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Soil Moisture Tension and Ice Segregation

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This paper deals with some of the fundamental aspects of frost actions in soils. It is suggested that the freezing point depression at the ice-water interface determines the soil moisture tension which acts as the driving force during frost heaving. The dimensions of the soil pores are responsible, at least in part, for the induced freezing point depression when a saturated soil specimen is frozen unidirectionally.

Moisture tensions in excess of 8,000 cm of water have been measured in closed systems containing fine-grained soils after heaving approached zero, but are much less in coarse-grained soils. Blended soils covering a wide textural range as well as fine Ottawa sand were used to prepare a series of test specimens. In general, the experimental findings are in harmony with the suggested theory.

THE PROBLEMS arising from frost action are well known to the soils engineer concerned with the construction and maintenance of highways and airports. The destructive action falls into two fairly distinct categories. One involves uniform or differential frost heaving as a result of ice segregation; the other concerns the loss of bearing strength when thawing occurs. A soil which exhibits either one or both of these conditions is known as a frost-susceptible soil.

The growth of ice lenses results from the migration of water to the freezing zone, from either a high water-table or the unfrozen portion of the soil which reduces its moisture content. The whole process of frost heaving, therefore, depends on the development of tension in the soil moisture at the freezing plane which acts as the driving force for moisture migration.

An investigation was undertaken to determine the dominant characteristics of the soil system responsible for the development of a moisture tension at the freezing plane. This paper reports soil moisture tension measurements under controlled freezing conditions for a number of soils, and also suggests a mechanism which is compatible with the experimental findings.

METHODS AND MATERIALS

Description of Frost Action Apparatus

The temperature conditioning apparatus has already been described (1). A modification was made to the frost cell containing the soil specimen to reduce further the frictional resistance to heaving. In place of a continuous sleeve around the upper portion of the soil specimen, a stack of lucite rings, each approximately ¼ in. in length, was used. A photograph of the frost cell is shown in Fig. 1. A sectional drawing of the frost cell incorporating the modification is shown in Fig. 2.

The temperature conditioning of the soil specimen is achieved by circulating temperature-controlled mixtures of ethylene-glycol and water through the compartments shown. The inlets and outlets to the compartments are set obliquely to the periphery of the cell but parallel to each other to facilitate the circulation of the cooling liquids, thus providing good heat exchange.

The frost cell was designed to give both a sharp temperature gradient at the frost line and a reasonably constant temperature over the lower two inches of the soil specimen. In this way, thermally activated vapour diffusion would be reduced to a minimum over the lower portion of the specimen. The characteristics of the moisture flow are interpreted in terms of liquid flow, recognizing some contribution to the total flow in the vapour phase.

Description of Soil and Sand Samples

The soil samples used in these experiments consisted of artifically blended soils.

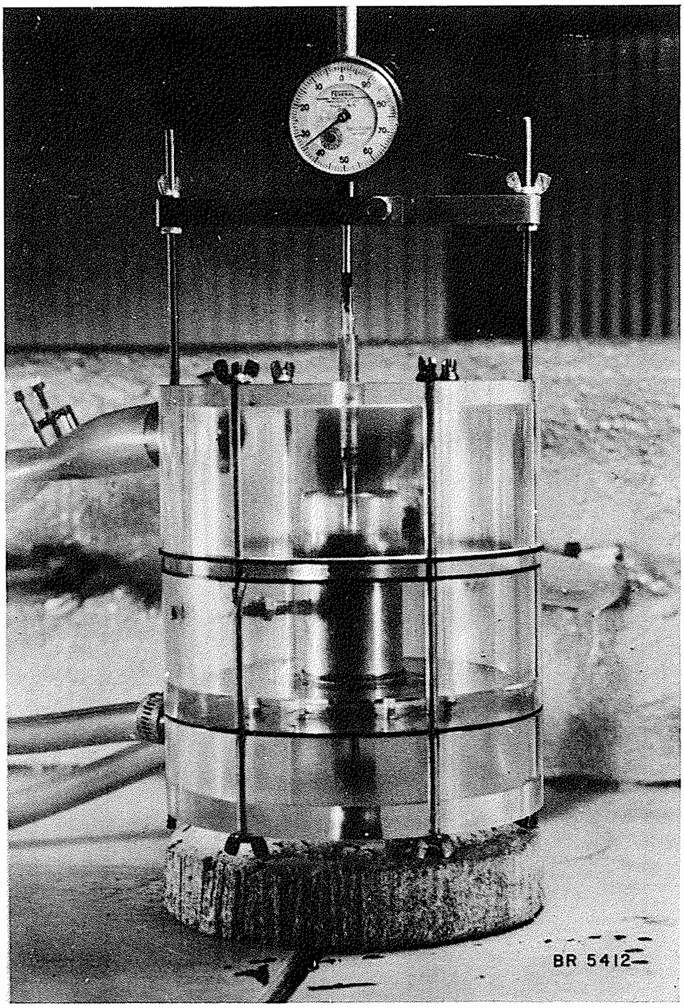

Figure 1. The frost cell in operation.

Leda clay was dried, crushed, then passed through a 100-mesh sieve and thoroughly mixed in an end-over-end shaker. This mix is designated as sample No. 4. Samples No. 1, 2, and 3 consisted of Leda clay and various proportions of Ottawa sand passing a 100-mesh sieve. Sample No. 6 was a mixture of Labrador silt and Leda clay. Samples No. 5 and 7 were composed solely of ground Ottawa sand; they differed in particle size only. Sample No. 5 was the portion passing the No. 325 sieve; No. 7 consisted of sand in the particle size range between the No. 200 and No. 325 sieves.

In all cases, the Ottawa sand was acid washed subsequent to grinding and finally washed with distilled water until the pH reaction of the filtrate was neutral. The hydrometer analyses for the soil samples are shown graphically in Fig. 3, with the exception of the two sand samples. Table 1 gives the Atterberg Limits for samples 1 to 4.

