SOME EFFECTS OF SONICS ON THE DRYING OF CEREAL GRAINS

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ABSTRACT

SOME EFFECTS OF SONICS ON THE DRYING OF CEREAL GRAINS

by Charles C. Huxsoll

Drying has long been used as a means of preservation for chemical and biological materials. However, drying techniques are continually being sought which improve the process by one or a combination of the following factors: increasing the drying rate, increasing the quality of the dried product, decreasing the cost, or by making it possible to dry products which cannot be dried by present techniques.

The use of sonic energy has been suggested as a relatively new drying technique. It has been observed that when a moist body is placed in an intense sonic field it will dry. Such drying occurs in the absence of a marked increase in the temperature of the drying product. Thus, the process has practical value for drying products with heat sensitive constituents.

An experiment was designed to determine the effect of sound on the drying of wheat and corn grains. Samples were conditioned to 30% dry basis moisture and dried for various intervals of time at several temperatures in a rotary dryer. The dryer was equipped with a sonic generator which produced a sonic field of approximately 165 db. at about 11.5 K.C. All tests made in the presence of the sonic field were

compared to tests made under identical conditions in the absence of the sonic field. This provided a measure of the effect of the sound only.

The results indicated that the sonic energy substantially increased the drying rates of these materials. The time required to dry the material was reduced by up to 50% when the sound was applied.

The drying of wheat and corn grains can be described in terms of a first-order reaction. The rate constant was substantially increased by the application of sonic energy. However, the rate constant was less affected by the temperature during sonic drying compared to drying in the absence of the sonic field. Therefore, when Arrhenius plots were made sonic drying produced a lower activation energy than conventional drying. The activation energies for both sonic and conventional drying were less than the heat of vaporization of water indicating that surface diffusion was the probable mechanism of moisture transfer within the material. Sonic vibrations caused a breakdown of the Van der Waal forces within the liquid filament which accounts for the lower heat of activation and the increased drying rate.

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SOME EFFECTS OF SONICS ON THE DRYING OF CEREAL GRAINS

By

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INTRODUCTION

Drying refers to the removal of moisture from a wet material until the moisture content of the material reaches some critical level. When the moisture content of the material is at or below this critical value, deterioration of the material is practically negligible. For this reason, drying processes have long been used as means of preservation.

Recently the food industry has been making a great effort to increase the use of drying processes. The drying of food products not only makes possible their preservation, but it often reduces transportation costs and gives rise to instantized products as well. The recent efforts in the areas of freeze-drying, spray drying, and foam-mat drying are evidence of this increasing emphasis on drying in the food insutry.

The general criteria of a good drying procedure are that the process is rapid, does not adversely affect the product quality, and yields a product that is easily and rapidly rehydrated.

In conventional heated air drying processes, drying rates are increased by increasing the temperature of the drying air. However, this often increases the temperature of the drying product which may, in turn, adversely affect the product quality if certain of its constituents are heat sensitive.

One drying process which offers some possibility for increasing drying rates without substantially increasing the temperature of the drying product is sonic drying. It has been observed that when a moist material is placed in an intense sonic field the material will dry. This drying takes place without a marked increase in the temperature of the drying product. Thus, the process has practical value for the drying of products with heat sensitive constitutents.

REVIEW OF LITERATURE

The sonic drying process has been investigated to only a very limited extent. Burger and Sollner (1936) first considered the use of ultrasonics for drying. Later, in a U. S. patent, Stephanoff (1938) observed that when a moist material is placed in the shock wave region of an air jet exceeding the speed of sound the material will tend to dry. Stephanoff surmised that under such conditions a nearly perfect vacuum would be created on the downstream side of the particle and, regardless of the temperature, boiling would occur there. Vang (1942) suggested that the application of sonic vibrations in a dryer would enhance drying by creating an artificial turbulence. In general, these early experimenters did not achieve substantial success with sonic drying.

However, with the advent in the 1950's of the much more powerful sonic generators for creating high intensity airborne sounds a new phase in sonic-drying came about. Whereas the early experimentation was done with lower intensity and sound merely had a secondary effect on drying, the much more powerful generators made it possible to consider that the sound alone might effect the drying process.

Boucher (1959) reported on a series of pilot tests and established the following facts:

"1) Intensity level is the main factor governing the evaporation rate; a minimum of 145 lb. is required for industrial processing.

2) Thinner material layers dry quicker. The maximum thickness suggested today is 1-2 inches.

3) The optimum operative frequency range lies between 6-10 KC judged on results obtained to date."

Boucher proposed a theory to account for the drying effect of sonic vibrations. He noted that the rate of evaporation at a liquid-gas interface is governed by the following law:

$$\frac{dm}{dt} = KS \frac{(P-p)}{H} \qquad (1)$$

Where:

m = moisture content
t = time
P = saturation vapor pressure at the temperature of the liquid
p = vapor pressure in the surrounding atmosphere
H = gas pressure in the surrounding atmosphere
S = surface of the material
K = a coefficient strongly dependent on the gas turbulence above the interface.

Boucher then pointed out that the sonic field would cause a considerable increase in the coefficient K. He noted, as well, that experience dictates that in a sonic field the effect of expansion always predominates over the effect of compression, which also leads to an increase in the rate of removal of moisture from the drying material. In addition, the extent of the surface subjected to the sonic field would, according to equation (1), have a marked effect on the drying rate. For this reason, Boucher suggested that a rotary type dryer would be the most ideal drying apparatus for drying wet powders and granular materials in a sonic field. The investigations by Boucher also showed that there is no common point between drying with airborne sound and the heating process since the only materials to show a heating affect due to sound were certain types of mineral wools.

P. Greguss (1961) also noted that there is no common point between sonic drying and the heating process. He further considered the effect of sound on the moisture conductivity and pointed out that sound does not have only a surface effect as would be the case in the previous theory of Boucher. As evidence, Greguss noted that in the drying of certain sugars it is extremely difficult to remove the moisture content below 1-2% by any method utilizing turbulence. However, where drying in a sonic field, this moisture could be removed in about 15 minutes. Therefore the sonic energy affected the moisture migration within the material.

Greguss categorized the moisture in a wet material according to adsorption as osmotic, capillary, and polymolecular attached moisture. In the region of osmotic moisture the theory outlined by Boucher would be operative; however, to explain the additional beneficial effects of sound in the other regions, the theory must be amplified.

In conventional drying once the so-called osmotic moisture is removed the drying rate suddenly decreases as the capillary moisture must be removed. In this region Greguss considered that sonic energy would have a very beneficial effect since it would diminish the decrease in the drying rate which occurs in conventional drying. To explain this, Greguss noted that several phenomena occur which could have the effect of increasing the rate of removal of capillary-attached moisture. First, when water is subjected to intense sonic vibrations its viscosity decreases. This can be explained on the basis of the hole-theory of liquids. The sonic vibrations cause an increase in the number of holes and therefore the viscosity decreases. In a liquid, the diffusion coefficient is given by the equation:

$$D = u \bar{k} T \qquad (2)$$

where:

u = mobility of the particle under consideration
$\bar{k}$ = Boltzmann constant
T = absolute temperature

Assuming that the particles are spherical in shape and the medium in which they are moving is continuous, the mobility is given by Stokes' formula:

$$u = \frac{1}{6\pi \eta r} \qquad (3)$$

where:

η = viscosity of the liquid medium
r = radius of the moving particle

Therefore,

$$D = \frac{\bar{k}T}{6\pi \eta r} \qquad (4)$$

Thus, the diffusion coefficient is inversely proportioned to the viscosity, and the application of an intense sound field can therefore cause an increase of moisture diffusion.

In addition, the cavitation caused by sonic vibrations could lead to the formation of small vapor or air bubbles within the capillaries. The work done on these bubbles by the sonic vibrations could cause them to expand and force the water from the capillary. The radiation pressure, which is a unidirectional pressure which arises in intense airborne sonic fields, was also suggested by Greguss as a contributing factor in the enhancement of drying. This pressure could lead to a pressure drop across the capillary which would tend to move the water filament toward the surface.

In the region in which the moisture is held by polymolecular adsorption, the drying rate is determined by the internal diffusiveness of the moisture. Greguss suggested that sonics would affect the moisture diffusiveness in this region in the same way as it affected diffusiveness in the region of capillary moisture.

Thus, while several theories have been proposed to explain the enhancement of drying due to sonic irradiation, no quantitative results have been reported which will substantiate these theories.

Many of the biological materials which are dried contain a major portion of their moisture in the region of polymolecular adsorbed water. Drying studies on materials such as wheat and corn grains have been extensive, and the moisture which is removed during most drying processes is entirely in the polymolecular adsorbed region. Adsorption studies have also been made on these materials, and the relative energy levels of the adsorbed water molecules are approximately known.

Becker and Sallans (1956) reported on the desorption isotherms of wheat. They observed that the desorption isotherm represented by a plot of equilibrium moisture content versus the relative vapor pressure of the surrounding atmosphere at constant temperature, could be divided into three regions. In the region corresponding to relative vapor pressures less than 0.35 the desorption isotherm could be described by an equation developed by Brunauer, Emmett, and Teller (1938), usually referred to as the B.E.T. equation. This equation is based on the assumption that the same forces that produce condensation are chiefly responsible for the binding energy of multimolecular adsorption. Many biological materials show a behavior described by this equation in the region of low relative vapor pressures.

In the region of relative vapor pressures between 0.35 and 0.50 the equilibrium moisture content displayed a linear variation with the relative vapor pressure. As the relative vapor pressure increased from 0.50 to 0.95 the moisture content showed a variation described by the Smith equation (1947).

By applying a form of the Clausius-Clapeyron equation to these water desorption isotherm equations Becker and Sallans were able to calculate the heat of desorption for various moisture levels and thus establish relative energy levels of the adsorbed water molecules.

A similar study by Rodriguez et al., (1963) on shelled corn showed that the Clausius-Clapeyron equation and the Othmer equation, which is a form of the Clausius-Clapeyron equation, could be used to evaluate the heat of vaporization for shelled corn. The B.E.T. equation was found applicable at moisture levels below 9%. The heat of desorption for shelled corn followed the same trend as that for wheat as the moisture content varied.

While the water sorption isotherms have been described or can be described relatively easily for most biological materials, the basic physical mechanism of desorption or drying is not clearly understood. The general trend in drying research has been to attribute all of the moisture removal to liquid diffusion. While it appears plausible that a major portion of the drying results from liquid diffusion, the assumptions which must be made in order to obtain working solutions of the diffusion equations, such as Fick's second law, seem rather gross for most biological materials.

Jason (1958) has reported on the drying of fish muscle. He observed that when the free moisture content was plotted versus time on semi-logarithmic coordinates two straight line regions appeared. If the free moisture content during each phase is divided by the initial free moisture content for each phase this ratio, called the free moisture ratio, also exhibits a straight line relationship with time when plotted on semi-logarithmic coordinates. Jason denoted the reciprocals of the slopes of these latter curves as "time-constants", and used them to characterize the drying curves. By treating the fish muscle as the shape of a brick Jason also showed that the solution to Fick's law could be written as:

$$\frac{Wt - We}{Wo - We} = (8/\pi^2) \quad \exp\left[\frac{\pi^2}{4} \left(\frac{D_x}{a^2} + \frac{D_y}{b^2} + \frac{D_z}{c^2}\right) t\right] \quad (5)$$

where a, b, c are the half-thicknesses of the slab in the directions of x, y, z, and Dx, Dy, and Dz are the corresponding diffusion coefficients.

By further assuming that the material was isotropic, it was possible to set the slope of the curve or the reciprocal of the time constant equal to: $\frac{-\pi^2}{4}(a^{-2} + b^{-2} + c^{-2})$ or

$$D = \frac{-4}{\pi^2 \tau}(a^{-2} + b^{-2} + c^{-2}) \quad (6)$$

A plot of D versus the reciprocal of the absolute temperature yielded a straight line on semi-logarithmic coordinates, indicating that an Arrhenius type of relationship existed.

Similar analysis have been made on the drying of wheat. Becker and Sallans (1955) based an analysis on Fick's law assuming that the wheat kernels were spherical in shape and that the moisture diffusion coefficient remains constant with position and moisture concentration over the range of practical drying. These researchers also deduced that the surface moisture content remains constant while the average moisture content remains between about 15-30% (dry basis). A later paper by Becker (1959) removed the assumption that the drying wheat kernels were of spherical shape. In the latter work, high vacuum drying was utilized and the surface moisture content varied slightly with temperature and was called the dynamic surface moisture content. These researchers also attempted to measure the temperature of the drying grains. Within the accuracy of their measurements they found that the surface temperature of the grain was practically identical to the internal grain temperatures and that the grain temperature asymptotically came to within two degrees of the drying air temperature after about eight minutes.

Earlier work on the drying of whole wheat kernels by Simmonds et al. (1953) yielded a linear relationship between the logarithm of the free moisture content and the drying time. This relationship is characteristic of the first order reaction rate processes defined by the equation:

$$\ln\left(\frac{M-M_E}{M_O M_E}\right) = -kt \tag{7}$$

where:

M = moisture content at time t
M_O = initial moisture content
M_E = equilibrium moisture content
k = rate constant of the process.

The value of M_E was defined as the dynamic moisture equilibrium content. This value differs from the equilibrium moisture content usually attributed to drying grains where the moisture content is determined after leaving the material exposed to certain atmospheres for several days. The dynamic equilibrium moisture value is several percent higher than the value determined by the static measurements. In a work dealing with the affects of the biological structure on the drying of wheat grains, McEwen et al. (1954) pointed out the different curves that resulted when the different values of the equilibrium moisture content are applied to the data. Figure 1 is a general reproduction of these curves. The dynamic moisture content was obtained by taking the asymptotic value of the moisture content on a semilogarithmic plot of moisture content versus time.

