E*)
$CaCO_3$	(*10E*)
$BaSO_4$	(*87E*)
KH_2PO_4	(*89E*)
$Mg(OH)_2$	(*82E*)
Hexamethylene tetramine	(*81E*)
Triglycine sulfate	(*43E, 55E*)
Thiourea	(*55E*)

TABLE I-F. EFFECTIVE DISTRIBUTION COEFFICIENTS

System	*Reference*
Se in GaSb	(*8F*)
Ag, Cu, Pb, Fe, Al, Si, Sn, Mg, Bi in Te	(*14F*)
In, Pb, Sb in Sn	(*15F*)
Ge in Ni	(*37F*)
Si, Al in Fe	(*29F*)
NH_4F in Ice	(*28F*)
Ca, Sr in NaI	(*10M*)

(*8Fa*) who emphasized the importance of the relationship between the solid–liquid interface shape and the entropy of fusion of each material. Tiller (*12Fa*) suggested that fundamental research on crystallization has produced a sufficient scientific base so that the crystallization field is rapidly becoming an engineering science. He cites single crystal growth, materials purification, and structure control of ingots and casting as areas where technological applications have occurred. Recent fundamental work in crystal growth from the melt has concerned itself with the interaction of interfacial kinetics and the transport of heat and material in the bulk phases, which has led to investigations of morphological instability and dendritic growth, of eutectic growth, and of solute or impurity distribution during solidification.

Interfacial kinetics of melt growth. There have been relatively few papers published in the past year dealing either theoretically or experimentally with the interface attachment kinetics of crystal growth from the melt. Chernov (*3Fa*) obtained an expression for the anisotropy of the crystal growth velocity by an analysis of morphological and kinetic data. In another paper (*2Fa*) Chernov reviewed his theoretical work on the attachment kinetics for solid solutions including a computer simulation of the growth of a binary crystal exhibiting an order–disorder transition. A theory of diffusionless crystal growth that occurs when the ratio of the diffusivity to the growth velocity is of the order of the interatomic distance was presented by Temkin (*9Fa, 10Fa*).

A new technique for employing a thermoelectric probe for measuring solid–liquid interface velocities and temperatures was introduced (*41F*), but very little new experimental data on the interfacial kinetics of pure materials have appeared in the past year. The growth kinetics of gallium were measured over a wide temperature range and the results compared to a number of crystal growth theories (*2F*). Another study (*22F*) found that the growth rates of salol and betol at low undercoolings followed a spiral growth mechanism. Hobbs and Ketcham (*11F*) discussed the growth of ice in slightly supercooled water. In another paper (*1F*) it was found that an acoustical field had no effect on the solidification rate of mercury. The kinetics of crystallization of a number of glasses were investigated (*5F, 7F, 9F, 21F, 38F*), and one paper (*11Fa*) dealt theoretically with the kinetics of bulk crystallization of glasses. DeLuca, Eagan, and Bergeron (*5F, 7F*) extensively investigated the relationship between crystal growth rate and viscosity in the system $PbO–B_2O_3$.

Electrical charge distribution during crystallization was considered in two papers. Cobb and Gross (*3F*) measured charge separation across the phase boundary during the freezing of ice