Specimen Preparation

The cylindrical specimens, $1\frac{5}{16}$ in. in diameter and approximately 3 in. long, were prepared directly in the frost cell in $\frac{1}{2}$-in. layers at the air-dry moisture content. Sample No. 4 was prepared at a density of 1.26 gr/cm^3 since this was the highest density

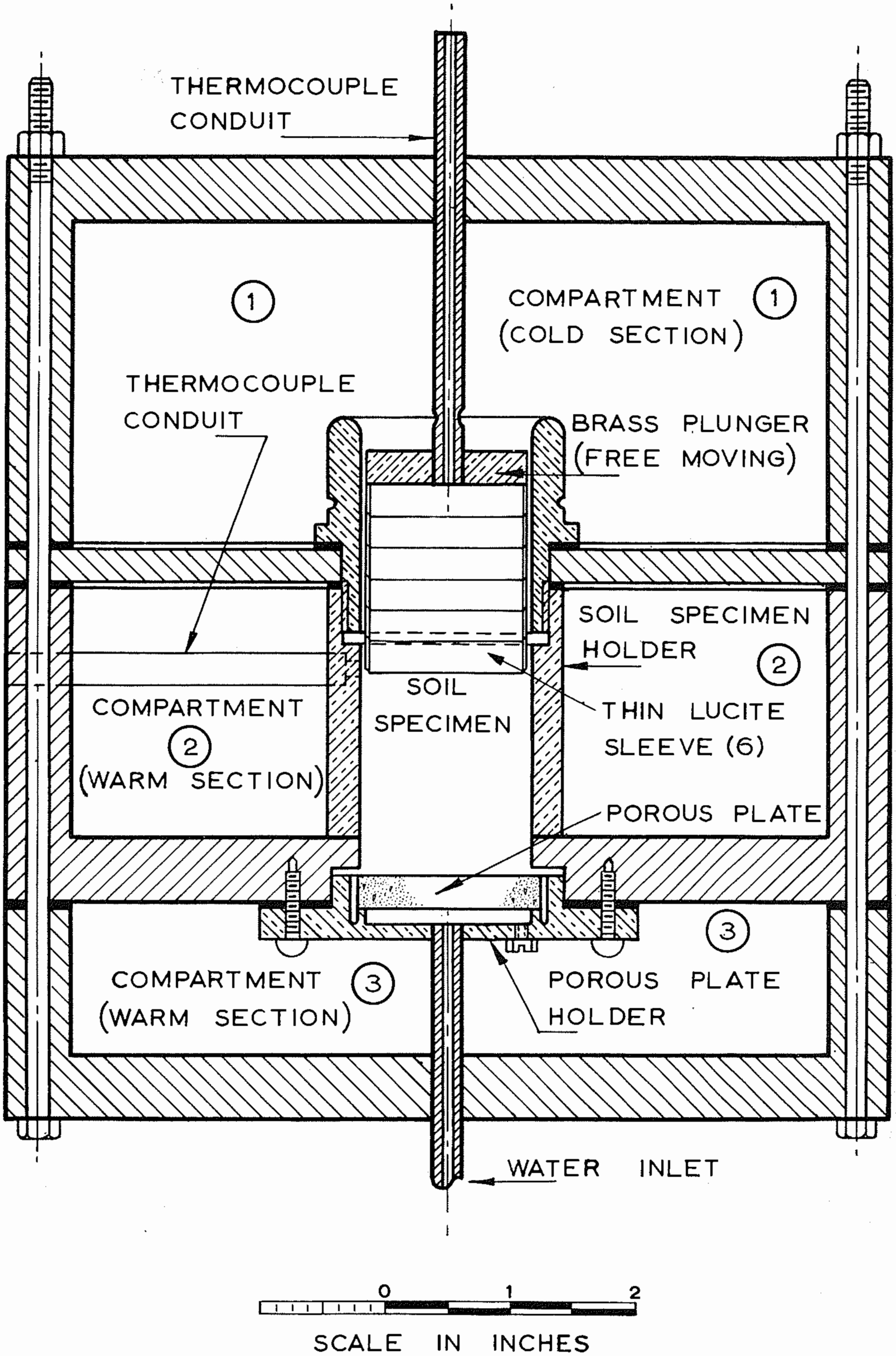

Figure 2. Section through frost cell.

TABLE 1

ATTERBERG LIMITS FOR THE FOUR FINE-GRAINED SOILS USED IN THE FREEZING EXPERIMENTS

Sample No.	Liquid Limit	Plastic Limit	Plasticity Index
1	23.7	16.1	7.6
2	33.2	20.7	12.3
3	39.0	23.8	15.2
4	54.2	31.8	22.4

TABLE 2

FINAL MOISTURE CONDITIONS OF THE UNFROZEN SOIL
(Temperature of 1.5C on Warm Side and -6C on Cold Side)

Sample No.	Percent Moisture Contents				pF[a]
	1	2	3	Ave.	
4	34.5	35.0	34.8	(34.8)	3.70
3	27.2	27.2	27.2	(27.2)	3.46
2	23.2	23.2	23.1	(23.2)	3.46
1	17.9	18.0	18.0	(18.0)	2.98
6[b]	27.8	28.0	28.2	(28.0)	2.60

[a] log moisture tension expressed in cm of water
[b] average of two experiments

TABLE 3

FINAL MOISTURE CONDITIONS OF THE UNFROZEN PORTION
(Temperature at Warm Side 1.5 C and -3 C on Cold Side)

Sample No.	Percent Moisture Content					pF
	1	2	3	4	Ave.	
4	34.6	34.6	34.6	34.4	(34.6)	3.70
6	26.2	24.9	24.6	25.6	(25.3)	2.66
5[a] sand pass 325	30.5	31.3	31.3	32.4	(31.3)	2.13
7 sand 200-325	31.4	30.9	29.6	30.4	(30.6)	1.5

[a] average of two experiments

TABLE 4

FINAL MOISTURE CONDITIONS OF THE UNFROZEN PORTION FOR SPECIMENS PREPARED OF TWO DIFFERENT SOILS

(Upper Half Containing Ice Lens Consisting of Sample No. 3 and Lower Half as Designated in the Table)

Sample No. (of lower half)	Percent Moisture Content (of lower half)				Percent Moisture Content Sample No. 3 (upper half)	pF
	1	2	3	Ave.		
6[a]	11.3	11.2	11.2	(11.2)	26.8	3.50
5[b] sand pass 325	4.0	4.4	4.3	(4.25)	27.6	3.44

[a] temperature of warm side 1.5 C, cold side -6 C

[b] temperature of warm side 1.5 C, cold side -3 C, and is the average of two experiments

obtainable in the air-dry state. Samples No. 5 and 7 were prepared at a density of 1.5 and 1.6 gr/cm^3 respectively, and all other samples at 1.4 gr/cm^3. No. 30 copper-constantan thermocouples were placed at ½-in. intervals in the bottom 2-in. portion and one was positioned at the top of the specimen. To avoid errors in temperature measurement due to heat conduction along the thermocouple wire, one complete turn equal to the circumference of the specimen was coiled at the level where the thermocouple was placed. Each thermocouple was positioned in the middle of the cylindrical specimen.