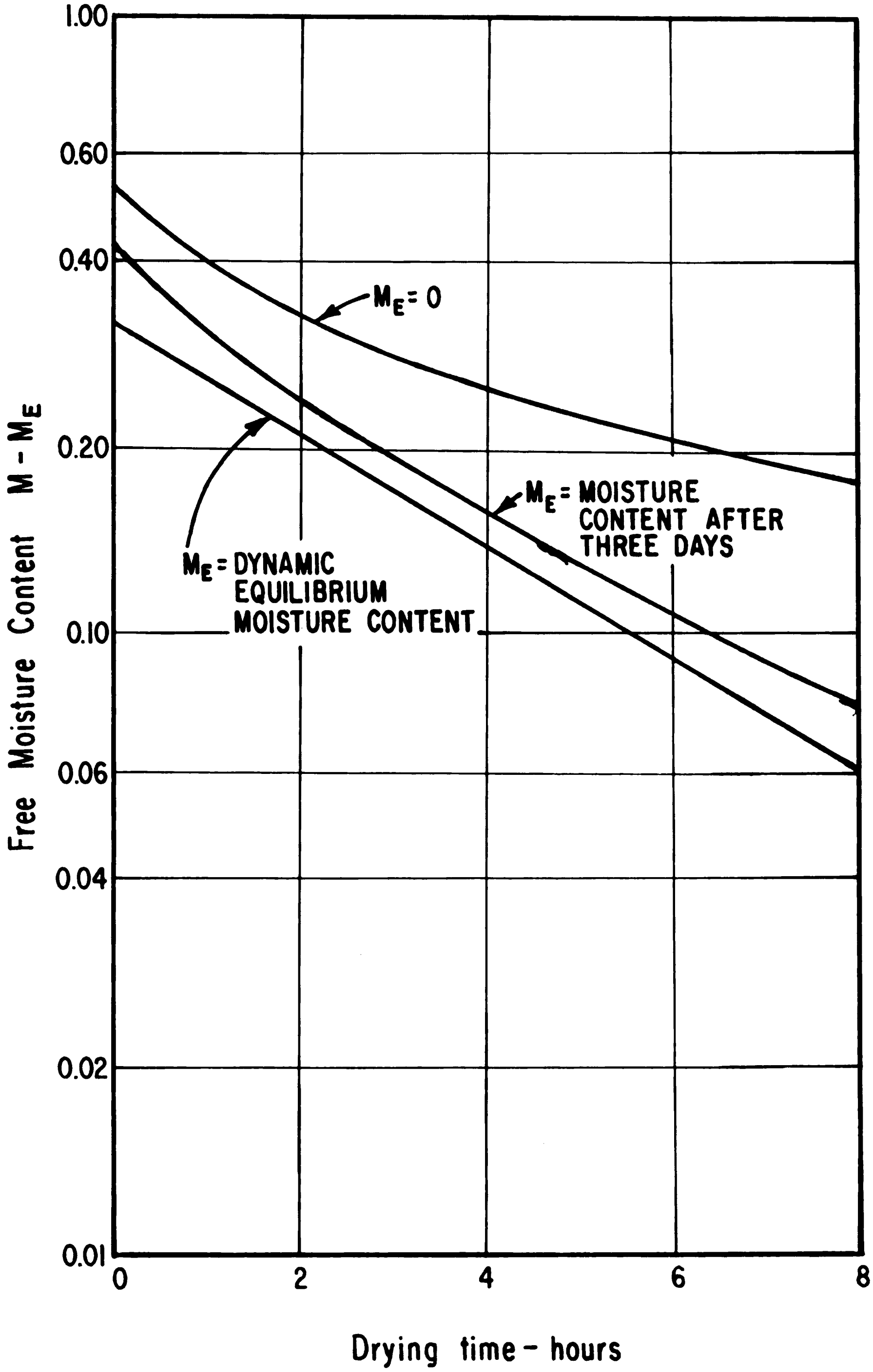

Figure 1. Free moisture content versus time for various values of equilibrium moisture content

McEwen et al. (1954)

McEwen et al. explained this anomaly in measurements of the equilibrium moisture content by noting that wheat grain is a living material in which physical and chemical changes continually occur. As the "free" moisture is removed the grain begins to adjust to its new environment. These adjustments are attributed to the fact that physical and chemical changes in the proteins and carbohydrates occur which liberate moisture. As a result, at least two equilibria are present, one for the free moisture and the other accompanying the physical and chemical changes.

Pabis and Henderson (1961) presented a critical analysis of the drying curve for shelled corn. These researchers proposed that moisture diffusion is the controlling factor in the drying of corn and applied Fick's law using the assumptions that 1) the kernel shape approximated a brick and 2) the shape approximated a sphere. In this work, the surface moisture concentration was assumed to be equal to the equilibrium moisture value as determined by static measurements. The advisability of using this value has been just previously discussed. However, Pabis and Henderson claim that the results indicated that the diffusion equation does apply. The equation based on a spherical shape produced a better fit for the experimental data when the diffusion coefficient was considered constant. The brick shape was more appropriate when the diffusion coefficient was regarded a function of time.

Finney et al. (1962) reported on the effects of conduction heating on the drying of shelled corn. They applied equation (7) and found that the relation between the logarithm of the free moisture ratio and the drying time was approximately linear when the value of M_E was set equal to the value suggested by the Henderson equation:

$$1 - R_h = \exp(-c\ T\ M_E^{n}) \qquad (8)$$

where:

R_h = equilibrium relative humidity, (a decimal).
M_E = equilibrium moisture content, percent dry basis.
T = temperature, R
c, n = constants, 1.10 x 10.5 and 1.90 respectively for shelled corn.

This equation has been used to correlate equilibrium moisture data for static equilibrium measurements. Finney also stated that the relationship between the free moisture ratio and drying time could be made linear by assuming a dynamic equilibrium moisture content rather than the value predicted by equation (8).

In tests involving a rate process which can be characterized by some constant, such as the diffusion coefficient or the rate constant it is common practice to examine the variation of this constant with temperature. This has been done as already mentioned by Jason for the drying of fish muscle. In addition, Henderson and Pabis (1961) have analyzed the affect of temperature on the drying coefficient for shelled corn, and Becker and Sallans (1955) have made a similar analysis of the diffusion coefficient for wheat.

The general approach to such an analysis is to plot the logarithm of the coefficient under study versus the reciprocal of the absolute drying temperature. Such a plot almost invariably shows a linear relationship to exist between the logarithm of the coefficient and the reciprocal of the absolute temperature. This indicates that an Arrhenius type equation is operative. The general form of the Arrhenius equation is:

$$c = A\exp(-E/RT) \tag{9}$$

where:

c = coefficient under examination, rate constant or diffusion coefficient
A = constant, denoted "frequency factor"
R = gas constant
T = absolute temperature
E = activation energy, experimentally determined from the slope of a plot of log c versus 1/T.

While such an analysis is commonly made in drying research, the analysis usually stops with a statement of the value of the activation energy without attempting to associate the magnitude of this value with the mechanism involved in the drying process.

OBJECTIVES

The objectives of this investigation are:

1) To compare the drying characteristics of wheat and corn grains in an intense sound field with those for conventional drying;

2) To make some inferences regarding the drying mechanism and how this mechanism may be affected by sound;

3) To determine if there is a practical potential use for sonic energy for drying hygroscopic materials.

APPARATUS

The apparatus used for making the tests is depicted schematically in Figure 2. A hopper contained the sample material which was fed into the drying chamber by a small auger. The auger was driven by a variable speed drive, and the drying chamber, which was a plastic tube 36 inches in length and 6 inches in diameter, was driven by an identical drive. A thermostat which could be set with a dial controlled a small electric heater which provided a small flow of air through the drying chamber. An air driven stem-jet whistle created a sonic field within the drying chamber. The stem-jet whistle is depicted in more detail in Figure 3. A pressure regulator controlled the pressure of the air to the whistle, and a pressure gauge was placed near the whistle to measure this pressure. The angle of elevation of the drying chamber could be adjusted by means of a hand screw.

Figure 3 depicts the stem-jet whistle which created the sonic field within the drying chamber. Compressed air entered the whistle and flowed through a tapered nozzle. The high velocity air which left the nozzle set up shock waves which maintained resonance in the resonator cavity. This maintained resonance in the resonance tube. The air which was used to drive the whistle was released from the sound field through ports on the whistle. A baffle plate prevented the air from re-entering the sound field.

The frequency and intensity of the output of the whistle as a function of the pressure of the driving gas are plotted in Figure 4. These calibration curves were supplied by the manufacturer of the apparatus, Branson Instruments, Incorporated, Stamford, Connecticut. The pressure of the driving air was set at 30 psig for all of the investigations. This maintained a sonic field intensity of about 165 db. (Ref: 0.0002 microbar) and a frequency of about 11.5 KC.

It was assumed that when the drying chamber was rotated, the flow of heated air through the chamber was turbulent and the temperature distribution across the chamber was uniform. The temperature distribution along the tube was also uniform.

