Production of Maltose Sirups from Granular Wheat Flour

AARON EFRON[1] AND R. H. BLOM[2]

Northern Regional Research Laboratory, U. S. Department of Agriculture, Peoria, Ill.

Experimental work is described concerning the preparation of maltose sirup from granular wheat flour which has been extracted with water prior to conversion. Several conversion agents were tested, and the effects of these materials on the quality of the converted liquors are reported. The filtration of wheat mashes converted with enzymes is difficult, and various processing techniques were investigated to improve this rate. Bentonite was employed in the refining of maltose sirups and appears to have a strong affinity to wheat protein.

THE continuing severe shortage of sugar in the United States has intensified industrial interest in sirup sweeteners of the maltose type, and concurrent restrictions on the use of grains for alcohol and alcoholic beverage production have prompted several brewers and distillers to convert to the production of maltose grain sirups. A recent report (*1*) estimated this new production of maltose sirup to be 200–250 million pounds annually; this may be compared with the total production, in 1945, of 1.9 billion pounds of corn sirup, and with the estimated deficit of 1 billion pounds for 1946.

The conversion of breweries and distilleries to the manufacture of sirup has been relatively simple; the only necessary equipment these plants might lack are filters and evaporators. The processing of grain to yield a bland sirup has been handicapped, however, by the sparsity of available information regarding the production of a grain sirup by malt or enzyme conversion. The small scale work reported here was undertaken to increase and advance the fund of information available to processors.

The objects of this investigation were to determine the effects of various refining agents in the production of a bland, light-colored sirup; and to develop a process for the production of sirup from granular wheat flour by enzyme conversion. Granular wheat flour was chosen because of its availability during the war and because it appeared to be a likely raw material for the production of bland sirup.

PROCESS

The manufacture of a maltose sirup was considered as comprising four distinct steps: (*a*) cooking the grain to render the starch accessible to the conversion agent; (*b*) converting the starch to maltose and glucose by means of an amylase-containing agent; (*c*) clarifying the mash; and (*d*) refining and concentrating the thin liquor to produce the sirup.

The cooking is a relatively simple step, involving the gelatinization and liquefaction of the starch by heat. In all the work reported here, the cooking was conducted for 10 or 15 minutes at the temperature corresponding to steam at 20 pounds per square inch gage. It was necessary to limit the concentration of granular wheat flour in the slurry to 26% to maintain a sufficiently fluid mass in the cooker. When it was desirable to cook at a higher concentration (for example, when the subsequent dilution was severe) a small portion, 5–10%, of the conversion agent was added before cooking in order to dextrinize and liquefy sufficient starch to retain the required fluid characteristic. In this manner slurry concentrations as high as 35% could be used. When ground whole grain, either wheat or corn, was used, the slurry concentration was limited to a maximum of 30%.

The conversion of starch to maltose and glucose is accomplished by a diastatic agent such as malt, which is generally used. Malt, however, is relatively expensive, and during the war period, when a supply was not available to fill all demands, other sources of amylase were developed. Two of these, mold bran (*2*, *5*) and fungal amylase liquor (*7*), as well as malt, have been used in this work.

The malt used was a typical distiller's barley malt in flour form, and of high diastatic activity. It has been recommended (*3*) that green (unkilned) malt be used to obtain a higher diastatic activity with lower coloring; green malt, however, presents shipping and storage problems which appear to make its use impractical unless the sirup factory is to be operated in conjunction with a malting house.

Mold bran is produced by growing the organism *Aspergillus oryzae* on acid-cooked wheat bran and drying the product. It has been used industrially in the production of grain alcohol. The mold bran used in this work was furnished by the Mold Bran Company, Incorporated, Eagle Grove, Iowa.

Fungal amylase liquor is the designation used here for an amylolytic agent developed recently at this laboratory. It is produced at present by growing *Aspergillus niger* in thin stillage from corn mashes which have been saccharified with malt and fermented to alcohol. It appears to be a relatively inexpensive source of amylase for a plant which is producing both grain alcohol and maltase sirup.

The converted mash is clarified most conveniently by filtering through plate-and-frame filters, or possibly rotary-drum filters; both types may be expected to yield a cake which has been sufficiently dewatered to be dried for feed. With granular wheat flour and with ground wheat, however, the filtration is difficult because of the glutinous protein content of the wheat. Considerable effort was made to improve this filtration to the point where it was believed to be practicable.