After preparation, the specimens were allowed to absorb water from a free water-table, level with the base of the specimen, until no further water was withdrawn from the reservoir.

The pF/Moisture-Content Relationship

The various mechanisms whereby water is held in the soil are still not known with certainty. There appears, however, to be general agreement that, at high moisture contents, moisture is held in the soil by surface tension, and that at low moisture contents the surface forces on the solids play a major role. Irrespective of the actual mechanism, many workers have shown that there is a continuous relationship between pF and moisture content from saturation to oven dryness.

The pF term introduced by Schofield (3), is the logarithm of the length of water column in cm required to give a certain moisture tension, i.e., pF=log h, where h is the water column length in cm. The pF/moisture-content relationship is dependent on the direction from which any equilibrium point is approached. As a result, for any porous material there is usually both a wetting and a drying curve for this relationship. Since the freezing of soil is a drying process, this so-called hysteresis phenomenon need not be further considered.

The pF/moisture-content curves for the soils used in these studies were determined by the porous plate (4) and pressure membrane (5) techniques. In the range pF=0 to

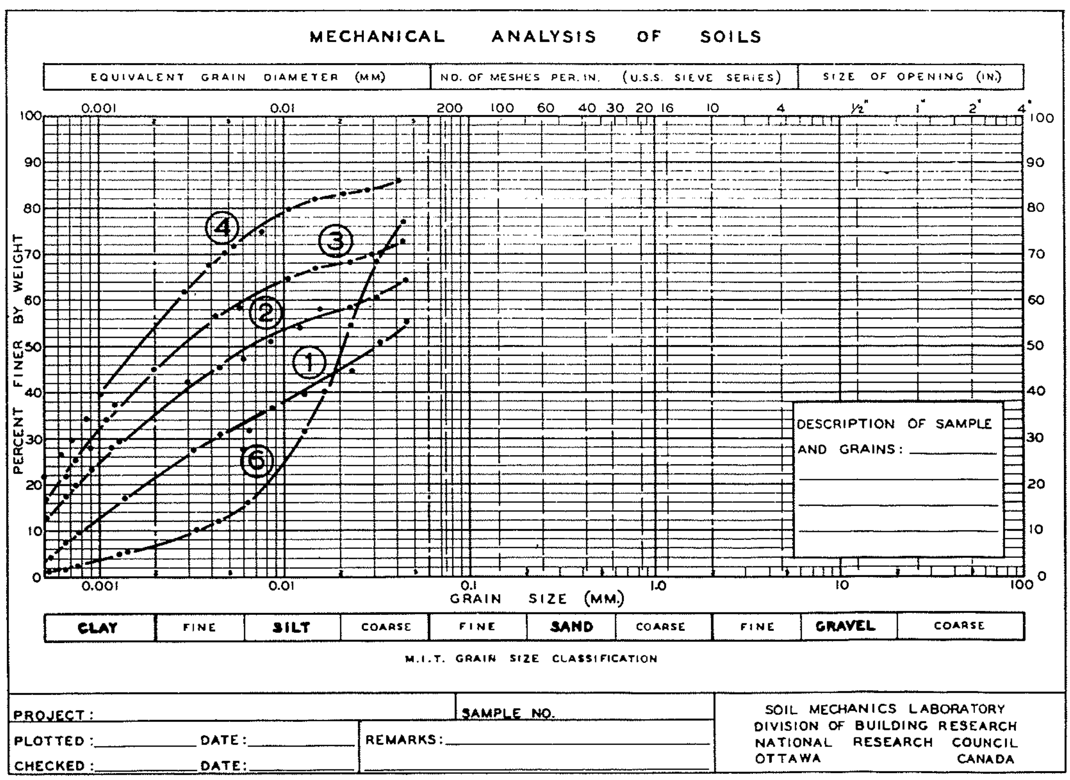

Figure 3.

pF=3, a previously saturated soil sample is placed on a porous ceramic plate across which a pressure difference is maintained. Provided a liquid contact exists between sample and porous plate, the sample will continue to release water until pressure equilibrium is established. The equilibrium moisture content is determined either by oven drying the sample or by measuring the moisture loss from the saturated condition. The limit of usefulness of a porous plate depends on the size of the largest pores. When air begins to pass through the plate, the pressure difference cannot be maintained and other techniques must be used.

For the range from pF=3 to pF=4, the porous plate is replaced by a porous membrane such as Visking sausage skin. In this way, sufficient points of equilibrium may be established to construct a continuous pF/moisture-content curve. The relationship between the largest pressure difference maintainable across a porous plate or membrane and the pore radius is given by the so-called height-of-capillary-rise equation:

$$h = \frac{2s}{rdg} \qquad (1)$$

where h = height of liquid column;
r = radius of curvature of liquid surface, also the radius of pore when maximum curvature is developed;
d = density of the water;
g = acceleration due to gravity; and
s = surface tension for air-water interface.

In the cgs system

$$\text{Schofield's pF} = \log h = \log\frac{(2s)}{(rdg)} \qquad (2)$$

The pF/moisture-content relationships for the soils used are shown in Fig. 4. To evaluate whether the relationship determined at 20 C could be used near the freezing point, some of the determinations were repeated at 1.5 C for three soils over the pF range 1.8 to 3.2. It can be seen that within the normal fluctuations caused by experimental error, the relationship is apparently similar at both temperatures. Hutcheon (6), after completing a literature review on the effect of temperature on moisture retention, has examined the applicability of the equations proposed by Edlefsen and Anderson (7) and Croney and Coleman (8). The equation proposed by the first-mentioned authors can be expressed in the form:

$$H_T = \frac{T_1 T_2 \ (\log H_2 - \log H_1)}{T\ (T_1 - T_2)} + \frac{(T_1 \log H_1 - T_2 \log H_2)}{T_1 - T_2} \qquad (3)$$

where T, T_1 and T_2 = absolute temperatures; and
H, H_1 and H_2 = relative humidity expressed as a fraction and at the appropriate temperature.

When applied to absorption data for Sitka spruce, Hutcheon obtained good correlation between predicted and experimental values at relative humidities below 90 percent.