TABLE I-G. MATERIALS PRODUCED BY EPITAXY OR THIN FILM GROWTH

Element or Compound Deposited	*Reference*
Ag	(*116Ga, 145Ga, 167Ga, 171Ga, 195Ga*)
on Ag	(*26Ga*)
(Ag,Bi) on rock salt	(*102Ga*)
on Alkali metal halides	(*131Ga, 158Ga*)
AgCl	(*42Ga*)
Al	(*94Ga, 103Ga, 133Ga*)
Al_2O_3	(*53Ga, 101Ga, 108Ga, 138Ga*)
Alumino-silicate	(*174Ga*)
Au	(*33Ga, 82Ga, 109Ga, 113Ga, 145Ga, 161Ga, 171Ga, 183Ga*)
Au–SiO Cermet	(*125Ga*)
Bi	(*78Ga*)
Ba Aluminate	(*17Ga*)
$BaTiO_3$	(*148Ga*)
$(Ba–Sr)TiO_3$	(*107Ga*)
C	(*55Ga*)
CaF_2	(*179Ga*)
Cd on Sb	(*51Ga*)
$Cd_xPb_{1-x}S$	(*194Ga*)
CdS	(*6Ga, 40Ga, 93Ga, 110Ga, 112Ga, 187Ga*)
Cu	(*114Ga*)
Fe	(*77Ga, 105Ga, 159Ga, 184Ga*)
Fe_2O_3 on MgO	(*16Ga*)
FCC Metals	(*61Ga, 70Ga, 132Ga*)
Fused glass films	(*46Ga*)
Ga	(*182Ga*)
(Ga–V) Compounds	(*99Ga*)
GaAs	(*8Ga, 11Ga, 14Ga, 25Ga, 54Ga, 74Ga, 100Ga, 111Ga, 127Ga, 143Ga, 156Ga, 190Ga, 191Ga, 196Ga, 197Ga*)
$GaAs_xP_{1-x}$	(*11Ga, 27Ga, 157Ga*)
$GaAs_{1-x}Sb_x$	(*36Ga*)
GaN	(*85Ga*)
GaP	(*11Ga, 92Ga, 162Ga*)
Ge	(*4Ga, 5Ga, 47Ga, 54Ga, 67Ga, 79Ga, 84Ga, 106Ga, 164Ga, 193Ga*)
$Ge(Ga_xIn_{1-x})$	(*86Ga*)
βHgS	(*160Ga*)
In	(*20Ga, 122Ga*)
InAs	(*88Ga*)
InSb	(*35Ga, 41Ga*)
In_2O_3	(*138Ga*)
MgF_2	(*134Ga*)
$MnFe_2O_4$	(*97Ga*)
$NaNO_3$	(*130Ga*)
NbZr, Nb–Zr–MO	(*149Ga*)
Ni	(*43Ga, 172Ga, 173Ga*)
(Ni,Zn) ferrite	(*73Ga*)
$NiCl_2$	(*75Ga*)
NiO	(*139Ga*)
Oxide films (various)	(*7Ga, 38Ga, 49Ga, 72Ga, 138Ga, 188Ga*)
Pb on Cu	(*142Ga*)
PbI_2	(*68Ga*)
$Pb_{1-x}Sn_x$ Chalcogenides	(*28Ga*)
$Pb_{1-x}Sn_xTe$	(*52Ga*)
PbS	(*151Ga, 198Ga*)
PbTe	(*168Ga, 198Ga*)
$Pb_{1-x}Ln_xTe$ (Ln = Eu,Yb)	(*169Ga*)
PbSe	(*198Ga*)
RbCl on oxides	(*3Ga*)
Rutite	(*59Ga*)
$SbSe_3$	(*48Ga*)
Sb_2Te_3	(*57Ga*)
Si	(*10Ga, 39Ga, 50Ga, 56Ga, 65Ga, 90Ga, 91Ga, 95Ga, 106Ga, 137Ga, 140Ga, 176Ga*)
(Si,Cr)	(*181Ga*)
$SiC(\alpha$ & $\beta)$	(*12Ga, 13Ga, 24Ga, 66Ga, 81Ga, 135Ga, 178Ga*)
$Si–Si_3N_4$	(*31Ga*)
Si_3N_4	(*2Ga, 30Ga, 31Ga*)
SiO	(*9Ga*)
SiO_2	(*37Ga, 170Ga*)
Sn	(*158Ga*)
SnO_2	(*49Ga*)
SnTe	(*198Ga*)
Sulfide films	(*72Ga*)
Te	(*146Ga*)
Ti	(*185Ga*)
TiO, TiN	(*69Ga*)
Th on W(100)	(*124Ga*)
βW	(*80Ga*)
$YCl_3 \cdot 6\ H_2O$	(*49Ga*)
(Zn,Cd)S	(*19Ga*)
ZnSe	(*54Ga*)
ZnO	(*128Ga, 138Ga*)
ZnS	(*134Ga, 177Ga, 192Ga*)
ZnTe	(*186Ga*)
on Zr	(*38Ga*)

from dilute (10^{-6}–10^{-3} M) ionic solutions. Mel'nikova (*23F*) reviewed the separation of charge during the crystallization of both dielectrics and aqueous solutions containing ionic impurities.

Morphological stability and dendritic growth. In contrast to the previous year theoretical analyses of morphological instability have abated somewhat. Sekerka (*7Fa*) reviewed the general phenomenon and theory of morphological stability and particularly discussed the paper of Hardy and Coriell (*10F*) who observed instabilities during the growth of ice cylinders. The growth rate and wavelength of the perturbations on the interface were in agreement with the predictions of stability theory. Hurle (*4Fa*) considered interfacial stability during the solidification of a stirred binary alloy. Brice (*1Fa*) analyzed the effect of interface kinetics on the stability of the growth interface of a pure crystal and found an anomolous effect of the kinetics under certain circumstances.