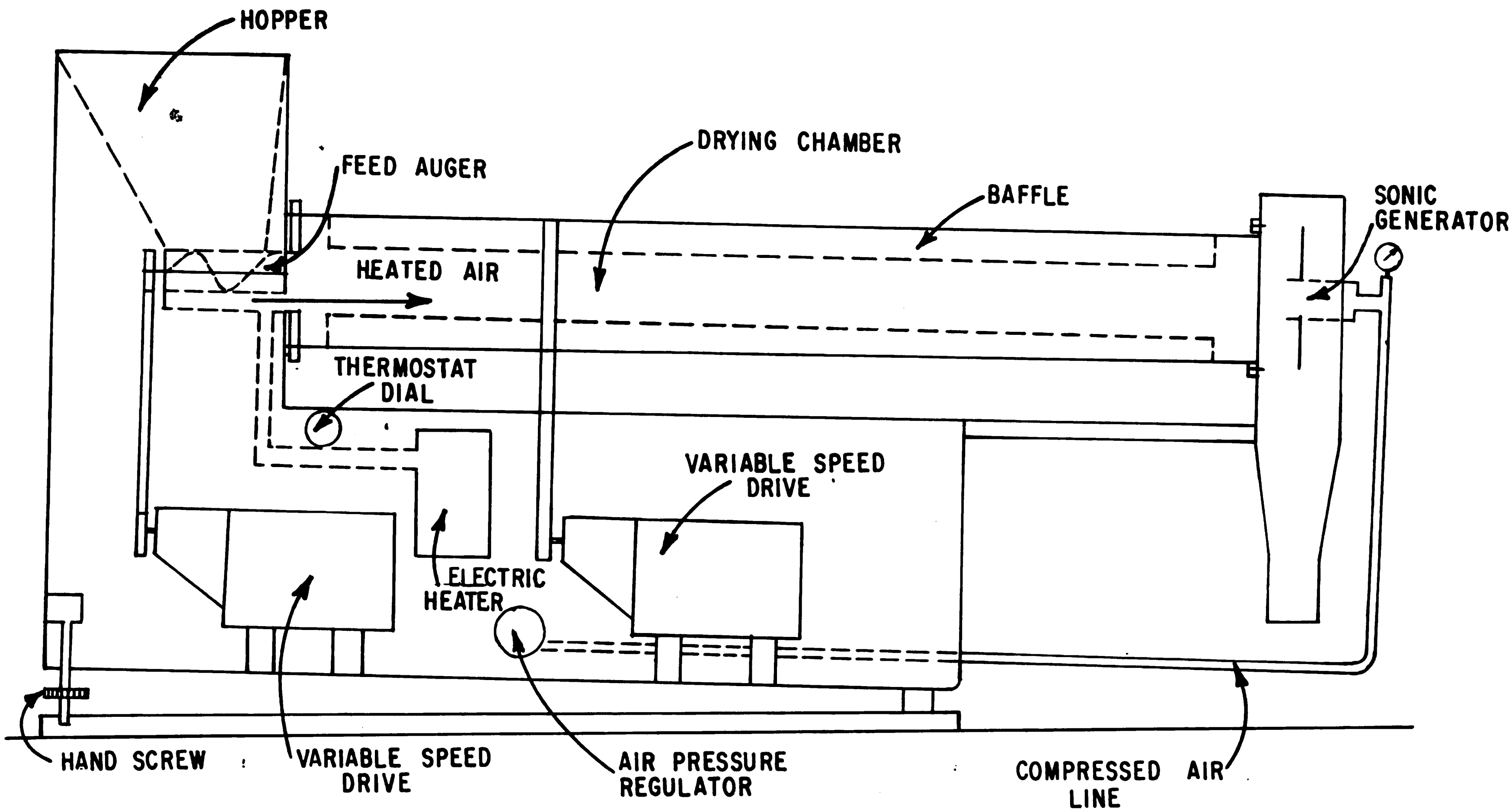

Figure 2. Schematic diagram of laboratory sonic dryer

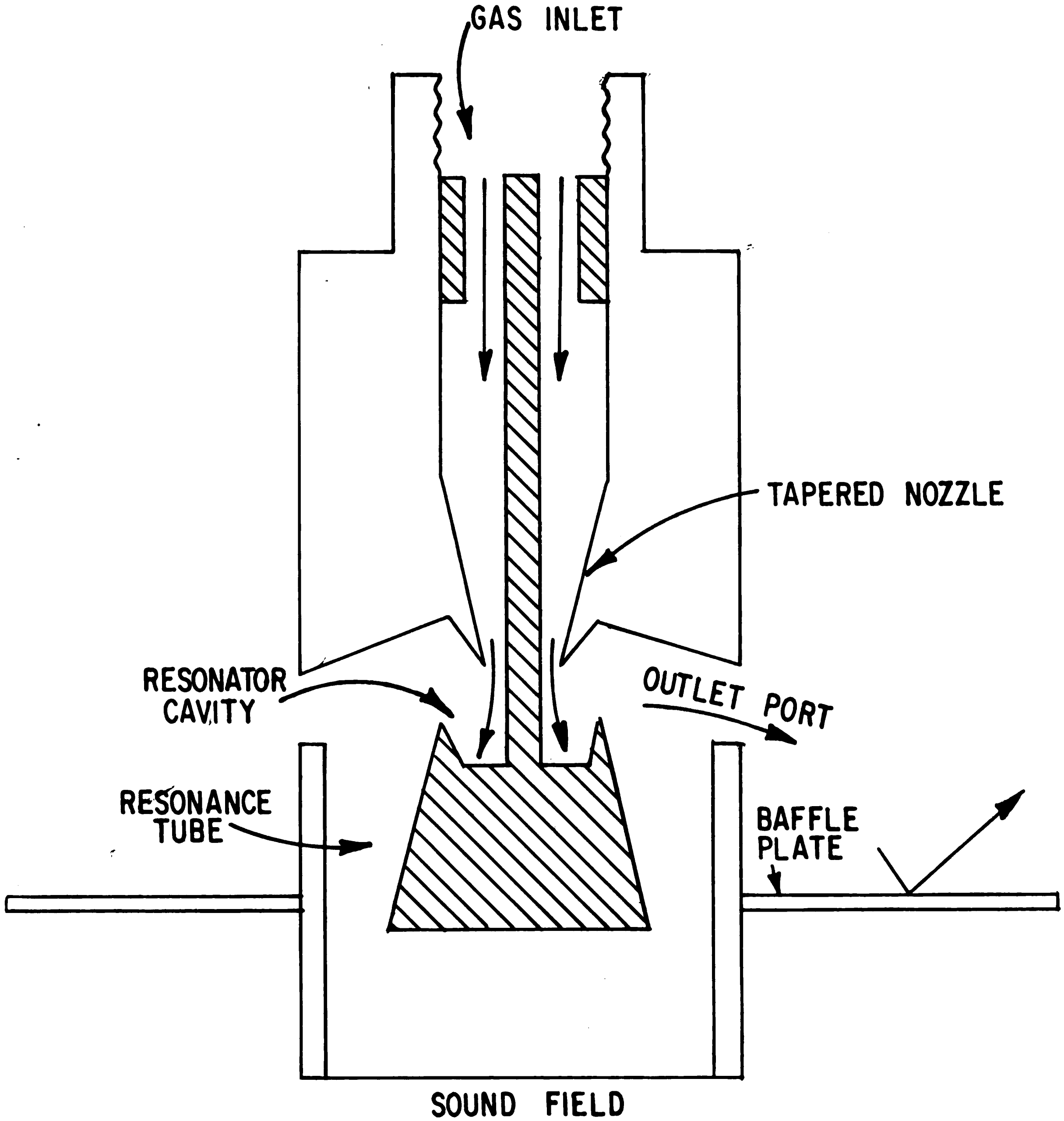

Figure 3. Schematic diagram of stem-jet whistle

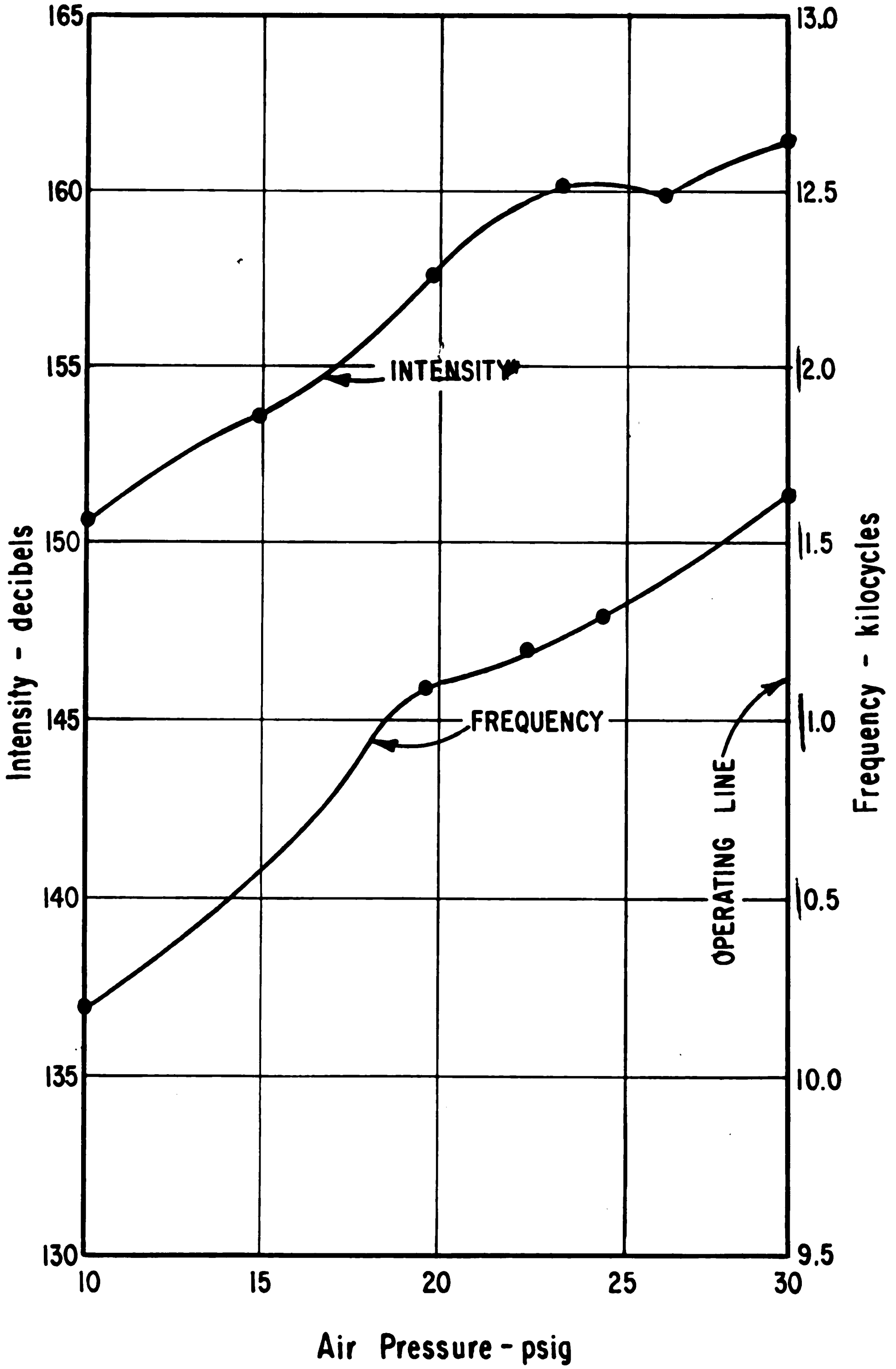

Figure 4. Calibration curves for sonic whistle

PROCEDURE

Soft winter wheat containing approximately 15% dry basis moisture was placed in 40°F. storage shortly after harvest. Approximately three weeks prior to making the drying tests, the wheat was placed in one-gallon jars and water was added to provide an average moisture content of about 30% dry basis. The samples were then sealed and again placed in the 40°F. storage. The samples were periodically shaken, while in storage, to ensure that the grains in the sample would be in equilibrium.

Hustrulid (1963) compared drying curves for naturally moist, re-moistened, and frozen wheat kernels and reported that treating the samples as described above did not alter the drying characteristics from those of naturally moist grains.

Shelled yellow corn was obtained at harvest time at a moisture content of approximately 22%. To complete the tests a small amount of corn was taken from storage at about 15% moisture content. The corn was reconditioned in a manner identical to that used for the wheat. Hustrulid (1962) also reported that treating corn grains in this manner altered the drying characteristics very little from those of naturally moist grains.

To determine if the effects of sonic energy on the drying rate was affected by size reduction of the material, a portion of the grain was dried as whole kernels, and a portion of the grain was reduced in size before drying. The degree of reduction was also of interest. Therefore, the material was categorized as "crushed" and "ground" material, with the "ground" material representing the greater degree of reduction. A standard sieve analysis was made as outlined by Henderson and Perry (1955).

The fineness modulus and the uniformity index characterize the reduced material. The fineness modulus provides a relative measure of the average particle size in the sample, with lower moduli corresponding to finer material, and the uniformity index provides a measure of the relative distribution of coarse, medium, and fine particles within the sample.

The crushed wheat had an average fineness modulus of 4.18 and a uniformity index of 4:6:0. Similarly crushed corn had a fineness modulus of 3.99 and a uniformity index of 4:5:1. The fineness modulus for ground wheat was 2.63 with a uniformity index of 0:6:4, and ground corn had a fineness modulus of 2.79 with a uniformity index of 1:6:3.

The air flow through the drying chamber was measured with a rotameter and maintained at 6 scfm for all drying tests. The temperature of the air flowing through the drying chamber was read from a dial thermometer near the inlet to the chamber. This reading was calibrated by measuring the temperature at several locations in the drying chamber with thermocouples. The air temperatures used in the drying tests were 70, 145, 175, and 200 F. The absolute humidity of the air was about 30 grains per pound of dry air.

A few preliminary tests were carried out by drying these materials for a period of ten minutes with different drum rotations. Although the values varied only slightly, a drum rotation of about 56 rpm produced the maximum drying rate. Evidently, this rotation resulted in a maximum exposure of the material to the sound field. Therefore, this rotation was used for all tests.

Prior to drying, the reconditioned grain samples were permitted to come into equilibrium with the room temperature. Samples of approximately 100 grams were used since this size of sample could be sparsely scattered through the drying tube without distorting the sound field, and each grain of material was therefore exposed to the sound field. At each drying temperature, one sample was used for each of five drying periods: 2, 5, 10, 20, and 40 minutes.

This procedure made it possible to obtain the relation between the moisture content and the drying time at each temperature. For crushed material, drying periods of 2, 5, 10, 15 and 30 minutes were employed, while for ground material the drying periods were reduced to 2, 4, 6, and 8 minutes. Three replications were made of each test.

The moisture contents of the samples were determined by the weight lost in an air oven at 212^{o}F. for three days. A Mettler balance which could be read to 0.0001 grams was used on all moisture content measurements.

This procedure made it possible to obtain drying curves of moisture content as a function of time. In addition, the sonic whistle could be turned on or off without affecting other drying conditions. Therefore, every test made using the sound generator was compared to a test without the sound generator. This technique made it possible to ascribe the differences in the drying so obtained to the presence of the sound field.

RESULTS

The results of the investigations on whole and crushed wheat and corn grains are depicted by the curves of moisture content versus drying time in Figures 5 through 8. In all cases, the solid line curves represent drying under given conditions of temperature, air flow, and drum rotation in the absence of the sound field, and the broken curves represent drying under identical conditions with the sound field applied. All points on these curves represent the mean of three separate tests. Such curves, as depicted in Figures 5 through 8, are common for the drying of hygroscopic materials such as wheat and corn.

It is noted that for a given drying condition the curve representing the sonic drying exhibits a lower moisture content than the corresponding conventional drying curve at any given time after the beginning of drying. This indicates that more moisture was removed in the given time under sonic conditions than was removed in the same time in the absence of sound. From a practical standpoint these curves give some indications about the relative increase in the drying rate which may be expected due to the sonic field. For example, from Figure 5 it is observed that whole wheat at 145°F. will dry from 30% to 16% in about 40 minutes in the absence of sound, whereas the same amount of moisture is removed in 30 minutes in the presence of sound. At 175°F. the corresponding times are about 23 minutes and 19 minutes, and at 200°F. there is only about one minute difference between sonic and conventional drying procedures in the time required for this drying. In short, the advantage of the sound diminishes as the temperature increases for the drying of whole wheat. The same effect is noted if the moisture contents at a given time are observed. Again in Figure 5, the differences in moisture contents between sonic and conventional drying after a drying period of 40 minutes show a steady decrease as the temperature is raised from 70°F. to 200°F.

Figure 6 shows similar results for crushed wheat. Crushed wheat dries more readily than whole wheat regardless of the temperature between 70°F. and 200°F. Figure 6 shows that drying crushed wheat at 70°F. may be practical, and the application of the sound field reduces the time required to dry from 29% to 13% from 30 minutes in the case of conventional drying to about 15 minutes for sonic drying. At higher temperatures, the advantage of the sound field was less pronounced than for the whole grains. It must be pointed out, however, that if it is desired to dry the crushed material to a much lower moisture the application of the sound field will have a distinct advantage even at higher temperatures. For example, crushed wheat will dry from 29% to 5% moisture in 30 minutes at 145°F. under conventional conditions, while the corresponding time for sonic-drying is only 18 minutes. In summary, the application of the sound field produces the same effect on crushed wheat as was shown for whole wheat except that the advantage of the sound field shifts to lower temperatures.

The curves of Figure 7 for whole corn exhibit a shape similar to those for wheat. The effect of the sound however does not diminish with increasing temperature as it did in the case of wheat. The advantage of sound was not as great at 70°F. for shelled corn as it was for whole wheat, but at 200°F. the trend was reversed, and the advantage of sound was greater for shelled corn than for whole wheat. In fact, the advantage of the sound appears slightly greater at 200°F. than at 70°F.

The curves for crushed corn, depicted in Figure 8, are very similar to those for crushed wheat. At higher temperatures, the advantage of the sound diminishes, and at 200°F. there is practically no difference between drying with and without sound. At all temperatures, the crushed wheat appeared to dry more readily than the crushed corn, and the advantage of the sound field appears somewhat greater for crushed wheat than for crushed corn.

While Figures 5 through 8 provide an idea of the practical advantages to be expected from applying a sound field to the drying process, these curves do not provide any ideas for more quantitative explanations of the effects of sound on the drying process. From the general shape of the curves of moisture content versus time, it would appear that the drying processes obey a relation similar to that given by equation 7:

$$\ln \left(\frac{M - M_E}{M_o - M_E}\right) = -kt \qquad (7)$$

Figures 9 through 12 are plots of the logarithm of the free moisture ratio, which is the term on the left side of equation (7), versus the drying time t. The slope of these curves is equal to -k, or the absolute value of the slope is k.