In the high humidity range corresponding to pF values less than 4.5, limited experimental data showing temperature dependence appear in the literature. The experimental results in Fig. 4 at two temperatures suggest that pF data in the high moisture-content region are applicable over a fairly wide temperature range. The effect of temperature on pF through surface tension was shown to be small by Croney and Coleman (8), in the following equation:

$$pF_1 - pF_2 = \log T_1 - \log T_2 + \log d_2 - \log d_1 \qquad (4)$$

where T_1 and T_2 = surface tension of air-water interface at two different temperatures;
d_1 and d_2 = density of water at two different temperatures. The equation predicts a change of approximately 0.10 units for a 10C change in temperature.

Sample Freezing Procedure

Upon completion of the water absorption process in the soil samples, the circulation

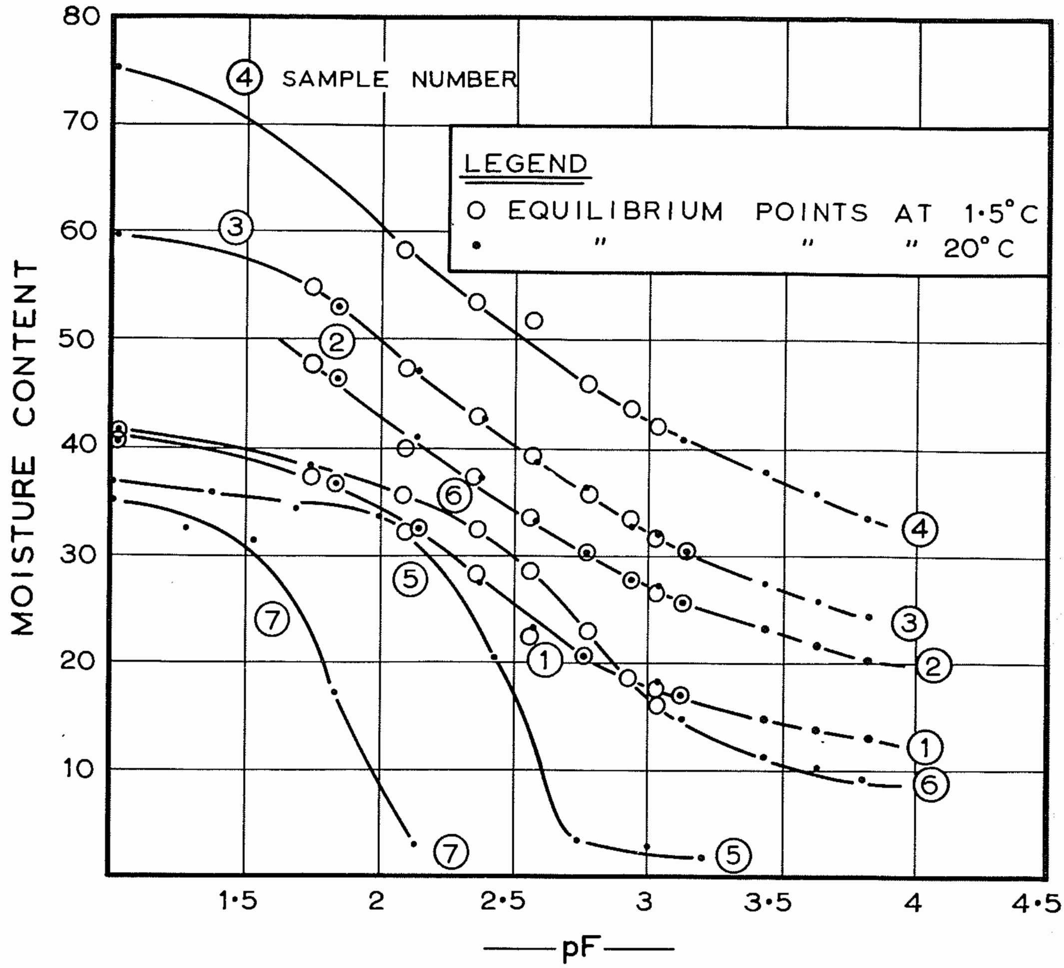

Figure 4. The pF/moisture-content relationship.

of the temperature conditioning fluid (1. 5 C) was started through compartments 2 and 3. When the specimen reached temperature equilibrium with the circulating fluid in compartments 2 and 3, circulation was started through compartment 1 using fluids from the second tank which were delivered to the compartment at either -3 or -6 C, depending on the particular experiment.

Crystallization was mechanically induced when the temperature at the top of the specimen indicated supercooling of approximately 2 C. An extensometer was mounted on the cell to measure the movement of the plunger which rested on the top of the specimen. The system was permitted to operate as an "open" system, with a free water-table at the bottom of the specimen, until the maximum heave rate under these conditions had been attained. During this period the rate of moisture flow into the specimen and the measurements of heave were obtained. After the establishment of the rate of maximum heave, the external source of water was removed. Further heaving occurred as a result of moisture loss from the unfrozen portion. The system now operated as a closed system.

When the extensometer readings indicated that heaving had almost ceased, i. e., that the heaving rate was approaching zero, the apparatus was dismantled and the unfrozen portion of the specimen was sectioned into ½-in. layers for determinations of moisture content. In soils where a high pF was induced a small residual amount of heaving was still evident at the time of sampling. This was attributed partly to vapour flow. With the pF/moisture-content relationship known for each soil, the soil-moisture/tension induced by ice segregation could be obtained indirectly from the moisture content of the sample. The sampling procedure was performed as rapidly as possible to minimize redistribution of moisture. In the light-textured soils, a small amount of melting seemed to occur at the ice-water interface. This was not considered serious enough to invalidate the conclusions.

Experimental Results and Discussion

In all the experiments discussed in this paper, the temperature of the temperature-conditioning liquids was not altered after the experiment was started. The freezing plane was allowed to penetrate naturally into the specimen. At the conclusion of the experiments, the length of the unfrozen portion was approximately 1.75 in. when the temperature on the cold side was held at -6 C. With a cold side temperature of -3 C, the length of the unfrozen portion was about 2 in.