Sharp and Hellawell (*35F, 36F*) observed solute distributions at planar, cellular, and dendritic interfaces during steady state and transient growth of Al–Ag alloys. They found the dendritic spacings to be inversely related to the growth velocity and also concluded that interfaces adjust their shape to minimize constitutional supercooling. Morris and Winegard (*26F*) observed the development of a cellular interface in a Pb–Sb alloy and noted that breakdown occurs at a lower velocity when defects are present at the interface. In a later study (*27F*) on the same alloy system, they observed the cellular–dendrite transition. Three papers (*16F, 24F, 32F*) dealt with the dendritic growth of ice from pure water, fluoride solutions, and brines.

Eutectic solidification. Interest continues in the mechanism and control of eutectic microstructures. Durand (*6F*) reviewed theories of eutectic crystallization with particular emphasis on the Al–Si system, while Hurle and Hunt (*12F*) observed the structure of directionally solidified semiconductor eutectics in the InSb–NiSb system. Jaffrey and Chadwick (*13F*) reported experimental observations of the effects of solid/liquid interface contours, growth variables, and impurity content on the lamellar and fibrous eutectic morphologies in the Sn–Zn system. In another paper (*19F*), a number of eutectic alloys were solidified at velocities up to 0.2 cm/sec and in all cases the lamellar spacing was found to be inversely proportional to the velocity to the 1/2 power. Rastogi and Rastogi (*31F*) examined the eutectic structure of a number of binary organic systems, and Wilcox (*39F*) obtained the formation of an oriented composite structure by directed solidification in an organic system containing a strong fiber-forming component. Other papers dealt with the control of microstructure with a magnetic field (*33F*) and by gentle scraping of the interface during solidification (*30F*).

Transport processes in the melt. Transport of heat and mass in the melt during solidification may occur by either convection or diffusion. One paper (*18F*) mathematically analyzes the influence of solute buildup on crystallization of binary mixtures by unidirectional cooling. They particularly noted and experimentally confirmed the importance of bulk motion of the liquid caused by diffusion of solute from the crystal surface. In the usual case, because of the complexity of the transport processes when convection is present, the mass transfer is described by an effective distribution coefficient that relates solid composition to bulk liquid composition. Table I-F lists systems in which effective distribution coefficients have been reported in the past year. Such coefficients are dependent not only on the thermodynamic properties of the system, but operating variables such as temperature gradient and geometry as well. Hurle and Jakeman (*5Fa*) have shown that temperature fluctuations during the measurement of distribution coefficients by directional solidification tend to decrease the average degree of solidification.

Experimental and Theoretical Studies of Crystallization from the Vapor

Thin films and epitaxy. Two recent symposia, The International Conference on Thin Films (Boston, 1969) and the National Symposium of the Thin Film and Surface Science Division of the American Vacuum Society (Seattle, 1969), are the source of most of the literature published in the field of epitaxy and thin film growth over the past year. Their proceedings are published in the *Journal of Vacuum Science Technology*, **6** (4), (1969), and **7** (1), (1970), respectively. The large number of papers presented is indicative of continued research interest in the technology of epitaxy and thin film growth. The vast majority of people working in this field are interested primarily in producing crystals that have desirable semiconducting or optical properties.