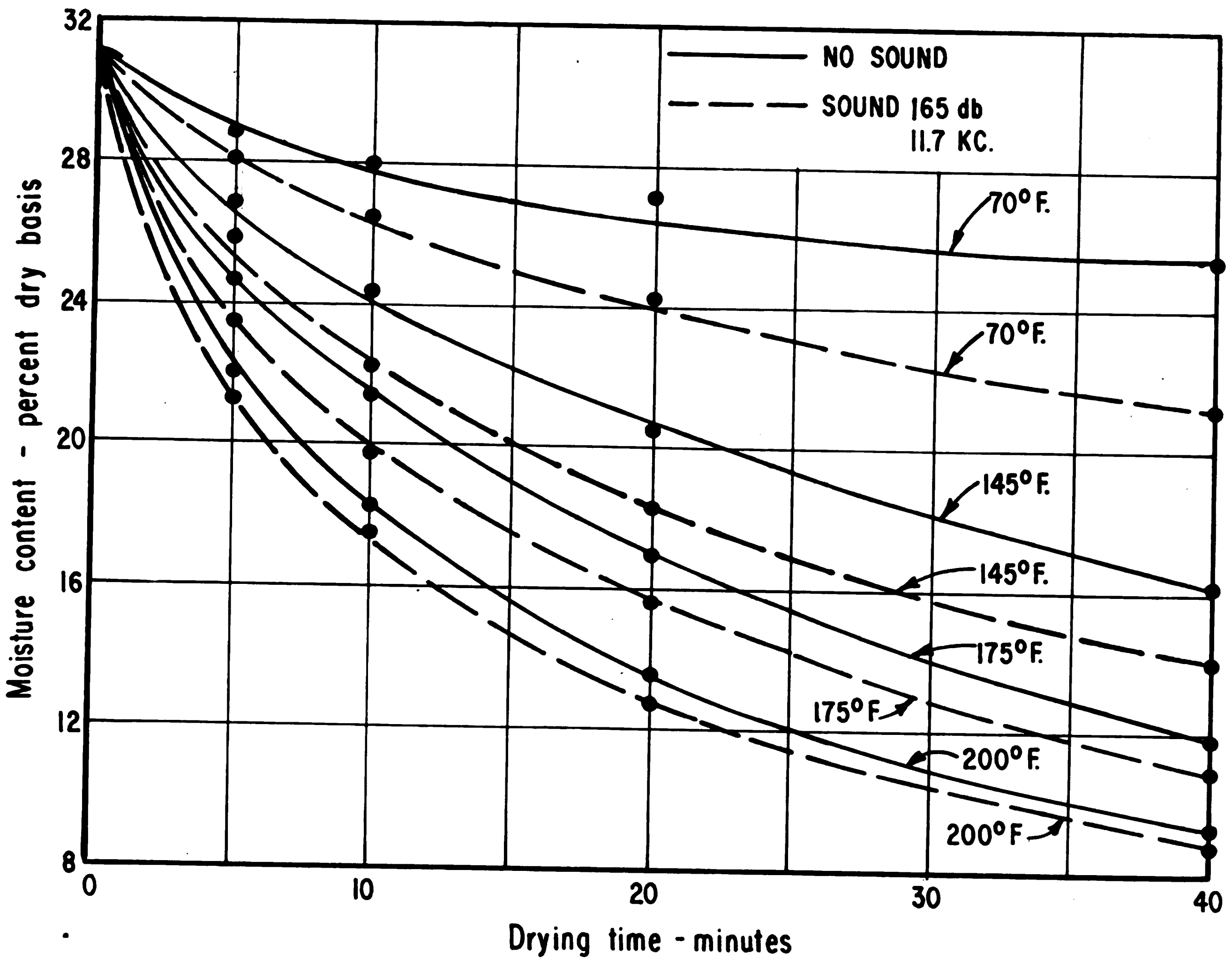

Figure 5. Moisture content versus time for whole wheat at various temperatures

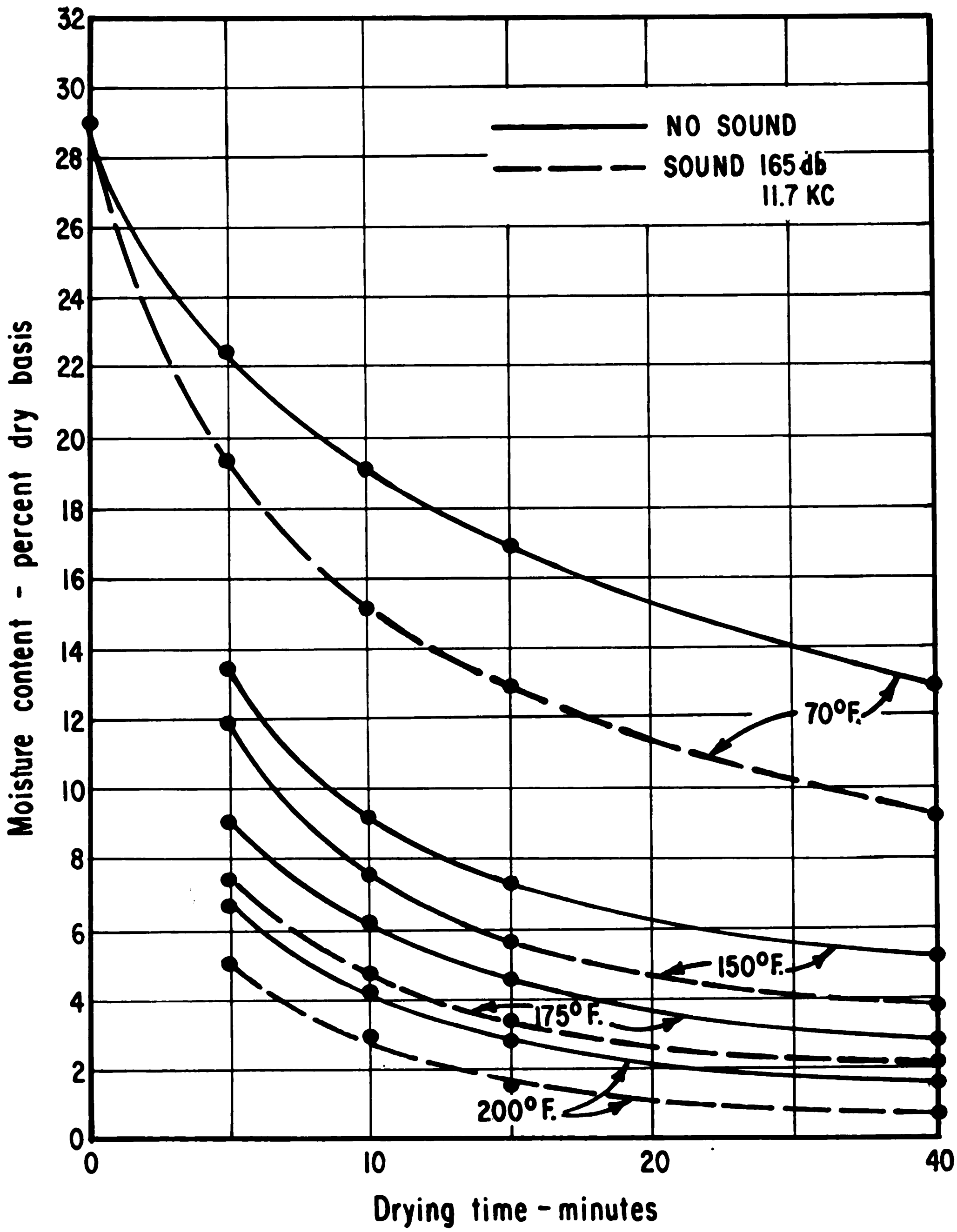

Figure 6. Moisture content versus time for crushed wheat at various temperatures

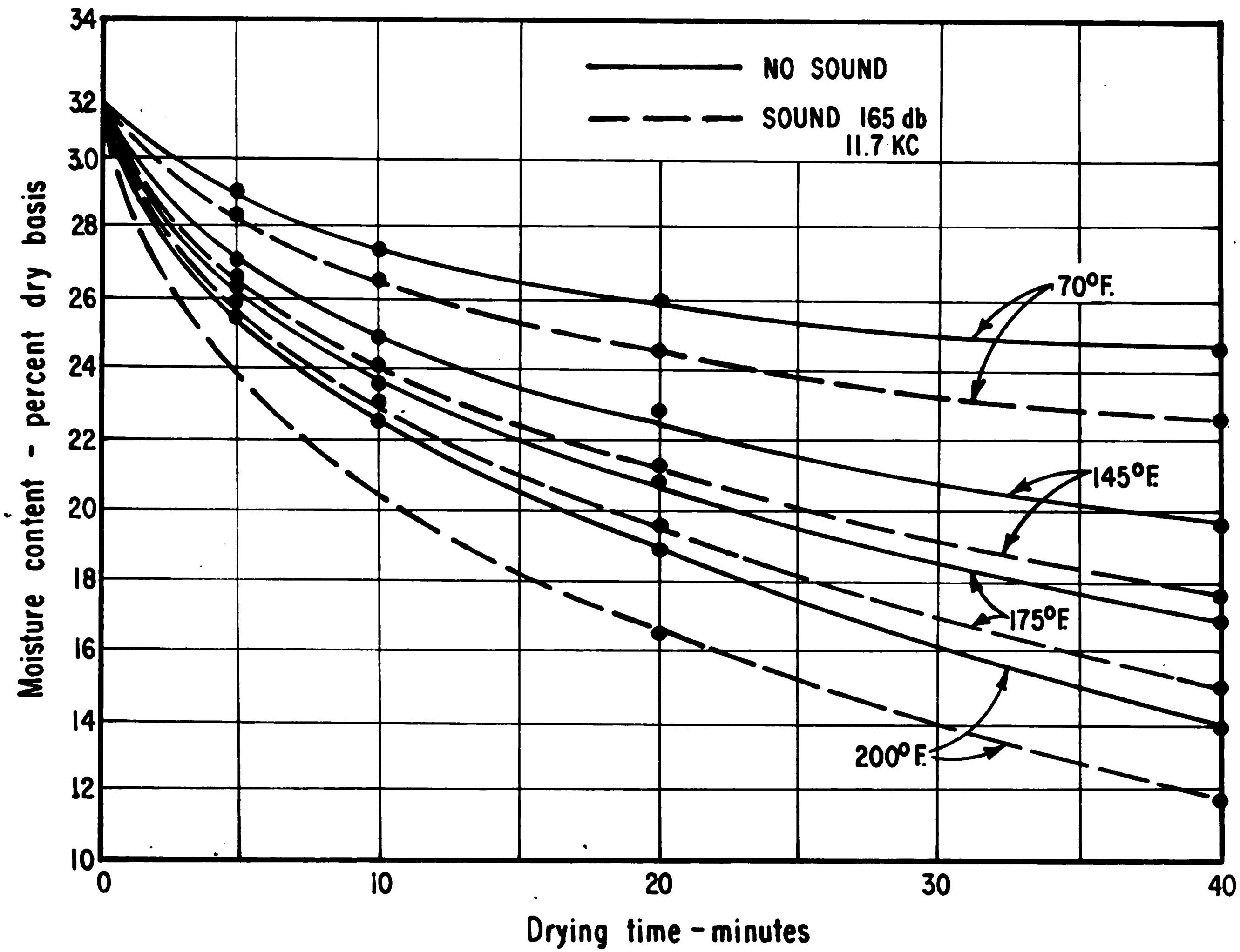

Figure 7. Moisture content versus time for whole corn at various temperatures

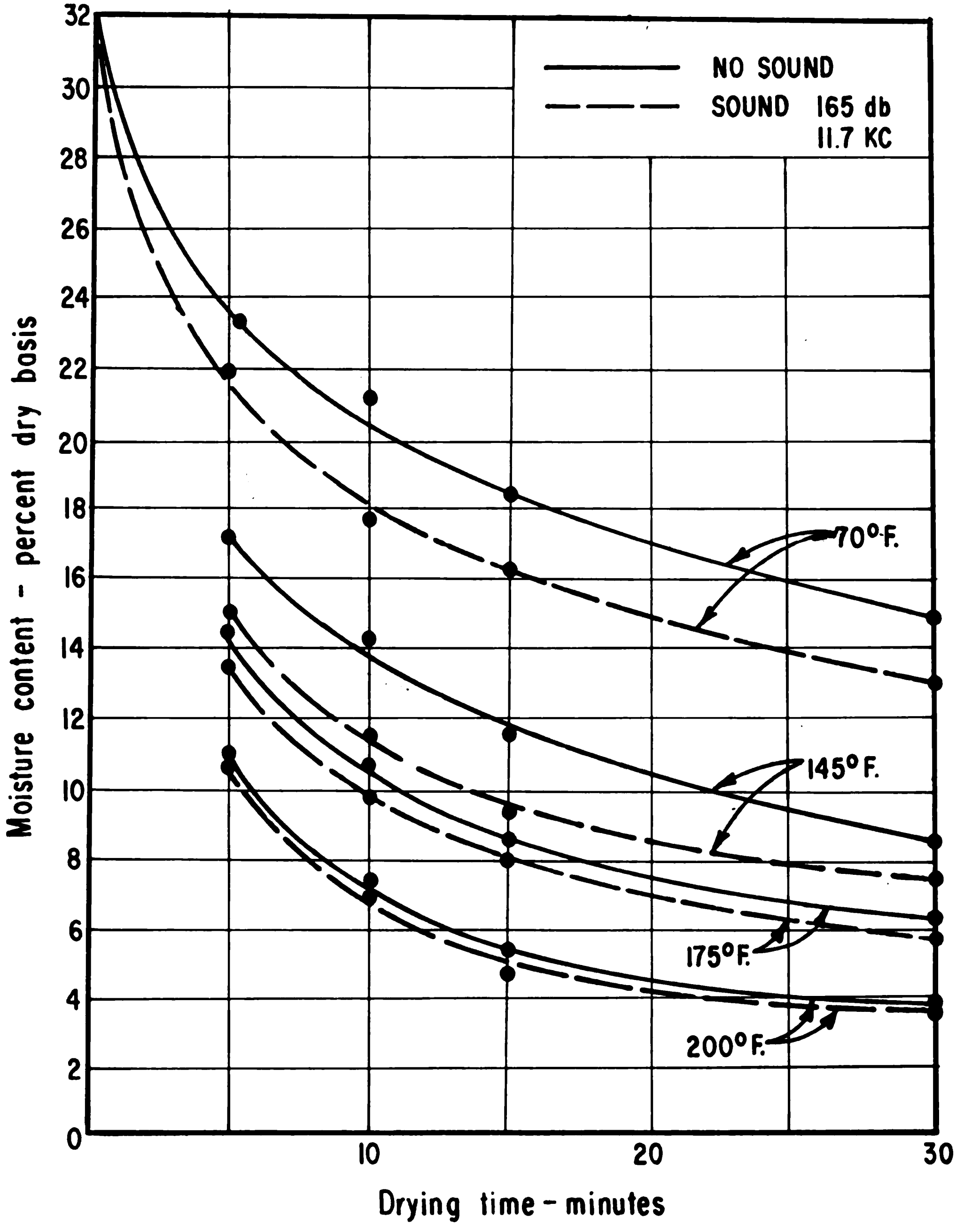

Figure 8. Moisture content versus time for crushed corn at various temperatures

The value of the initial moisture content, M_o, was not taken as that measured by the oven method, but instead, it was this value diminished by 1 to 3%. Do Sup Chung et al. (1961) observed that when grains such as wheat absorb moisture, about 2 to 3% moisture is absorbed on the surface of the grains. This moisture is transferred very rapidly, and it would not seem likely that it would obey equation (7) in the same way as the other moisture. In addition, the curves of Figures 9 through 12 are based on the dynamic value of the equilibrium moisture content. Crushed material dries very rapidly in the first few minutes of drying, and at temperature levels other than the 70^{o}F. level, the drying process cannot be described by equation (7). Therefore, for the crushed material equation (7) was altered by shifting the time axis by two minutes since after two minutes the surface moisture appeared to be removed, and the rate of drying was limited by the internal movement of moisture. The resulting equation is:

$$\ln \left(\frac{M-M_E}{M_2-M_E}\right) = -k\ (t-2) \qquad (7a)$$

where:

M_2 = moisture content after two minutes

When these corrections were made, the experimental data for whole and crushed material plotted as the logarithm of the free moisture ratio versus the drying time exhibited a linear relationship with little deviation. From these curves, values of the rate constant, k, were determined.