Although the frost cell was designed to give a constant temperature over the unfrozen portion, this objective was not completely achieved. Two typical temperature distributions are given in Fig. 5 for sample No. 6 at the two different cold-side temperatures. In general, the temperature distributions were the same for all samples. There were, however, minor differences due to swelling which displaced the thermocouples to some extent. The temperature distribution in the frozen layer was not measured since it was found that, as the thermocouple wires were frozen in, there was some interference with the heaving process. It can be seen that the largest temperature gradient in the unfroz-

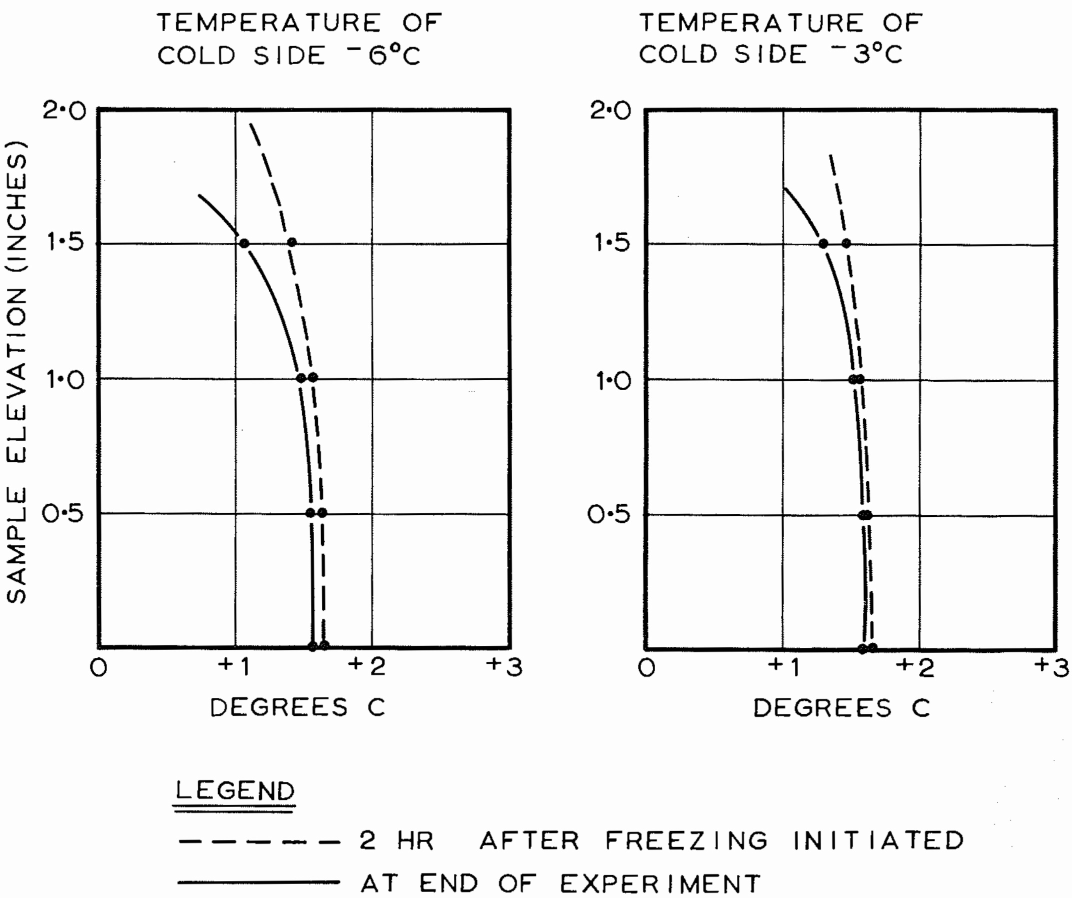

Figure 5. Temperature distribution in unfrozen portion of sample 6 for two different temperatures at the cold side.

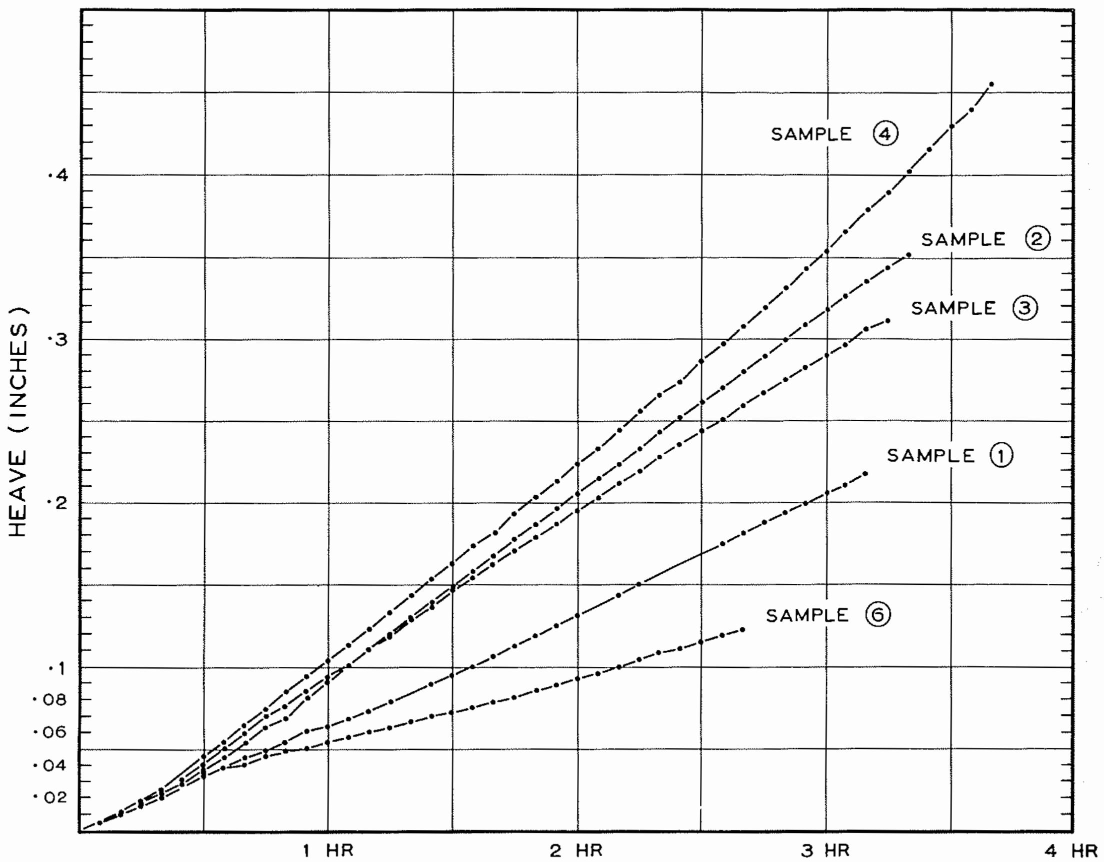

Figure 6. The amount of heave versus time with free water table at the base of the specimen (temp. of warm side 1.5 C, cold side -6 C).

en portion of the sample occurred immediately beneath the freezing zone. No influence of this temperature gradient upon moisture contents could be detected, i. e., the moisture contents were essentially the same in all sections of the unfrozen sample at sampling time.