TABLE II-G. GENERAL TOPICS IN EPITAXY AND THIN FILM GROWTH

Subject	*Reference*
Contact and Interconnection Technology	(*96Ga*)
Effect of Electrical Relief Structures	(*42Ga*)
Effect of Ion Bombardment Cleaning	(*155Ga*)
Epitaxial Growth Apparatus and Techniques	(*10Ga, 25Ga, 50Ga, 133Ga, 141Ga, 144Ga, 149Ga, 175Ga, 198Ga*)
Epitaxial Growth Mechanisms	(*58Ga, 59Ga, 139Ga*)
Film Formation and Structure Sensitive Properties	(*15Ga, 29Ga, 34Ga, 87Ga, 89Ga, 104Ga, 117Ga, 118Ga, 119Ga, 123Ga, 126Ga, 129Ga, 150Ga, 152Ga, 153Ga, 154Ga, 166Ga, 175Ga, 180Ga, 198Ga*)
Gunn Effect Devices	(*14Ga*)
Initial Stage of Growth	(*8Ga, 32Ga, 64Ga*)
Influence of Impurities	(*61Ga, 65Ga*)
Lattice Misfit	(*62Ga*)
Microzone Recrystallization	(*18Ga*)
Noncrystalline films—Preparation and Properties	(*98Ga, 180Ga*)
Nonsingular Surfaces	(*44Ga, 45Ga*)
Polytypism	(*68Ga*)
Positive Coefficient for Thin Thermistor Films	(*107Ga*)
Role of Surface Migration	(*67Ga, 140Ga, 166Ga*)
Surface Structure Studies	(*166Ga, 151Ga, 132Ga, 131Ga, 34Ga, 70Ga, 76Ga, 78Ga, 180Ga, 44Ga, 45Ga, 86Ga, 104Ga, 105Ga, 123Ga*)
Temperature Control	(*1Ga*)
Theory of Condensation and Nucleation Processes	(*21Ga, 63Ga, 83Ga*)
Theory of Thin Platelet Growth	(*23Ga*)
Thin Film Devices	(*88Ga, 115Ga, 121Ga, 141Ga, 189Ga*)
Uniformity of Deposition	(*22Ga, 60Ga, 120Ga, 126Ga, 147Ga, 163Ga*)
Vapor Deposit Replication	(*71Ga*)

Basically, epitaxy or overgrowth consists of depositing a layer of crystalline material on a well-defined crystallographic surface of a substrate. The orientation and structure of the final layer is dependent on the nature of the substrate as well as the deposited atoms. Table I-G classifies the references according to particular compounds or elements, which are deposited either as epitaxial layers or thin films. Table II-G lists some of the general topics pertaining to the field.

An annotated bibliography on semiconductors (*150Ga*) was presented in a U.S. Government Report. Runyan (*137Ga*) and LaChapelle *et al.* (*91Ga*) reviewed silicon deposition technology and recorded over 250 references in that area. Runyan (*137Ga*) concluded that despite the numerous unsolved problems and the importance of Si deposition technology to the electronics industry, the field is showing signs of stagnation. Zamel (*198Ga*) presented several ideas on general methods employed in the production of epitaxially deposited compounds, *e.g.*, PbS, PbSe, PbTe, SnTe.

In addition, a number of investigators (*8Ga, 32Ga, 64Ga*) described the initial stage of epitaxial deposition, including the importance of morphology and surface structure on the growth process. Chapman and Jordan (*32Ga*) introduced the fractional nucleation rate concept, which is useful in determining values for the energy parameters intrinsic to nucleation and growth. Aleksandrov (*8Ga*) analyzed 2- and 3-dimensional nucleation theory in relation to surface purity, shape, orientation, and gas supersaturation. Several investigators (*22Ga, 60Ga, 120Ga, 147Ga, 163Ga*) treated the concept of uniformity of film thickness making comparisons between theoretical predictions and experimental values.

Kenty and Hirth (*83Ga*) considered epitaxy and heterogeneous nucleation from a theoretical standpoint. Relative nucleation rates were determined for "epitaxial and random" cap shaped nuclei, which have different values for the nucleus–substrate interfacial free energy; impurity effects were discussed; and comparisons made with available experimental data. Several other papers (*15Ga 29Ga, 58Ga, 63Ga*) were also concerned with the kinetics, structure, and mechanistic aspects of epitaxy.

Mayer (*104Ga*) investigated the first period of layer formation on amorphous substrates by microbalance techniques in ultrahigh vacuum and showed that a phenomenon of delayed condensation existed, as well as a variation in the condensation coefficient from 0 to 1.0 as deposition continued. Braunstein and Kickuchi (*21Ga*) used the path probability technique to estimate nucleation rates.

Hanoka *et al.* (*68Ga*) presented direct experimental evidence for polytypism, postulating that the screw dislocation which produces higher polytypes resulted from epitaxial growth on a foreign nucleus. By electron microprobe investigation, they were able to show that Ag impurities, acting as foreign nuclei, were present in significant amounts in gel grown PbI_2 crystals; and that there was a possibility that PbI_2 grows on an AgI "coated" Ag particle rather than on a pure Ag particle.

Haidinger and Courvoisier (*67Ga*) discussed the role of surface migration in epitaxy by considering the importance of imperfections on Ge epitaxy over a wide range of supersaturations.