The thin liquor obtained from the filters was treated with various refining agents to lessen the color and improve the appearance and flavor. It was then concentrated to a heavy sirup in vacuum evaporators. The decolorizing agent usually employed is activated vegetable carbon; the authors found in this work, however, that, in addition to carbon, it was necessary to use bentonite to reduce the protein content to avoid an adverse effect in flavor and clarity.

EXPERIMENTAL EQUIPMENT

The cooker used was an 18-gallon horizontal steel cylinder with suitable openings, a spiral-ribbon agitator, and several steam-injection fittings. The filter press was 12 inches in size, with 1-inch frames, top feed, and open discharge. From seven to ten frames were used for a run, depending on the amount of cake expected. The filter press was fed with a rotary pump driven at a variable speed.

Clarified liquors were concentrated in a glass evaporator operating at 25 to 27 inches vacuum, with steam at atmospheric pres-

[1] Present address, University of Michigan, Ann Arbor, Mich.
[2] Present address, Mold Bran Company, Inc., Eagle Grove, Iowa.

TABLE I. CONVERSION AND CLARIFICATION

Run No.	Pretreatment of Raw Material[a]	Saccharifying Agent		Conversion[c]			Filtration Rate	Compn. of Thin Liquor, % Dry Substance Basis			
		Substance[b]	Relative amt.	Temp., ° C.	pH	%		Reducing sugar as maltose	Dextrin	Protein	Ash
6	None	FM	10% of bill	65	5.8	65.1	Fair	69.0	31.3	2.9	...
20	None	FM	8% of bill	65	5.5	69.4	Fair	78.3	15.2	2.9	...
24	None	FM	8% of bill	65	5.5	72.4	Fair	71.5	22.4	2.9	...
25	None	FM	8% of bill	65	5.9	69.6	Fair	67.4	21.6	2.8	...
28	Steeped 15 hr.	FM	8% of bill	65	5.5	70.1	Good	71.3	19.1	2.7	...
35	Steeped 15 hr. and extd.	FM	8% of bill	65	5.6	73.7	Good	74.8	12.9	1.7	0.81
37	None	WFAL	208 Q-units	60	5.6	73.1	Good	85.5	13.1	3.6	3.20
38	None	WFAL	208 Q-units	60	5.3	77.5	Fair	82.5	25.8	3.5	...
42	Steeped 15 hr.	FM	10% of bill	65	5.2	64.4	Fair	66.6	24.0	2.3	...
43	None	WFAL	121 Q-units	60	5.6	69.9	Good	78.8	22.6	3.1	1.34
44	Steeped 15 hr. and extd.	FFAL	212 Q-units	60	5.2	58.5	Fair	78.6	25.6	2.5	...
47	None	FFAL	185 Q-units	60	5.5	57.1	Good	67.6	33.8	1.5	1.54
48	Steeped 22 hr. and extd.	FFAL	121 Q-units	60	5.4	51.8	Fair	66.9	36.9	1.6	0.81
49	Steeped 22 hr. and extd.	FM	10% of bill	65	5.5	68.1	Good	77.5	16.5	2.0	0.51
52	Steeped 22 hr. and extd.	FFAL	180 Q-units	60	5.5	60.2	Good	79.6	23.9	2.2	...
53	Steeped 22 hr. and extd.	MB and FFAL	3% of bill 109 Q-units	55	5.5	63.1	Fair	75.4	23.4	5.3	0.97

[a] Granular wheat flour was the raw material in all experiments reported except for run 47 in which corn was used.
[b] FM = flour malt; WFA = whole fungal amylase liquor; FFAL = filtered fungal amylase liquor; MB = mold bran.
[c] Time of conversion: run 7, 2½ hours; all others, 2 hours.

sure. The evaporator consisted of several vertical glass tubes sealed into a steam jacket and discharging into entrainment separators. The construction was similar to that of the long-tube-vertical industrial evaporator with natural circulation.

ANALYSES

The principal analytical determinations included in this work were (*a*) reducing sugars calculated as maltose, (*b*) glucose equivalent of the total sugar, (*c*) protein, (*d*) ash, and (*e*) amylolytic activity of the fungal amylase liquors.

Sugars were determined by a modified Shaffer-Hartmann method (*6*). Reducing sugars were calculated as maltose; and total sugars, after hydrolysis, were calculated as glucose. The dextrin contents were obtained by the difference between the total and the reducing sugar values. Since dextrose-maltose ratios were not determined, the dextrin values are approximate only. Nitrogen was estimated by the Kjeldahl method, and crude protein was calculated as N × 5.7 for wheat products or N × 6.25 for corn products. Ash was found by igniting at 525° C.