Figure 6 shows the heave-time relationship, plotted at 5-min intervals, when the cold side was maintained at -6 C and with a free water-table at the base of the specimen. There appears to be a relationship to soil texture evident in the comparison of Fig. 3 with Fig. 6. The finer the texture of the sample, the greater is the rate of heave.

The open system part of the experiment was terminated when it became evident that the maximum rate of heave had been obtained. (In the case of sample No. 4, the maximum rate was probably not obtained before the external source of water was cut off, as indicated by the shape of the heave-time relationship.) The end of each curve marks the time when the external source of water was cut off; further heaving could occur only as a result of internal water loss. It was then possible to determine the moisture tension induced in the unfrozen portion of each soil due to a freezing plane.

Figure 7 shows the results of the heave measurements during the entire experimental period which were terminated when the heave rate approached zero. As already noted, a very small amount of heaving was still evident in the heavy-textured samples when the experiment was terminated; a contribution attributed, in part, to vapour flow. Figure 8 shows that the dominating characteristics of frost heaving are determined by the soil at the ice-water interface. These experiments were carried out in the same way as those summarized in Fig. 7. The lower curve shows the heave versus time relationship for sample No. 6. The results shown in the upper curve are for a specimen containing sam-

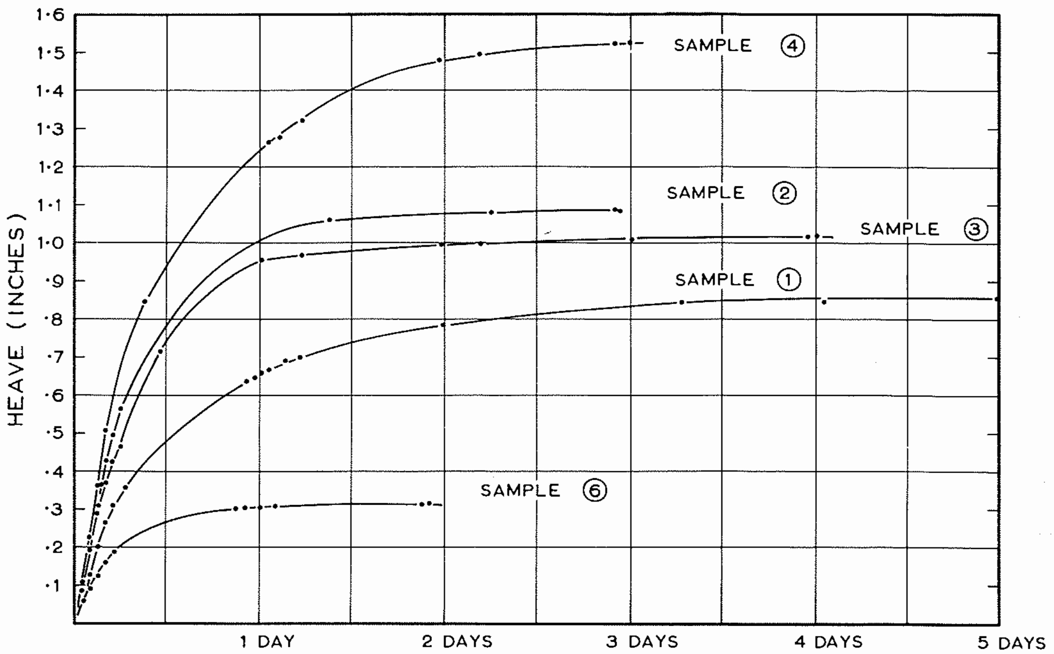

Figure 7. The amount of heave versus time for a number of different soils (temp. of warm side 1.5 C, cold side -6 C).

ple No. 6 in the lower half and sample No. 3 in the upper half. The freezing zone remained in the portion of the specimen containing sample No. 3, but when heaving approached zero, it had penetrated to within ½ in. of sample No. 6. The upper curve shows that the heaving characteristics were dominated by the soil at the ice-water interface (sample No. 3), and that sample No. 6 acted merely as a transmission zone for

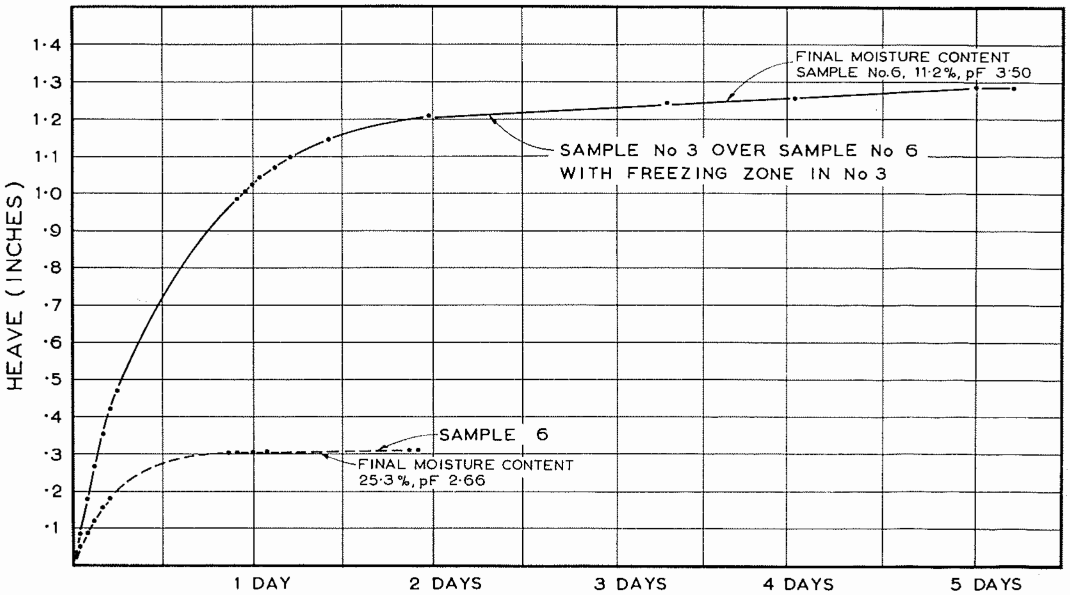

Figure 8. The amount of heave versus time showing the added heave from sample No. 6 when the freezing zone is located in a layer of finer textured soil (No.3).

moisture flow when it operated as an open system, and as a source of water supply in a closed system.