Physical vapor transport (homogeneous condensation). Work in this area has proceeded mainly on the experimental level. A number of patents appeared in the literature either describing specific methods for producing crystals CdS (*2Gb*) and SiC (*11Gb*), or general techniques of value such as thickness control by impurities (*3Gb*) and continuous sublimation (*6Gb*). Mamykin *et al.* (*9Gb*) discussed the effect of carbon on MgO crystal growth. Carbon apparently had a significant effect on the mechanism and kinetics of growth of filamentary MgO crystals. This was believed to be caused by precipitation of carbon, thus reducing the growth rate and giving rise to multidimensional overgrowth. Toyama and Sekiwa (*17Gb*) and Shiozawa *et al.* (*13Gb*) studied the growth of II–VI compounds including CdS, CdSe, and ZnTe.

Table III-G summarizes the work done on PVT by indicating the materials produced and the corresponding reference.

Chemical vapor transport and chemical vapor deposition. Chemical Vapor Transport (CVT) and Chemical Vapor Deposition (CVD) provide useful methods of growing crystals avoiding the high temperatures and/or pressures required in homogeneous transport (PVT). In CVT, the desired element (compound) is reacted in one isothermal section of the growth cell with iodine, chlorine, or similar carrier gas, *e.g.*, $2\ Ga + Cl_2 \rightarrow 2\ GaCl_2$; then the resultant vapor diffuses to a different section of the cell maintained at a different temperature, and is deposited on a suitable substrate by a shift in the thermodynamic equilibrium created by the temperature difference. In contrast, pure CVD requires a reaction but not necessarily gas transport, *e.g.*, $SiCl_4 + CCl_4 + 4\ H_2 \rightarrow SiC + 8\ HCl$ (*10Gd*). Obviously, combined CVT and CVD processes are quite commonly used and are sometimes advantageous in producing high quality crystals (*2Ge, 3Ge, 5Ge, 11Ge*). Additional examples are contained in the bibliography under Combined Processes.

Gibart (*6Gc*) treated the theoretical aspects of single crystal growth by CVT and gives some specific examples of crystals grown. These are listed in Table III-G along with other compounds and elements produced by CVD and/or CVT. Jona (*9Gc*) described research efforts aimed at improving the mechanistic understanding of CVT. Rustamov and Geidarov (*14Gc*) discussed mechanisms for $A^{III}B^{VI}$ and $A_2^{III}B_3^{VI}$ crystal growth. Wehmeier (*17Gc*) considered the thermodynamics of chemical transport of some ternary compounds including yttrium iron garnet.

CVD saw increased use in producing crystals by organometallic decomposition. Thomas (*15Gd*) grew GaP by reacting gallium triethyl and phosphorous triethyl at 485°C. Kaplan and d'Heurle (*5Gd*) deposited Mo and W by carbonyl decomposition. Gabor and Blocker (*3Gd*) used *in situ* transmission electron microscopy to observe the growth of Fe and Ni by carbonyl CVD.

Evaporation. Several general articles covering the entire field of evaporation phenomena are discussed. Comparisons between evaporation and condensation (growth) phenomena can provide some insight into the actual molecular processes that are occurring on the crystal surfaces. In the past, most of the work on evaporation was concentrated in the area of equilibrium vapor pressure measurements under Knudsen or similar conditions, or Langmuir free evaporation studies. Primarily, weight-loss and/or torsion-effusion studies were utilized in the measurements. In recent years, however, increased emphasis has been placed on the morphology of the crystal and how that relates to the qualitative and quantitative aspects of evaporation. For example, Budke (*1Gf*) considered the morphology of evaporating NaCl crystals in detail and showed that the etch pits are directly related to surface contamination and not emergent dislocations, in contrast to Somorjai's (*21Gf*) analysis for NaCl. Munir and Hirth (*13Gf*) discussed the morphology of sublimated CdS and ZnS and showed that it is at least qualitatively consistent with the terrace–ledge–kink (TLK) model for evaporation.