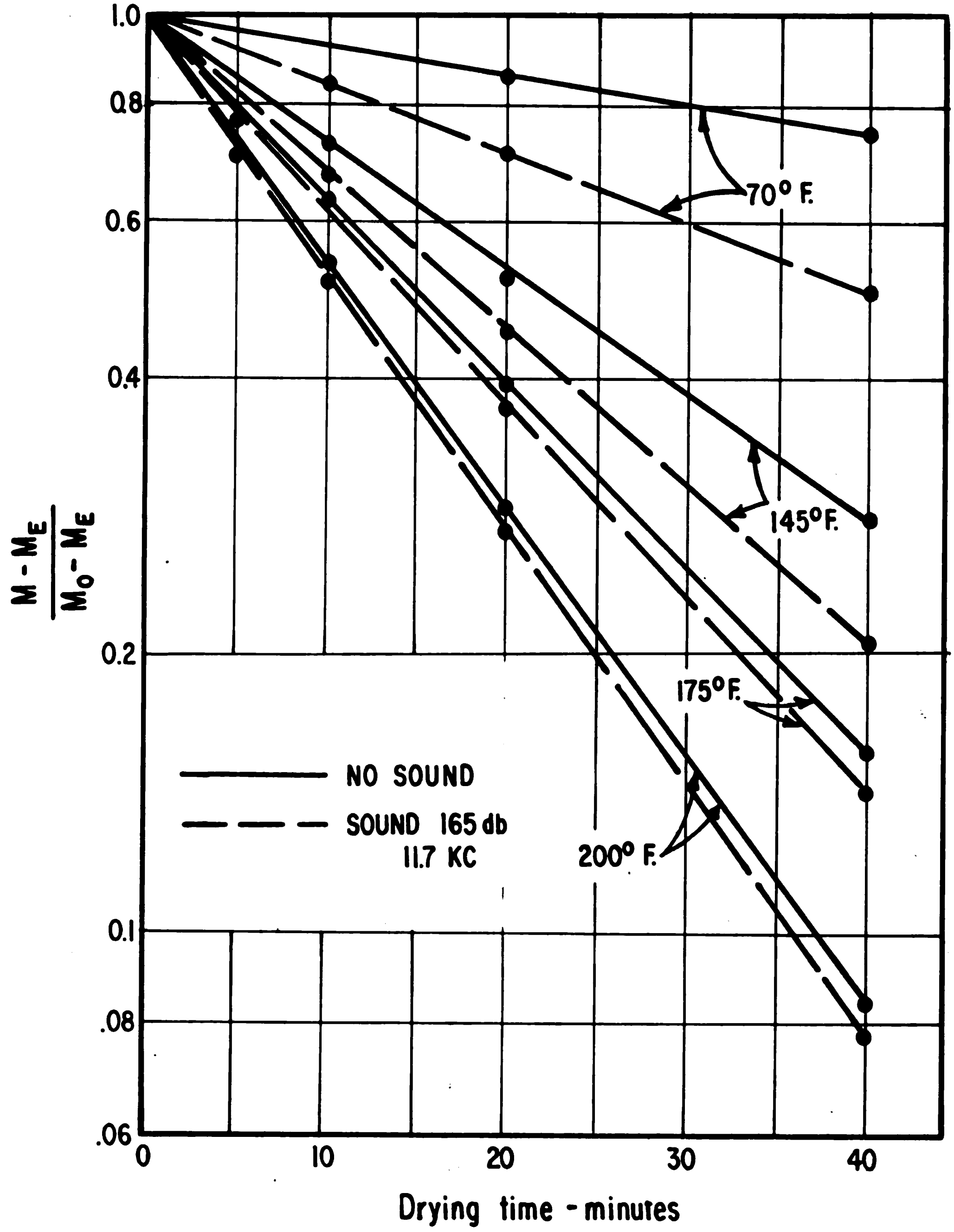

Figure 9. Free moisture ratio versus time for whole wheat

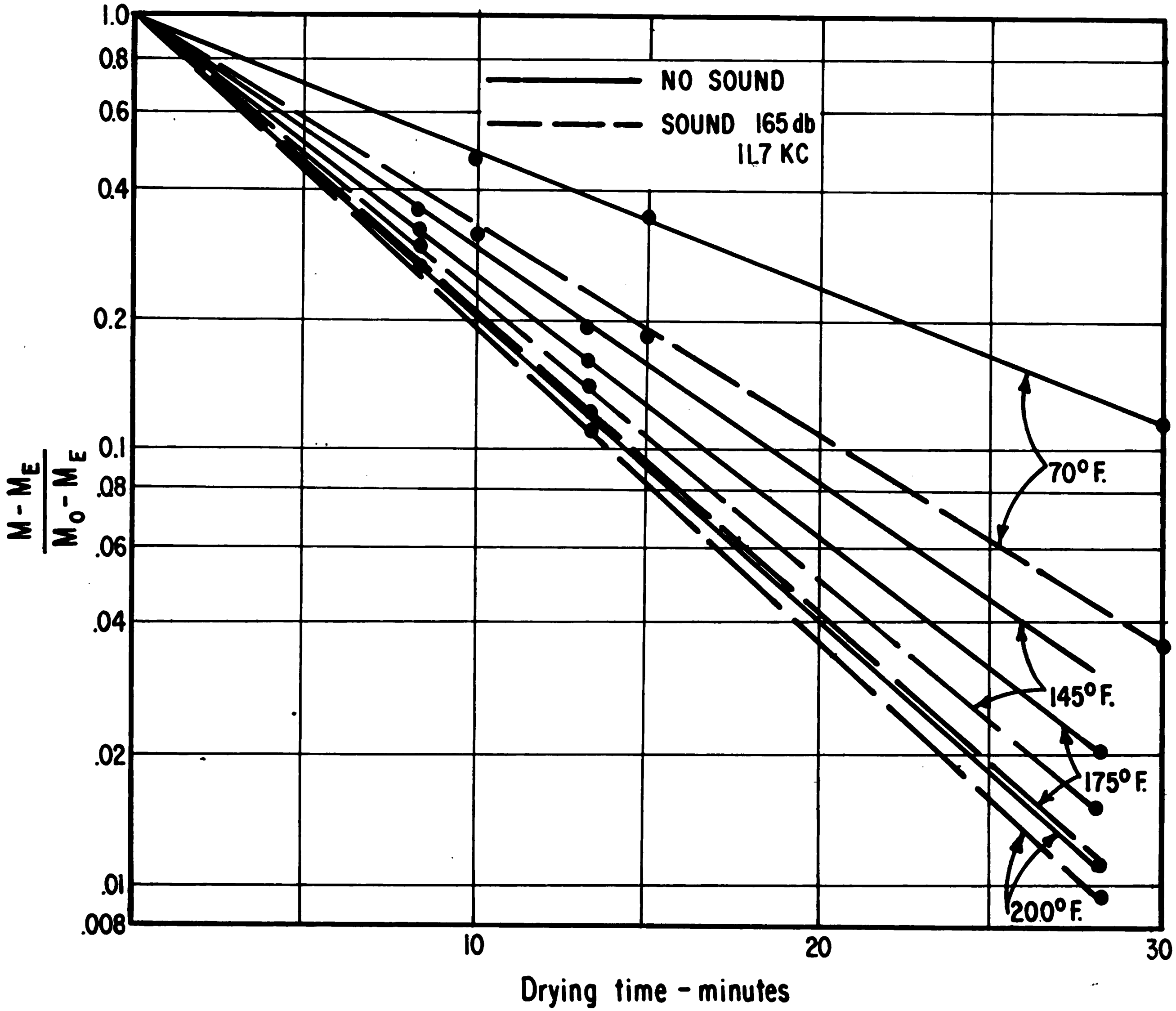

Figure 10. Free moisture ratio versus time for crushed wheat

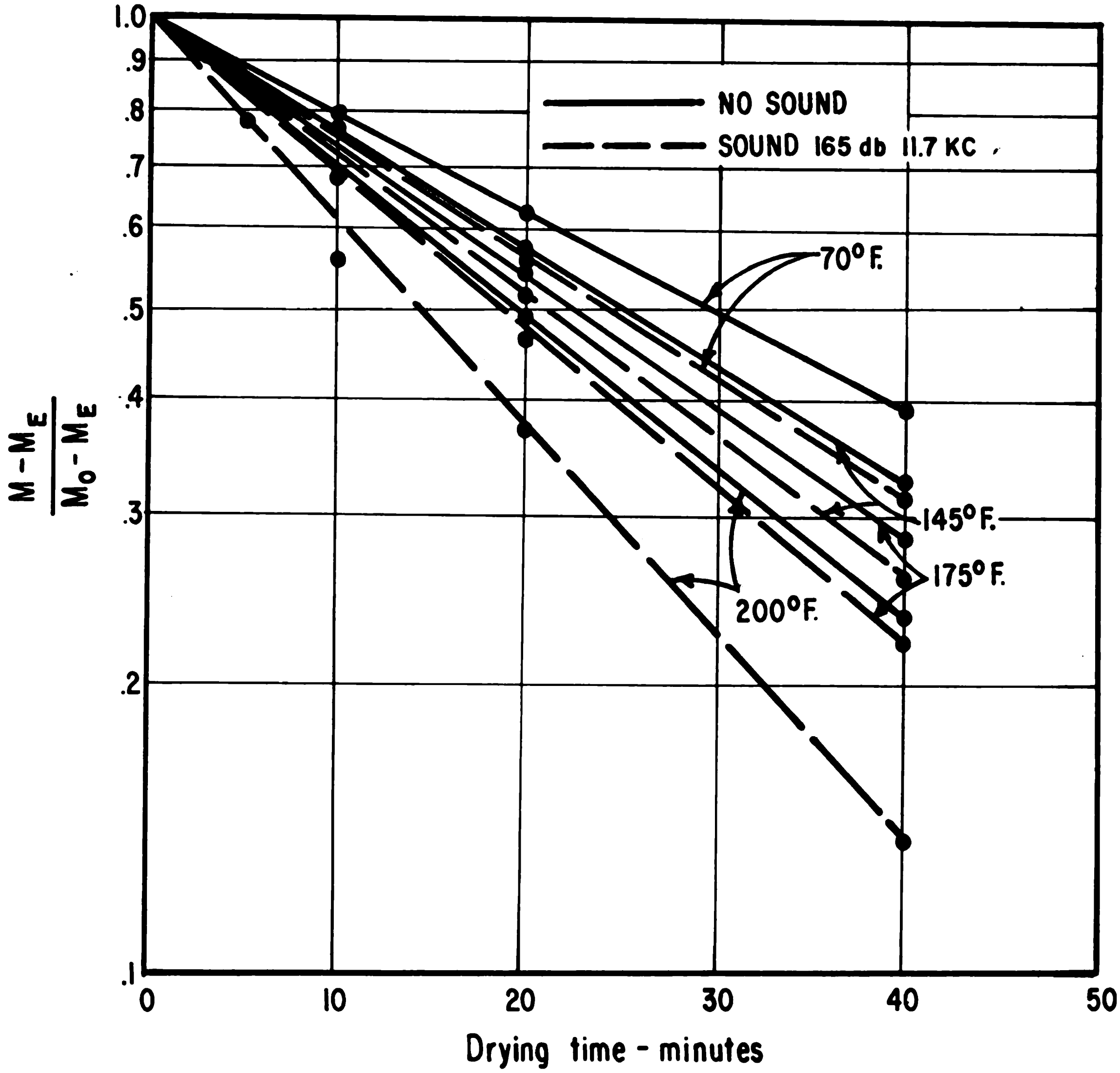

Figure 11. Free moisture ratio versus time for whole corn

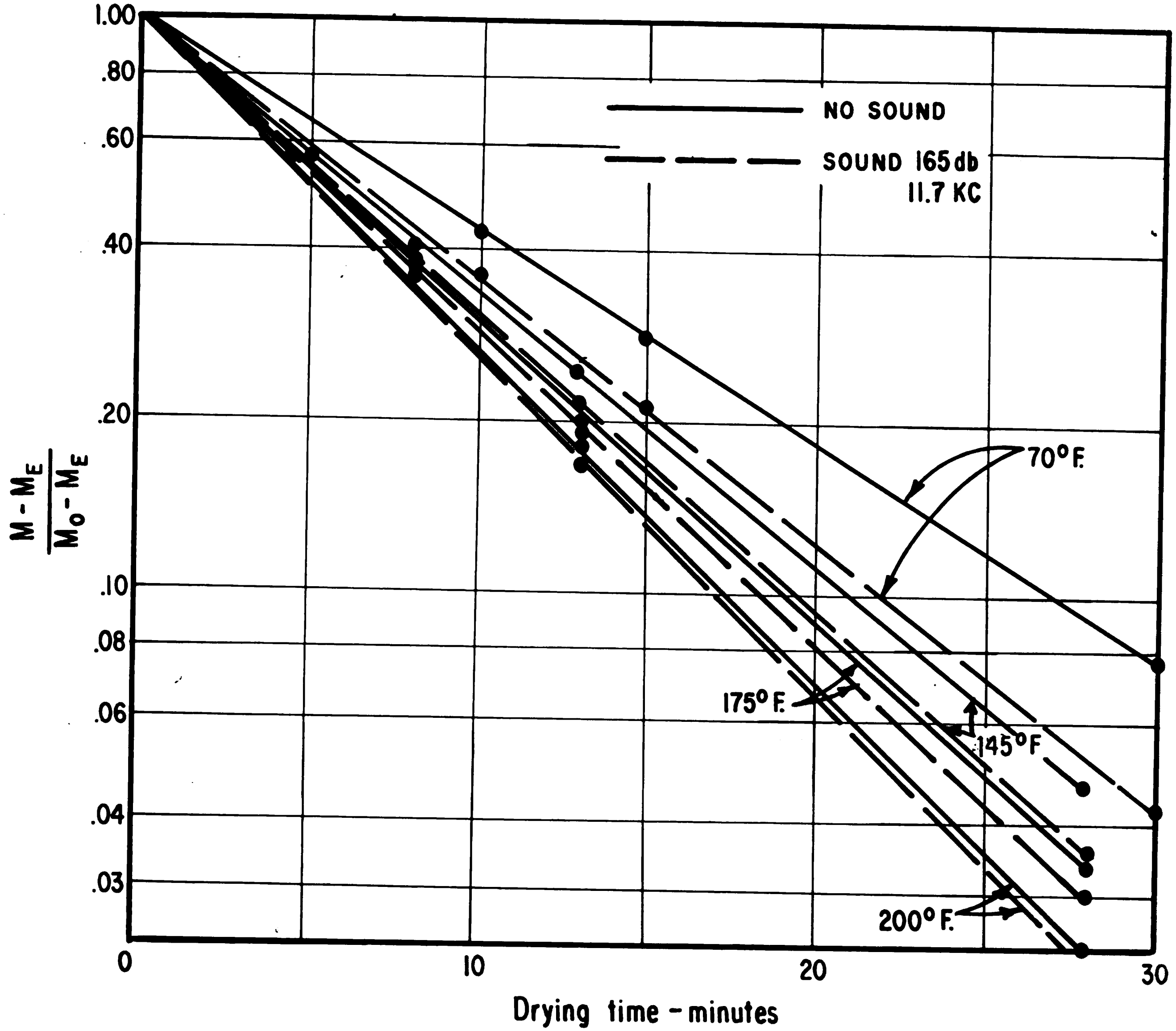

Figure 12. Free moisture ratio versus time for crushed corn

Table 1 is a summary of the dynamic equilibrium moisture values for whole and crushed wheat and corn.

Table 1. Values of the dynamic equilibrium moisture content (Percent dry basis)

	Temperature			
	70°F.	145°F.	175°F.	200°F.
Whole wheat				
No sound	13.4	10.8	8.6	7.6
Sound	13.4	10.5	8.1	7.6
Crushed wheat				
No sound	10.9	4.8	2.6	1.4
Sound	8.6	3.7	2.2	.62
Whole corn				
No sound	20.5	15.5	12.0	9.1
Sound	19.8	13.6	11.7	9.1
Crushed corn				
No sound	13.6	8.1	5.65	3.6
Sound	12.2	7.1	5.57	3.6

Table 2 summarizes the k values as taken from Figures 9 through 12.

The rate constant, k, has the dimension of reciprocal time; therefore, the reciprocal of the rate constant, denoted by τ, has the dimension of time and can be considered the characteristic time constant. After a drying period of τ minutes the right hand side of equation (7) is equal to unity. The value of M at this time is denoted M* and may be considered a characteristic moisture value.