Similar experiments with other soils confirmed these conclusions. The coarsest sample used as a transmission zone was sample No. 5 (the portion of finely ground sand passing the No. 325 sieve). These experiments indicate the ability of coarser-

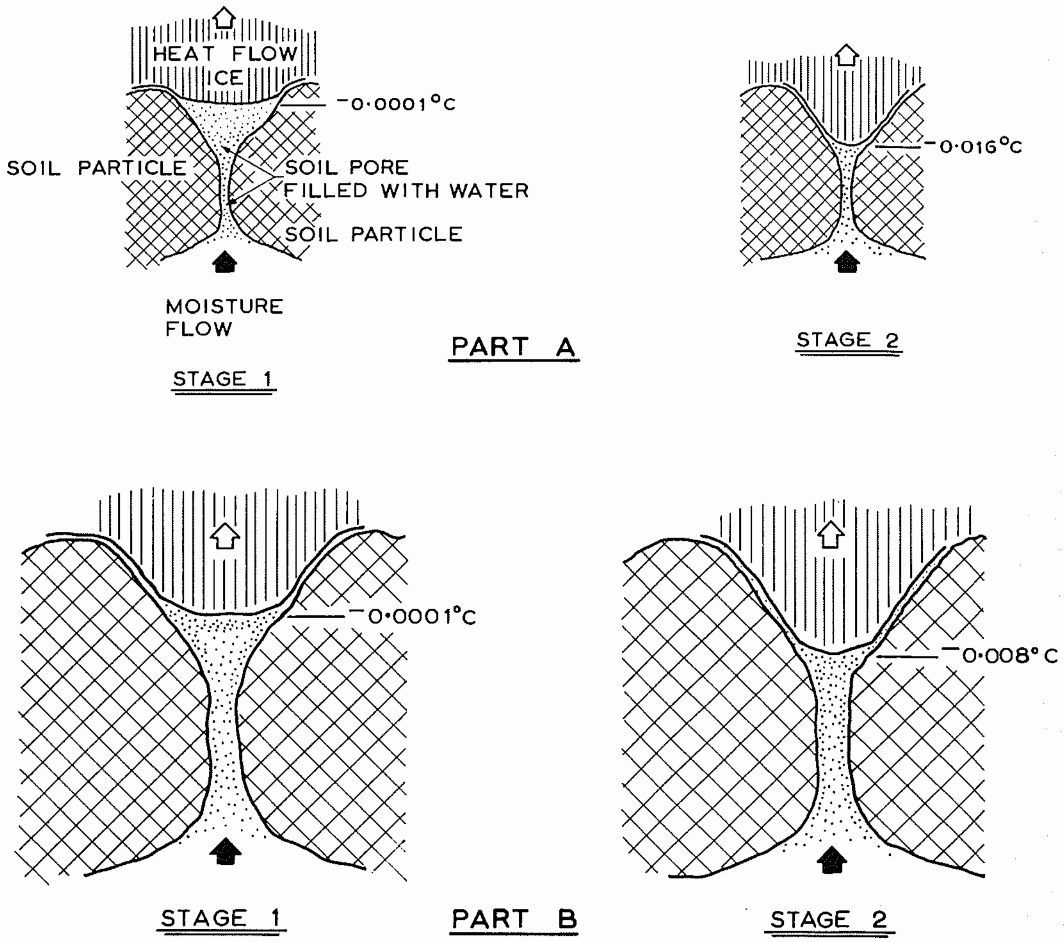

Figure 9. An enlarged schematic diagram of two different pore sizes showing stages in the movement of the ice/water interface with assumed freezing point depressions.

grained soils to act as effective transmission zones or as sources of water supply, if the frost line intercepts small layers or islands of heavier-textured materials.

Tables No. 2, 3, and 4 show the moisture contents and corresponding moisture tensions when heaving approached zero in a closed system. The numbers along the top of the table designate the position of the layers in the soil column, No. 1 being the bottom ½-in. layer, No. 2 the second ½-in. layer, etc. It should be noted that the final moisture content was essentially constant throughout the unfrozen portion of the specimen for any given soil. The moisture contents of the unfrozen portion, which are essentially the same in all layers, reflect the moisture tension induced by the freezing zone through the liquid phase. The last column gives the moisture tension in terms of Schofield's pF. This pF value was obtained from Fig. 4, which gives the pF/moisture-content relationship for each sample.

Table No. 3 gives similar data to that in Table No. 2, but for a temperature of -3 C on the cold side instead of -6 C. A comparison of the results for samples No. 4 and 6 in Tables 2 and 3 shows that the final tension in the soil moisture is not influenced by the temperature on the cold side.

In the case of the lower temperature, the frost line penetrated a little more than ¼ in. further. The transient soil moisture tensions however, appear to be dependent on rate of heat removal.

When the specimen was composed of layers of two different soil samples, the moisture tension induced was shown to depend on the soil type in which the freezing zone was located. These results are shown in Table No. 4. By referring the moisture contents in Table No. 3 to Fig. 4, it can be seen that the moisture content of sample No. 6 was drawn down to that corresponding to the pF of the soil above it which contained the freezing zone, in this case sample No. 3. A pF of approximately 2.6 was induced when the entire specimen consisted of sample No. 6. This sample has an average moisture content of 25.3 percent at this pF. With the freezing zone in sample No. 3 above it, the equilibrium moisture content was 11.2 percent, which corresponds closely to the pF induced in sample No. 3. Similarily for the sand, in sample No. 5 (passing No. 325) the moisture content was 31.3 percent (Table No. 3), but with the freezing zone in sample No. 3 (Table No. 4), its moisture content was reduced to 4.25 percent. According to the pF/moisture-content relationship for this sample, the moisture content should have been reduced to about 2 percent at a pF of 3.44. That this failed to occur may have been due to a number of causes, such as sample No. 3 shrinking away from the sand or that, at this very low moisture content, transmission of moisture was very low and the equilibrium conditions were not quite attained. Nevertheless, the amount of moisture released during frost action in closed systems appears dependent on the pF induced at the ice front and this, in turn, depends on soil type. The finer the texture of the soil, and consequently the smaller the pores, the greater the pF induced. The pF's induced when heaving approached zero ranged from 1.5 for a sand (sample No. 7) to 3.7 in Leda clay (sample No. 4). These tensions expressed in psi are 0.98 and 120 respectively.