Das, Cocks, Wolff and their associates (*4Gf, 5Gf*) considered the

TABLE III-G. MATERIALS PRODUCED BY PHYSICAL VAPOR TRANSPORT, CHEMICAL VAPOR TRANSPORT, AND/OR CHEMICAL VAPOR DEPOSITION

Element or Compound Produced	*Reference*
Alum alloys	(*4Ge*)
Al alloys	(*4Gc*)
Anthracence	(*10Gb*)
As	(*4Gc*)
B–Si–C	(*8Gd*)
B_4C	(*7Gd*)
BP	(*10Gc*)
$CeFeO_3$	(*9Gd*)
CdS	(*13Gb, 16Gb, 1Gc, 2Gc, 11Gc, 7Ge, 8Ge, 17Ge*)
CdSe	(*13Gb*)
CdTe	(*11Ge*)
$(CH_2)_6N_4$	(*16Ge*)
CO_2	(*1Gb*)
$CoAs_2$	(*19Ge*)
$CoCr_2S_4$	(*15Ge*)
$CoCr_2Te_4$	(*6Gc*)
$Co_{.67-.80}Te$	(*6Ge*)
Cristobalite	(*18Ge*)
Cr alloys	(*14Gd*)
Cr_3Te_4	(*6Gc*)
$CuAlS_2$, $CuAlSe_2$	(*7Gc*)
CuO	(*18Gb*)
$CuNbO_3$, $CuTaO_3$	(*14Gb*)
Fe	(*3Gd*)
$FeCr_2S_4$	(*15Ge*)
Fe_2Te_3, FeS_2, FeS	(*6Gc*)
FeP, Fe_2P, $FeAs_2$	(*19Ge*)
Ga alloys	(*4Gc, 14Gc*)
GaAs	(*9Gc*)
GaP	(*15Gd, 11Ge*)
GaS	(*12Ge*)
GaSb	(*4Ge*)
Gd_2S_3	(*13Gc*)
Ge	(*9Gc*)
$HgCr_2Se_4$	(*15Gc*)
In	(*12Gb, 5Gc*)
La_2S_3	(*6Gc*)
MgO	(*9Gb*)
Mo	(*5Gd*)
MoClS	(*17Gd*)
MnS, MnSe	(*18Gc*)
Ni	(*3Gd*)
Oxy-halide phosphors	(*16Gd*)
Phosphides (various)	(*5Gc*)
α-Pu	(*8Gb*)
Sb	(*5Gc*)
Si	(*9Gc*)
SiC	(*4Gb, 11Gb, 15Gb, 6Gd, 10Gd, 10Ge, 13Ge*)
SnO_2	(*2Gd*)
TiC	(*13Gd*)
VF_2	(*11Gd*)
W	(*1Gd, 5Gd*)
Yttrium iron garnet	(*17Gc*)
Yttrium silicates	(*4Gd*)
ZnS	(*8Gc*)
ZnSe	(*17Gb, 12Gc, 16Gc*)
ZnC–P	(*2Ge*)
$ZnSiP_2$	(*3Ge*)
(Zn,Cd)S	(*7Ge*)

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evaporation of 2 ZnO → 2 Zn + O_2 in detail by combining an experimental program of microbalance measurements, optical and electron microscopy, and laser scattering, with a theoretical program of morphological habit prediction. They were able to correlate changes in morphology with evaporation rate changes and found different evaporation rates for (00.1) and the (00.$\bar{1}$) faces of ZnO. By a theoretical treatment of habit modification they constructed some general principles that could be used in predicting the morphology of noncentro-symmetric materials of chalcopyrite structure. Leonard and Searcy (*11Gf*) also studied ZnO evaporation and recorded their findings in a short note. Munir and his associates (*6Gf*, *12Gf–14Gf*) studied PbTe, CdS, and ZnS single crystals and presented a relatively complete picture of the interrelationships between morphology, activation energy, and rates of evaporation for those compounds.

Rosenblatt and his coworkers (*16Gf*, *17Gf*) continued to report their studies on arsenic and antimony evaporation. Arsenic and antimony are unusual because they undergo surface rearrangement reactions to form As_4 and Sb_4 tetrahedra, which are found almost exclusively in the vapor phase. This particular step is rate controlling and believed to be located at the kink site on a stepped arsenic or antimony surface; it causes an evaporation coefficient that can be considerably less than 1.0 [for As $\sim 10^{-5}$, for Sb $\sim 10^{-2}$]. Rosenblatt cites the importance of dislocations in determining the location of etch pits on the surface.

Winterbottom (*26Gf*) investigated evaporating single crystalline Ag. He found considerable morphological effects caused by adsorbed gases on the Ag surface and developed a model based on his kinetic measurements which showed that capillarity forces were probably responsible for the hill and valley surface roughness observed. The experimental orientation dependence of surface morphology and vaporization faceting illustrate the possibility that a cusped minimum might exist in the surface free energy at the (111) and (100) planes.

In summary, morphology studies incorporated with weight loss and other dynamic techniques including mass spectrometry are being used to more clearly characterize the mechanism for solid crystalline evaporation.

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