Table 2. k- values for whole and crushed wheat and corn (min^{-1})

	Temperature			
	70°F.	145°F.	175°F.	200°F.
Whole wheat				
No sound	0.00749	0.0282	0.0470	0.0625
Sound	0.0172	0.0395	0.0488	0.0664
Crushed wheat				
No sound	.0725	0.124	0.140	0.165
Sound	0.112	0.149	0.162	0.167
Whole corn				
No sound	0.0238	0.0286	0.0312	0.0363
Sound	0.0290	0.0336	0.0377	0.0499
Crushed corn				
No sound	0.0867	0.110	0.120	0.133
Sound	0.106	0.119	0.127	0.133

If the dimensionless moisture ratio $M-M_E/M^*-M_E$ is plotted as a function of $t/\hbar$ the curve should theoretically have the shape of exp $(-^t/\hbar +1)$. Figure 13 is such a plot for all the data of these investigations. The close agreement between the data points and the theoretical line may be interpreted as a verification that the drying process is indeed a first order reaction as suggested. Therefore, it is reasonable to suspect that the rate constant k may be related to the temperature of the reaction by the familiar Arrhenius equation as expressed in equation

$$k = A\exp(-E/RT) \qquad (9a)$$

Figures 14 through 17 are plots of the logarithm of the k- values versus the reciprocal of the absolute drying temperature for whole and crushed wheat and corn. With the exception of whole corn, these plots

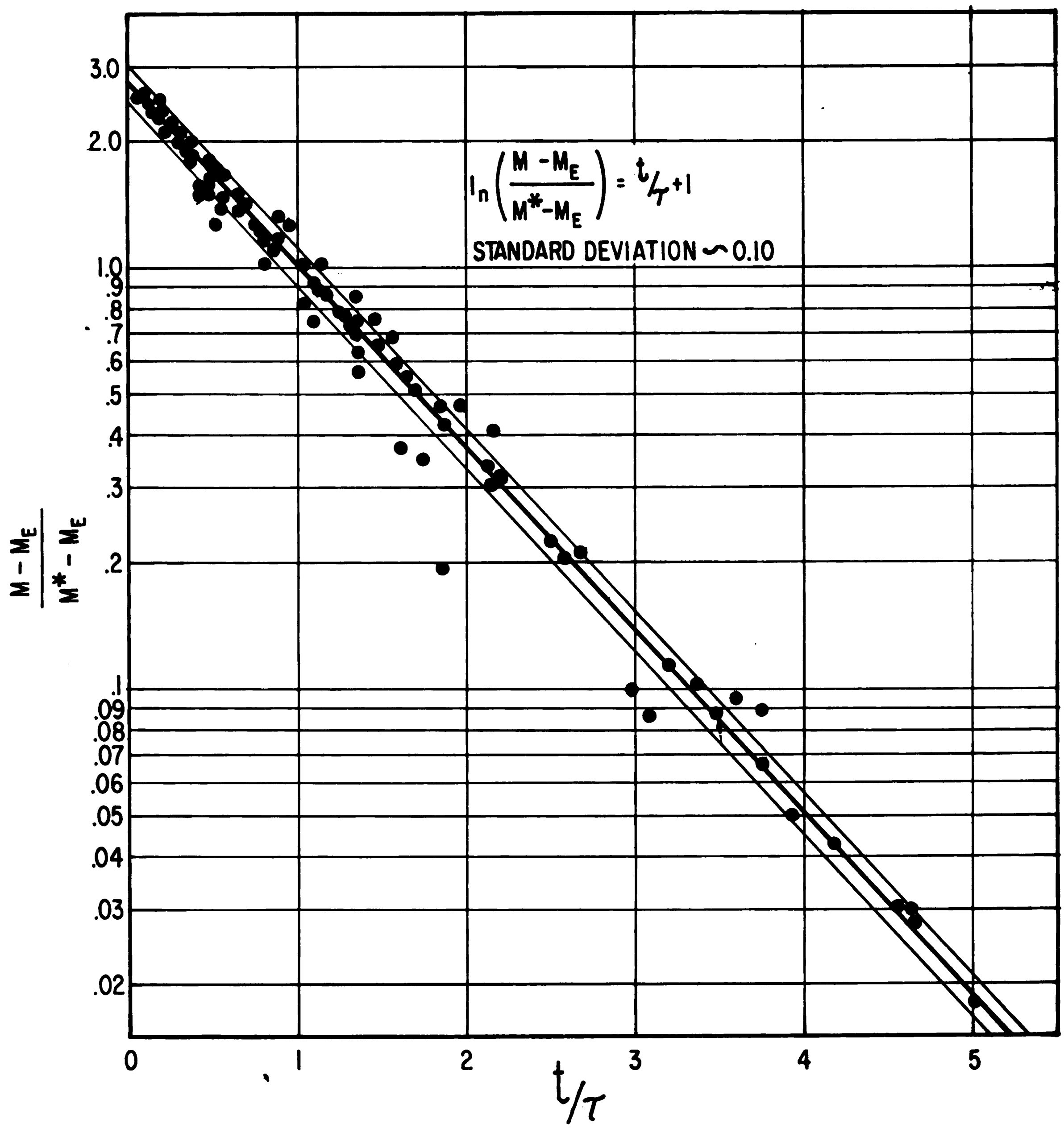

Figure 13. Dimensionless moisture ratio versus dimensionless time ratio for 128 experimental values