In the light of these experimental results, attention is now invited to some of the significant features of a soil system which appear to be dominant in the development of soil moisture tensions during unidirectional freezing.

Suggested Mechanism in the Development of Tensions

It will be recalled that the free energy of a system always decreases in a spontaneous change. If the temperature is lowered in an ice-water system originally in equilibrium, the specific free energy of each phase at constant pressure will change with temperature according to the following equation: $df = -sdt$. The phase with the greatest specific entropy, s, will have the greatest free energy for a given decrease in temperature. The spontaneous change will be the conversion of water to ice which is made possible by the temperature drop.

The size of a stable spherical crystal of a solid in its own melt is known to be temperature dependent. The relationship is given by the following equation used recently by Sill and Skapski (9):

$$\Delta T = \frac{2T_m \, \sigma_{sl}}{r \, \rho_s \, Q_f} \tag{5}$$

where r = radius of curvature of the crystal (critical size);

ρ_s = density of the solid;

σ_{sl} = interface tension between solid and liquid;

Q_f = heat of fusion;

T_m = the temperature of melting at zero curvature of the solid/liquid interface; and

ΔT = the depression in freezing point below T_m.

Application of this relationship to the unidirectional freezing of saturated salt-free soils in a closed system, shows that the temperature at which an ice-water interface can advance into a soil pore depends on the size of the pore, i. e.,

$$\Delta T \propto \frac{1}{r}$$

(where r is now considered to be radius of the soil pore). Thus it can be seen that the radius of the soil pore sets the size of the advancing tongues of ice. It is then clear that the advance of the ice-water interface will occur at a lower temperature in a soil containing mostly small pores than in a soil in which large pores predominate.

Winterkorn (10) has used the second law of thermodynamics to give a quantitative measure of the free energy or maximum amount of work available during frost action. More recently Jumikis (11) has compared these theoretical values with experimental results obtained under dynamic conditions. The law is used here to describe the amount of work solely as a result of the depressed freezing point. Under these circumstances, the amount of energy available is directly proportional to the freezing point depression and gives a method of evaluating ΔP_w, the induced soil moisture tension.

$$W_{max} = Q_f \frac{(T_2 - T_1)}{T_2} = \frac{Q \Delta T}{T_2} = V_w \Delta P_w \qquad (6)$$

where W_{max} = maximum work available or free energy;

Q_f = latent heat of fusion;

T_2 = the freezing point of free water and is equal to T_m in equation (4);

T_1 = the temperature at which freezing is occurring at the water-ice interface;

ΔT = freezing point depression;

V_w = specific volume of water; and

ΔP_w = change in soil moisture tension at equilibrium as a result of ΔT.

It has been assumed in Fig. 9 that a microscopic section of the soil-water-ice system can be represented schematically for purposes of clarity. These sketches are not to scale and are merely intended to help to illustrate the pertinent features under discussion. Stage 1 in both sketches shows that the water is freezing close to 0 C in the wide portion of the soil pore. In order for the ice to invade the narrow neck, a certain freezing point depression must take place, i. e., the 0 C isotherm must advance ahead of the freezing plane to fulfill the requirements of equation (4). In the case of the larger pore in Stage 2 of Fig. 9B, ΔT is much smaller at the soil-water interface. Ultimately, as the size of the pore considered is increased to the size that exists in coarse sand, the temperature of the freezing plane will be very close to 0 C and a correspondingly small tension would be induced in the soil moisture. In the period between Stages 1 and 2, a sufficient flow of water to the freezing zone may retard the advance of both the zero isotherm and the freezing plane. In this situation the rate of heat removal from the freezing plane will largely control the rate of heave; it is at this stage that ice lens growth takes place. When the flow of moisture is restricted, the temperature will drop at a given point in the soil, larger pores may be emptied through connecting channels, and the process will continue in the soil below.

A phenomenon which is also possible is that crystallization may occur in soil pores below the existing ice-water interface before the water freezes in the smallest pores. Many cases of unfrozen layers have been reported in the literature. The invasion of ice by way of large surrounding pores may also occur and the complete development of freezing point depression, demanded by the smaller pores, may be interrupted. These considerations generally appear to account for the high moisture tensions developed in fine-grained soils and the relatively small tensions in coarse-grained soils which were originally saturated identically and subjected to the same temperature conditions.

There are still many uncertainties that prevent the useful application of this concept. In equation (5), which relates the minimum radius of curvature of the solid-liquid interface to freezing point depression, only approximate values for interface tension are available. It is indeed uncertain whether the equation can be rigidly applied to minute pores where the force fields occupy a considerable portion of the pore or when the soil moisture is under tension in unsaturated systems. This point is discussed in some detail by Gold (12). While it is generally true that the dimensions of the soil pores decrease with decreasing particle size no exact mathematical solution of pore size or pore size distribution is possible and, in the development of moisture tension, the existence of some large pores may tend to nullify the effect of the smaller pores.

Pore sizes in swelling soils will vary with changing moisture content. Since the freezing phenomenon is a drying process, the soil moisture tension induced in this way will tend to decrease pore size. The freezing point depression due to dissolved solids in the soil moisture is superimposed on the depression due to pore size. Since salts will tend to accumulate during the flow of moisture to the freezing front, a satisfactory allowance for this effect is problematic.

CONCLUSION

The experimental results herein presented show that: (a) for the materials at the densities used, the rate of moisture flow due to unidirectional freezing in an open system is greatest for the heaviest-textured soil and least for sand; this could occur only as a result of higher moisture tensions induced at the freezing zone; (b) the moisture tension developed in a closed system appears to be largely dependent on soil texture; the greater the proportion of fines, the higher the moisture tension, and thus, more moisture is made available for heaving; (c) at two different cold-side temperatures, the moisture tension of the unfrozen portion was the same but caused only a shifting of the freezing plane; and (d) in specimens prepared in layers from two different materials, the moisture tension in the unfrozen portion is dependent on the material in which the freezing zone is located.

Theoretical considerations show that moisture tensions can develop in soils during freezing as a result of freezing point depressions. The freezing point at the ice-water interface and its radius of curvature must decrease as smaller pores are invaded by ice. As a consequence, higher tensions are developed in soils with small pores than with large pores. This appears to be in agreement with the experimental results presented.

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