The amylolytic activity of the fungal amylase liquors was estimated by the method of Olson, Evans, and Dickson (*4*).

CONVERSION AND FILTRATION

The experiments on conversion and clarification are summarized in Table I, which presents the conversion conditions, the rate of filtration, and the composition of the resulting thin liquor.

Cooking and mashing procedures depended on whether malt or fungal amylase was used as the saccharifying agent. For malt conversions, 22.5 pounds of granular wheat flour were mixed with 62 pounds of water in the pressure cooker. The pH of the mixture was adjusted to 5.5 with dilute sulfuric acid and the slurry was heated to 126° C. with live steam. The mixture was held at 126° C. for 10 to 15 minutes; it was then cooled to 65° C., and a slurry consisting of 1.96 pounds of malt in 8 pounds of water was added.

In the fungal amylase conversions, 22.5 pounds of granular wheat flour and 40 pounds of water were mixed and held at 70° C. for 10 minutes. One quart of amylase liquor was added, the pH was adjusted to 5.3, and the mixture was cooked for 10 to 15 minutes at 126° C. The cooked mash was cooled to 60° C., and 4.75 gallons of the amylase preparation were added.

The principal problem was to obtain a satisfactory filtration rate; hence data are given in Table I for only those runs which were reasonably successful in this respect. A filtration rate was designated "good" if it equaled or exceeded 1 gallon per hour per square foot with the formation of a solid cake which filled the frames. The notation "fair" means a rate lower than good, or the formation of a wetter cake, or both. (A filtration rate of 1 gallon per hour per square foot is low and consequently of little significance quantitatively but it is not impractical if a good cake is obtained.) Numerous unreported runs were made in which fresh slurries of granular wheat flour or cracked whole wheat were saccharified with malt or malt extract, and in which the conversion temperature and pH were varied over wide ranges, but invariably these mashes could not be filtered. Heating the mash, after conversion, under various conditions was tried, with poor results. Time and temperature were varied from 10 minutes at 85° C. to 2.75 hours at 100° C. with no significant effect on the rate of filtration.

In an attempt to lower the protein content of the converted liquor, the granular wheat flour was extracted with cold water to remove its soluble protein content. This treatment lowered the protein content approximately 40%. For example, run 24, without extraction, gave a thin liquor with 2.9% protein on a dry substance basis (D.S.B.) whereas run 35, with extraction, but otherwise comparable to run 24, showed 1.7% protein (D.S.B.) in the thin liquor. It was further found that steeping, incident to the extraction, resulted in greatly improved filtration rates for the converted mash, and accordingly, several subsequent runs were preceded by the steeping-extraction treatment, with steeping times ranging from 10 to 22 hours. Of 10 runs thus made, 8 gave practicable filtration rates; in 4 of these 8 runs, the average filtration rate was over 1 gallon per hour per square foot. In general, the longer steeping times resulted more consistently in a practicable filtration. The steeping-extraction treatment resulted in the loss of the free sugar and soluble protein of the wheat flour; this loss was found to average 2 pounds of dry substance per bushel.

In several experiments fungal amylase liquor was used in place of malt. In the earliest of these experiments the fungal amylase liquor was used unfiltered, as it came from the fermenters. It was soon found, however, that converted liquors of lower color and protein could be obtained when the fungal amylase liquor was filtered before use. The fungal amylase liquor, in these experiments, was used in amounts ranging from 5.1 to 12.5 gallons per bushel of grain, or 115–212 Q-units. Q-units are defined as

$$\frac{(\text{gal. fungal liquor used}) \times (\text{units } \alpha\text{-amylase/ml.})}{\text{lb. starch in batch}}$$

The protein content and filtration characteristics were found to be comparable when either malt or fungal amylase was used for conversion. The color and flavor of the converted liquor were always stronger than for comparable malt-converted liquor. The ash contents of fungal conversions were more than twice those from malt conversions.

Several miscellaneous experiments were made to test other raw materials and converting agents. One run was made on