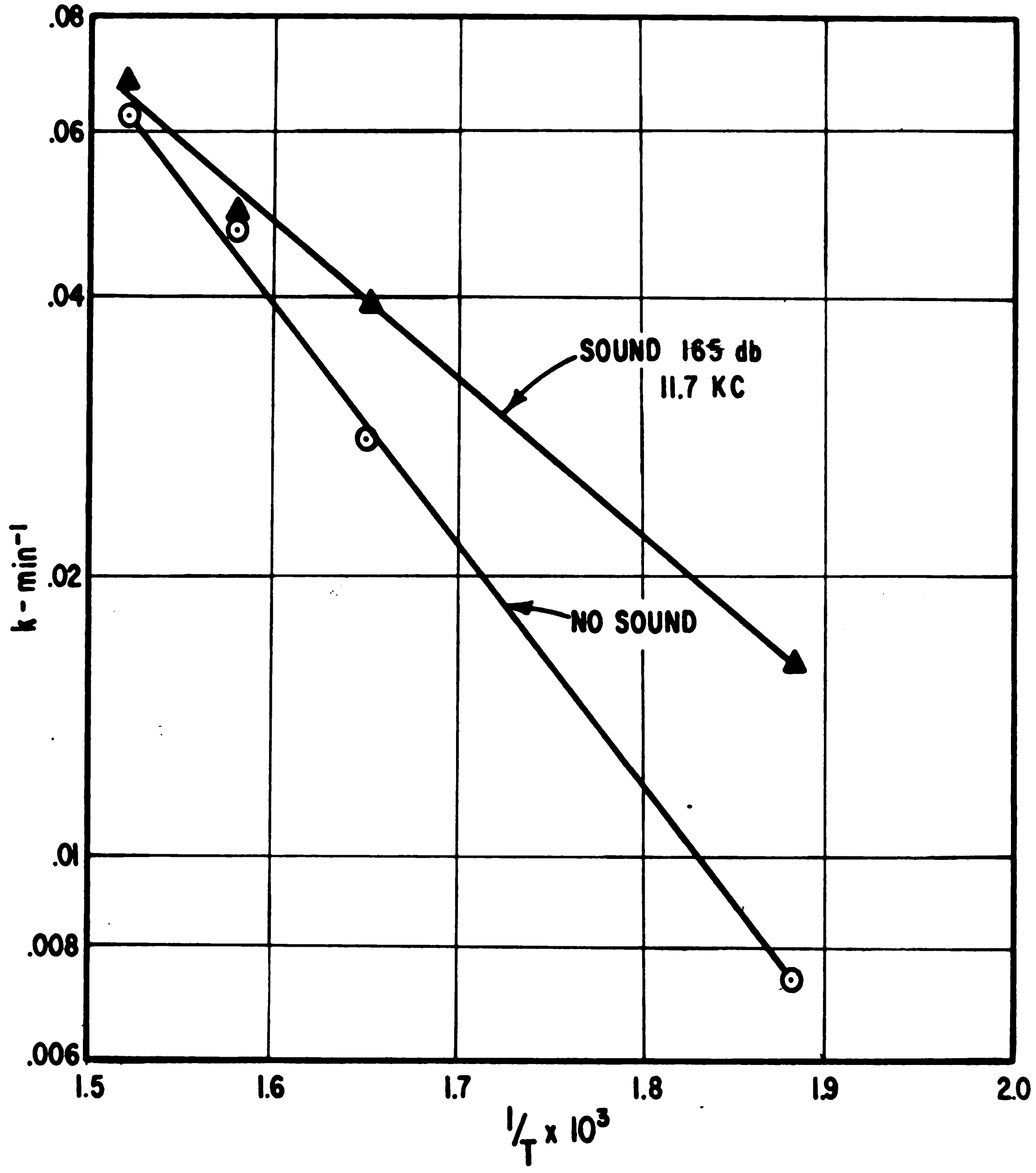

Figure 14. k versus the reciprocal of the absolute temperature for whole wheat

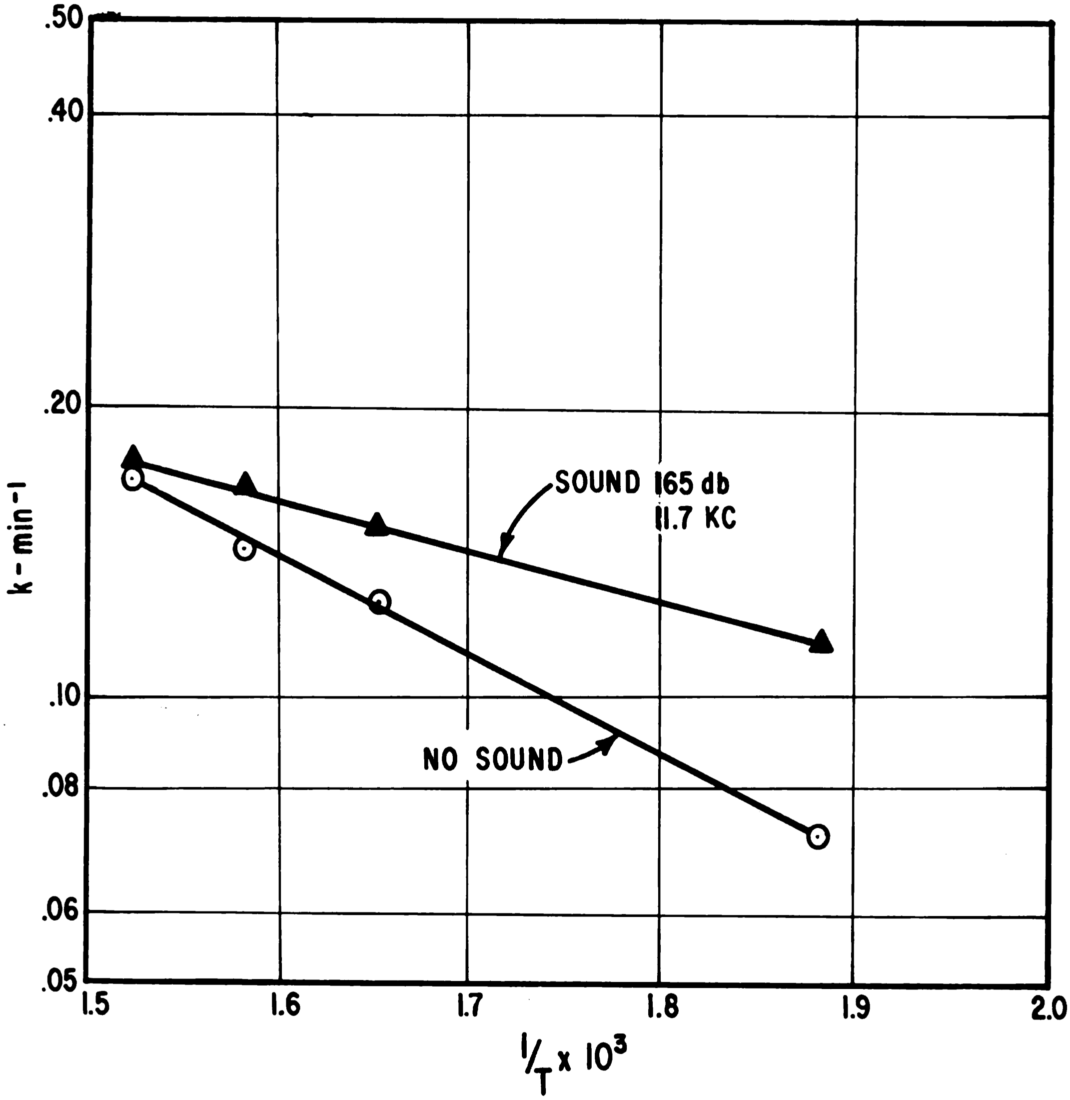

Figure 15. k versus the reciprocal of the absolute temperature for crushed wheat

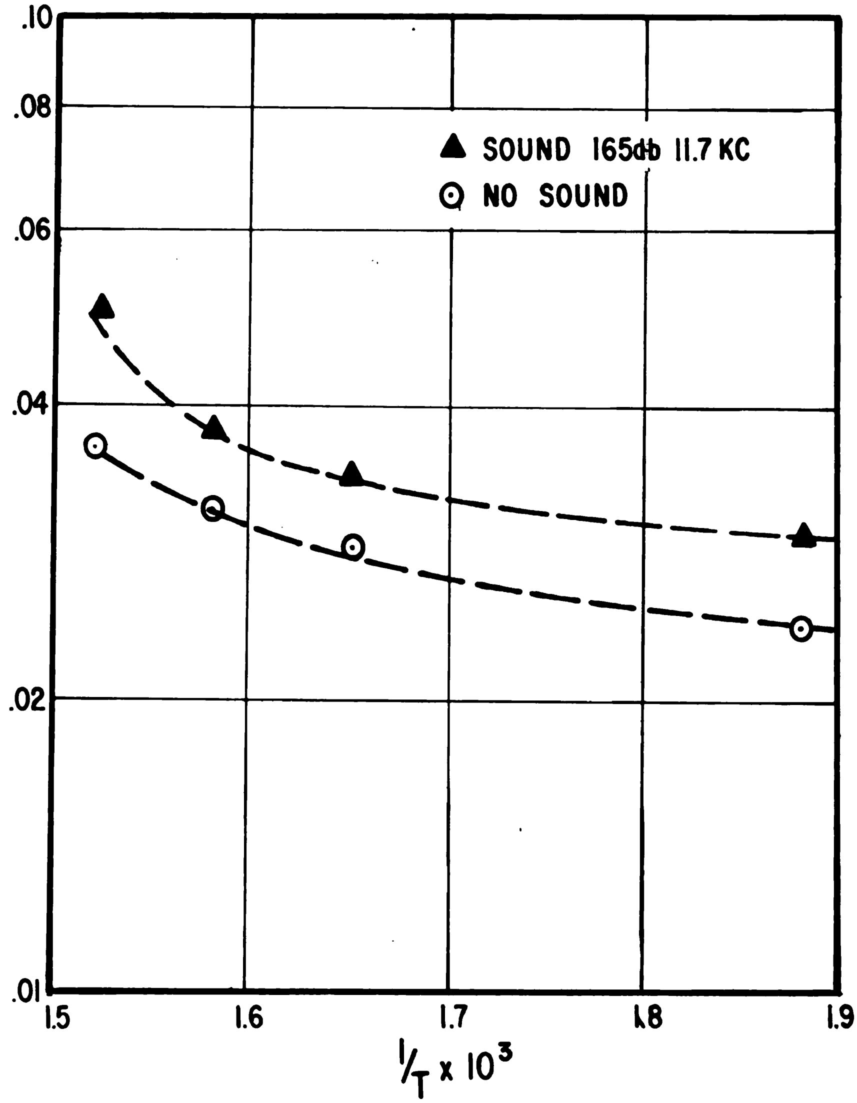

Figure 16. k versus the reciprocal of absolute temperature for whole corn

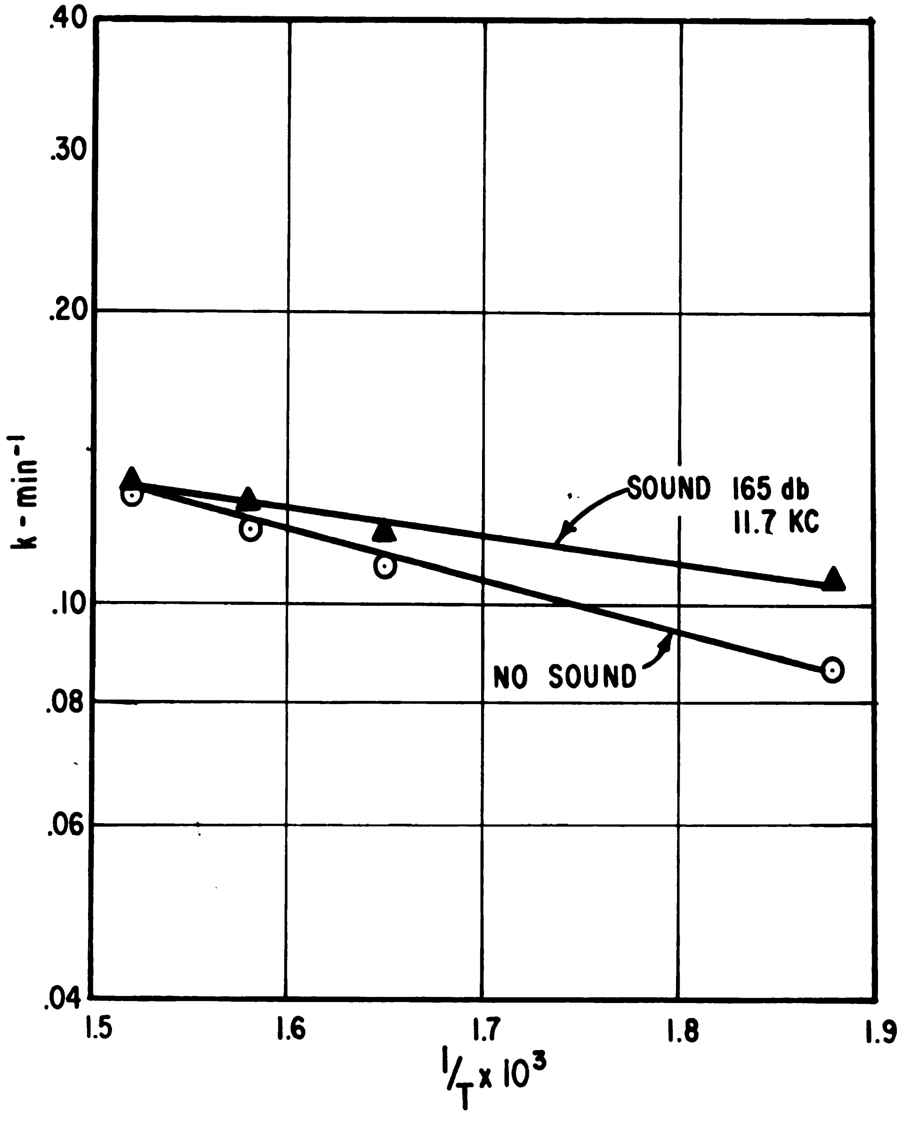

Figure 17. k versus the reciprocal of absolute temperature for crushed corn

are linear and permit an evaluation of E, the activation energy of the process. From these plots, the activation energy is given by the slope of the curve multiplied by the gas constant R.

Table 3 lists the values of the activation energies of whole wheat and crushed wheat and corn.

Table 3. Activation energies (B/lb. -mole) for the drying of whole wheat, crushed wheat, and crushed corn.

	No sound	Sound
Whole wheat	11,300	7,220
Crushed wheat	4,390	2,130
Crushed corn	2,310	1,275

The results of the investigations with ground material showed the same general trends as those for the crushed materials. Although, with ground material the drying was so rapid in the first two minutes of the drying process that it was not possible with the equipment and procedure used to ascertain whether or not an equation such as equation (7a) could be used to correlate the data. In addition, slight errors in the measurement of the drying time could lead to substantial errors in the results. Table 4 lists the moisture contents of ground wheat and corn after drying for periods of 2, 8, and 60 minutes.

Table 4. Comparison of the moisture contents of ground wheat and corn after 2, 8, and 60 minutes of drying.

	Temperature											
	70°F.			145°F.			175°F.			200°F.		
	M_2	M_8	M_{60}	M_2	M_8	M_{60}	M_2	M_8	M_{60}	M_2	M_8	M_{60}
Wheat												
No sound	19.6	14.8	5.3	11.3	5.7	2.3	7.9	3.3	1.2	4.9	1.7	.90
Sound	17.7	10.7	3.9	10.6	3.5	1.1	5.9	2.0	.79	2.8	0.6	.35
Corn												
No sound	24.7	17.0	8.2	15.1	7.8	3.8	11.5	6.4	2.5	9.3	5.2	1.8
Sound	19.9	13.6	7.7	12.2	7.5	3.5	10.6	6.3	2.1	7.6	3.3	1.3

DISCUSSION

The results of these investigations indicate that an Arrhenius equation may be used to express the variation of the specific rate of the drying process with temperature. This indicates that some type of activation must occur before the drying process can occur, and an equation of the form of equation (9) is applicable. Such processes are extremely common, and the theory of rate processes based on such reactions has been covered in many texts on reaction kinetics. An excellent treatment is given by Glasstone, Laidler, and Eyring (1941).

Briefly, the theory of rate processes as proposed by Arrhenius is based on the hypothesis that an equilibrium exists between inert and active reactants and that only the active are capable of reacting. The term "activated complex" is used to denote an active molecule which is the same as other molecules except that it contains an excess of energy equal to E/mole where E is the activation energy.

The supposition that an equilibrium exists between the reactant molecules and the activated complexes which go on to form products can be symbolically stated as:

$$\text{Reactants} \rightleftharpoons \text{Activated state} \longrightarrow \text{Products.}$$

If $K^{\ddagger}$ is used to denote the equilibrium constant between the activated state and the reactants, the rate constant can be expressed by the following equation:

$$k = \frac{RT}{N_h} \times K^{\ddagger} \tag{10}$$

Where:

R = universal gas constant
T = absolute temperature
N = Avogadros' number
h = Planck's constant

The term RT/Nh has the dimension of frequency and is sometimes referred to as the "universal frequency" since it is a constant dependent only upon the temperature and completely independent of the nature of the reactants or the activated state.

From classical thermodynamics, the following equations can be written:

$$\Delta F^o = -RT\ln K, \tag{11}$$

and

$$\Delta F^o = \Delta H^o - T\Delta S^o, \tag{12}$$

Where:

ΔF^o = standard free energy difference between products and reactants
ΔH^o = standard enthalpy difference between products and reactants
ΔS^o = standard entropy difference between products and reactants
K = equilibrium constant of the equilibrium between reactants and products.

Equation (11) can be rearranged as:

$$K = \exp(-\Delta F^o/RT), \tag{13}$$

and on substituting for ΔF^o from equation (12)

$$K = \exp(\Delta S^o/R) \exp(-\Delta H^o/RT). \tag{14}$$

Then equation (10) can be written as:

$$k = \frac{RT}{NH} \exp(\Delta S^{\ddagger}/R) \exp(-\Delta H^{\ddagger}/RT), \tag{15}$$

Where $\Delta F^{\ddagger}$, $\Delta S^{\ddagger}$, and $\Delta H^{\ddagger}$ are the standard free energy, entropy, and heat of activation respectively, that is

$$\Delta F^{\ddagger} = \Delta H^{\ddagger} - T\Delta S^{\ddagger}. \tag{16}$$

This derivation shows that it is the free energy of activation which determines the specific reaction rate at a given temperature. Thus, the higher the free energy of activation, the slower the rate of reaction at a given temperature.

If the heat of activation, $\Delta H^{\ddagger}$, is considered approximately equal to the experimental energy of activation, E, then the frequency factor, A, of equation (9) can be identified with the remainder of the term on the right hand side of equation (15).

That is:

$$A = \frac{RT}{Nh} \exp(\Delta S^{\ddagger}/R) \tag{17}$$

With these analogies between the heat of activation and the experimental energy of activation and that given by equation (17), the common Arrhenius equation (9) and equation (15) are the same. The discussion here will be made in terms of equation (15) since it is the more basic equation.

The energy diagram, Figure 18, is a comparison of the various energies of activation for the drying processes with the heat of vaporization of water. It is evident that the energies of activation are all much less than the heats of vaporization of water from wheat and shelled corn, and they are also much less than the heat of vaporization of pure water.

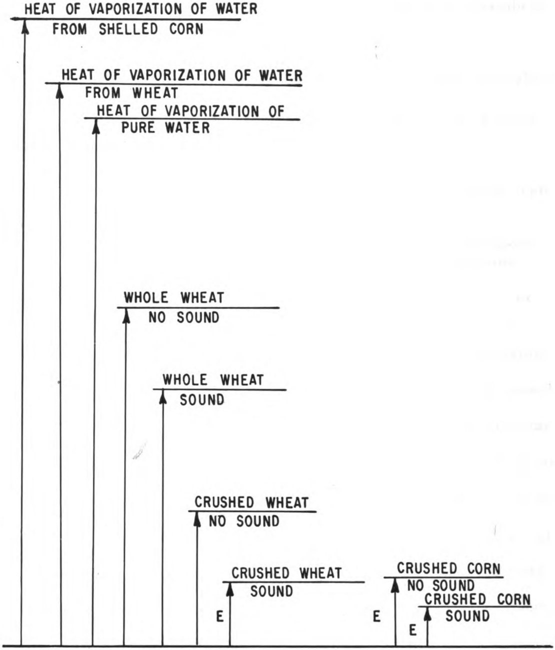

Figure 18. Comparison of heats of vaporization with energies of activation of the various drying processes

The investigations described here are not compatible with a detailed analysis of drying mechanism due to the non-homogeneity of the materials involved and the general nature of the investigations. However, from the values of the experimental activation energies it is possible to offer some qualitative suggestions regarding the process.

Barrer (1951) discusses the energy of activation and its relation to diffusion mechanisms. A classification of diffusion mechanisms according to Barrer can be made as follows:

(1) lattice diffusion, a structure insensitive diffusion through the lattice of the solid,

(2) grain boundary diffusion, a structure sensitive diffusion through breaks and openings of molecular dimensions,

(3) surface diffusion, a migration in the surface layer over the solid.

Although Barrer's discussion is primarily with metals, the same general type of migration may be expected in other types of materials.

The activation energy for lattice diffusion is the highest of the three mechanisms, followed by grain boundary diffusion, and surface diffusion.

According to Barrer, surface diffusion occurs with an activation energy much less than the heat of vaporization of the diffusing material.

Since the activation energies for the drying of wheat and corn grains have been found to be much less than the values of the heat of vaporization of pure water, surface diffusion is suggested as the mechanism whereby drying occurs in these materials. Jason (1958) observed that the energy of activation for the drying of fish muscle was less than the heat of vaporization of water and suggested that surface migration of adsorbed molecules along the protein fibrils in the fish muscle accounted for the process. It should be recognized that the surface considered here is not the exterior surface of the material, but instead it is the much more expansive internal surface of the material which is being considered.

For materials such as wheat and corn grains which do not approach a homogenous material it is unlikely that any one given mechanism would account for the entire drying process at any given time. It seems more probable that all mechanisms operate simultaneously, but that one mechanism is predominant at a given time. The activation energy then is an aggregate value for the total process.

If moisture movement in a liquid film is assumed to account for the majority of the drying process, it is necessary to consider how sound may affect the movement of this film. A liquid may be considered as a quasi-crystalline material which differs from ordinary crystalline materials in that it contains vacant sites or "holes" within the lattice. This is the well-known "hole theory" of liquids and is covered thoroughly by Frenkel (1946). The energy required to form a hole of molecular size in a liquid is equal to the energy of vaporization per molecule. This theory has been successful in predicting fluidity or viscosity and parameters such as compressibility in many liquids.

In diffusion processes, if the diffusion arises by holes moving through the volume of the liquid, it would require the energy of activation to be at least equal to the energy of vaporization of the liquid. However, if diffusion could occur by forming a hole of less than molecular size the energy of activation could be less than the energy of vaporization.

Glasstone, Laidler, and Eyring state that surface diffusion requires only about one-half as many bonds to be broken on a molecule in order for it to diffuse and therefore the energy of activation for surface diffusion is only about one-half of the energy of vaporization. This was based on metals diffusing over metal surfaces and probably was a unimolecular layer. On the other hand, if a polymolecular layer migrates over a surface the number of bonds which must be broken per molecule will be much less, and as a result, the energy of activation will be much smaller. This may explain the very low activation energy for the crushed materials.

There is an apparent discrepancy between the energy of activation for the drying processes of whole and crushed materials. If both processes are occurring by the same mechanism it would be logical to expect that the energies of activation would be approximately equal. Nevertheless, the discrepancy does exist and may be explained as follows. As previously stated, no one mechanism can be assumed to operate exclusively during the drying process. Whole kernels of wheat and corn grains are relatively tight and compact materials consisting of starchy material on the inside which is covered by a protective coat. Even though surface diffusion may account for the major portion of the internal moisture movement during the drying process, it is reasonable to expect that a substantial amount of the drying may result from bulk or lattice diffusion within the kernel. On the other hand, when a material such as moist wheat is crushed or ground it forms a light, fluffy material. Much of the starchy material in the grain is directly exposed to the atmosphere after the grains are crushed, and a "loosening up" of the starchy material occurs. These conditions allow for a more free movement of surface films, and, as a result, one would expect a greater portion of the drying to occur by surface diffusion in crushed material than in whole material. Therefore, the energy of activation for the aggregate drying process may be expected to be greater for the drying of whole grains than for crushed grains, which is the case.

When a liquid body is subjected to intense sonic vibrations the process of cavitation or "cold-boiling" occurs. Several authors have discussed cavitation in sonic fields, and Hueter and Bolt (1955) present a very good discussion of the basic aspects of the cavitation process. In the dilational phase of the sound wave the negative pressure may be sufficient so that the thermal agitation of the molecules may override the cohesive forces of the liquid. In water, these cohesive, or Van der Waal forces, are very high, of the order of 10^3 atmospheres. However, the presence of nuclei or weak points within the liquid will cause the rupture threshold to move to much lower values.

When a liquid film is moving over a surface it may therefore be expected that even small vibrations may enhance the breaking of the bonds required for the occurrence of the process. In this way the sonic field may enhance the drying of materials by diffusion processes.

Unfortunately, the results of the investigations do not lend to a quantitative analysis; although, it is of interest to examine the ratio of the activation energies for sonic and conventional drying E sound/E no-sound. These ratios are:

0.64-----whole wheat

0.49-----crushed wheat

0.55-----crushed corn

By assuming that the same number of bonds must be broken per molecule for both sonic and conventional drying, it could then be concluded that for crushed material only about one-half of the energy is thermal energy and the remainder is mechanical energy. For whole wheat the mechanical energy of the sound accounts for about 35% of the total energy required. The greater influence of sound on the crushed material may be assumed to be due to the ability of the sound to penetrate through a greater portion of the crushed particles. The impedance match may also be better between the airborne sound waves and the crushed material than it is between the sound waves and the whole material.

Figure 20 shows that for shelled corn an ordinary Arrhenius plot does not appear linear over the temperature range of these investigations. Instead, a greater increase in the rate constant is apparent above 175°F. than would be expected for an Arrhenius plot. When whole kernels of corn are dried at high temperatures irreversible processes occur which are revealed by "stress cracks" after the kernals cool. Presumably, these changes are such that drying is enhanced.

While the experimental energy of activation relates the change in the rate constant with changes in the temperature, the value of the rate constant is also dependent upon the entropy of activation as expressed in equation (17). Table 5 is a list of the entropy of activation for the several drying processes.

Table 5. The entropies of activation (B/lb-mole ^{o}F) for the drying of whole wheat, crushed wheat, and crushed corn.

	No Sound	Sound
Whole wheat	-55.6	-61.6
Crushed wheat	-63.9	-67.2
Crushed corn	-67.4	-69.1

These quantities are relatively large negative values for the activation entropy. Qualitatively, this indicates that the activated species must contain considerably more order than the inactive species. This is also the case when a reaction occurs involving several molecules. During surface diffusion several molecules may be required to form the activated species. Thus, the values of the activation entropies tend to substantiate the conjecture that surface diffusion accounts for a majority of the process. The larger decreases in entropy which occur for crushed materials also suggest that the activated species for these materials may be more complex than for the whole materials. In addition, the application of the sonic energy resulted in a further decrease in entropy of activation. This may be expected since sonic energy is a more ordered form of energy than thermal energy. Due to the lack of a knowledge of the nature of the activated species one cannot make precise statements regarding the significance of the activation entropies; however, the values determined lead to a substantiation of the theory suggested by the activation energies.

The ground material dries more rapidly when sound is applied. This was shown in Table 4. The very rapid decrease in moisture content within the first two minutes is due to the rapid disappearance of the moisture on the external surface of the ground particles. The grinding process greatly increases the total external surface. In the initial drying phase of the ground material the application of sound resulted in marked increase in the degree of drying, as accounted for by the theory of Boucher. As the drying time increased to one hour and the moisture content became very low, the application of sound still resulted in lower moisture values. In this region the sonic vibrations affect the cohesive forces between the molecules of water and the dry material. This shows the advantage of using sonic energy when very low moisture contents are desired in practical drying.

SUMMARY AND CONCLUSIONS

The application of high intensity airborne sonic waves may be used to enhance drying processes. From a practical standpoint, the time required to reduce the moisture content of wheat and corn grains from about 30% moisture content to a level below the critical value can be decreased by up to 50% when sonic energy is applied.

Products such as wheat and corn obey a semi-logarithmic drying law, which is indicative of a first order reaction rate, for both conventional and sonic drying. The variation of the rate constant with the reciprocal of absolute temperature indicates that the sonic drying requires a lower energy of activation than the conventional drying processes.

The mechanism of the drying process for these materials is primarily surface diffusion as evidenced by the low values of the activation energies. By the process of cavitation, the cohesive forces between the water molecules are reduced by the sonic vibrations, and hence, the movement of the water layer is enhanced.

Finely ground material dries very rapidly by both sonic and conventional drying. The increase in the drying rate due to sound in the early portion of the drying period is due to the cavitating effect of the sound waves on the surface moisture. When ground material was dried for extended periods of time the moisture content for sonic drying was lower than for conventional drying. This was presumably due to the rupture of the adsorbed water film within the particles when sound was applied.

When compared with thermal energy, sound is a noble energy form. Therefore, in practical drying processes sound would not be used if high temperatures would not have a deleterious effect on the product quality. On the other hand, if products are heat sensitive, the use of sonic energy provides a method of increasing the drying rate.

Therefore, it may be concluded that:

(1) Both sonic and conventional drying of cereal grains may be represented by a first order reaction process.

(2) Sonic drying requires a lower activation energy than conventional air drying.

(3) The mechanism of drying in wheat and corn grains can be explained by surface diffusion.

(4) Sonic energy is a relatively noble energy form, but has practical use for difficult drying problems where low temperatures are desired.

RECOMMENDATIONS FOR FUTURE RESEARCH

Sonic drying has been investigated to only a limited extent, and the investigations reported here can only be considered a small portion of the potential research in this area. A valuable contribution would be made to this field if basic information could be obtained on the mechanism of sonic drying. This would require homogeneous materials of regular shape for which practically all of the physical properties are known. Such information could lead to some prediction of the affect of sound on the drying characteristics of less homogeneous material.

From a practical standpoint, information regarding the side effects of the sound on the material being dried would also be necessary. For most materials, these side effects are probably not great; however, certain material may be sensitive to the sonic energy.

Combining sonic drying with other drying processes such as freeze drying, spray drying, and foam-mat drying may also lead to fruitful results. Again, a better understanding of the mechanism of sonic drying would be helpful in appraising the relative increase in drying rates when sound is used in conjunction with other drying processes.

In all cases, it is important to remember that drying is a complex process and failure to obtain sophisticated conclusions must not be interpreted as a failure of the investigation.

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