TABLE II. SIRUP REFINING

Sirup No.	From Liquor No.	Age of Liquor	Refining Treatment Agents[a], Amt. (Dry Substance Basis) and pH	Compn. of Sirup, % Dry Substance Basis: Reducing sugar as maltose	Dextrin	Protein	Ash	Appearance	Flavor
8	42	Fresh	1% C and 1% FA at 5.2; 1% C and 1% B at 4.2; SO_2 to 3.0; lime to 7.0	47.7	43.5	0.97	...	Clear, yellow	Pleasant, with sulfite aftertaste
9	42	Fresh	1% C and 1% FA at 5.2; 1% C and 1% B at 4.2; lime to 7.0	73.2	18.6	0.99	...	Slightly hazy, yellow	Pleasant
10	42	2 days	1% C and 1% FA at 5.2; 1% C, 1% B, 0.01% papain, 0.01% $Na_2S_2O_5$ at 4.2; lime to 7.0	55.1	38.4	0.90	...	Clear, yellow	Pleasant, with sulfite aftertaste
11	42	2 days	1% C and 1% FA at 5.2; 1% C, 1% B, 0.02% papain, 0.02% $Na_2S_2O_5$, 0.02% NaOAc at 4.2; lime to 6.0	74.0	23.7	1.06	...	Clear, yellow	As 8 and 10, but more bitter
12	43	Old	1% C and 1% FA at 4.4; lime to 7.0; 1% C and 1% FA at 7.0	76.6	20.0	1.92	1.42	Cloudy, dark	Strong, but palatable
12a	43	Old	As sirup 12, and in addition: 0.01% $Na_2S_2O_5$ and 0.01% papain at 7.0	74.6	23.8	2.40	...	Cloudy, dark	Sulfite taste
12b	43	Old	As sirup 12a, and in addition: 1% C and 1% B at 7.0	79.1	21.8	1.57	...	Hazy, dark	Similar to 12
13	44	Fresh	1% C and 1% FA at 5.3; lime to 7.0; 1% C and 1% B at 7.0	81.3	22.8	1.45	...	Clear, light amber	Pleasant
16	47	Fresh	1.5% C and 1.5% FA at 5.6; lime to 7.0; 1.5% C and 1.5% B at 7.0	71.8	33.2	0.97	1.36	Clear, light amber	Pleasant
17a	48	Fresh	1% C and 1% FA at 5.5; lime to 7.0; 2% C and 2% FA at 7.0	67.0	33.6	0.68	...	Cloudy, light amber	Pleasant
17b	48	Fresh	1% C and 1% FA at 5.5; lime to 7.0; 2% C, FA 2 % and 2% B at 7.0	67.2	35.8	0.40	0.73	Clear, light amber	Pleasant
18a	49	Fresh	3% C, 2% FA and 1% B at 4.7; Na_2CO_3 to 6.5; HCl to 5.2	74.0	18.5	0.66	...	Clear, yellow	Pleasant
18b	49	Fresh	As sirup 18a	67.6	15.8	0.62	...	Clear, yellow	Pleasant
18c	49	Fresh	As sirup 18a, except finished at 4.9 with HCl	73.3	18.1	0.61	0.65	Clear, yellow	Pleasant
19a	52	3 days	2% C, 1% FA and 0.5% B at 5.9; HCl to 5.0; 1% C, 1% FA and 1% B at 5.0	80.9	22.3	0.84	...	Hazy, light amber	Strong, but palatable
19b	52	3 days	2% C, 1% FA and 0.5% B at 5.9; lime to 7.0; 1% C, 1% FA and 1% B at 7.0	78.1	22.0	0.84	...	Clear, light amber	Pleasant
20	53	Fresh	1% C, 1% FA at 5.5; lime to 7.0; 1% C, 1% FA and 1% B at 7.0	88.4	24.0	4.4	1.09	Cloudy, dark	Pleasant

[a] C = Activated vegetable carbon; B = bentonite; FA = refined filter aid.

cracked whole wheat; the results were not significantly different from comparable runs on the granular wheat flour.

Coarse-ground whole corn was converted with fungal amylase liquor in one run (No. 47). The mash filtered readily, as would be expected, because of the coarse grind and the absence of glutinous protein.

In two experiments mold bran was used to convert granular wheat flour. Both resulted in poor filtrations, although one included a 16-hour steeping treatment. The protein content of the thin liquors was found to be more than twice that resulting from malt or fungal amylase conversions. In another experiment (run 53) a mixture of mold bran and fungal amylase liquor was used to convert steeped granular wheat flour. A fair filtration was obtained. As in the two mold bran conversions, the protein content of the thin liquor was found to be high—5.3% protein as compared with 2.0% protein (D.S.B.) in malt liquor.

REFINING OF SIRUP

Several of the filtered, converted liquors (thin liquors) were refined and concentrated to sirups in the laboratory. In general, the refining treatment consisted of heating with various grades of activated vegetable carbon, refined filter aid (diatomaceous earth), and bentonite; the carbon served to reduce the color and the bentonite the protein. Papain, sodium bisulfite, and sulfur dioxide were tested as refining agents but were not found to be advantageous.

A typical sirup run was made as follows: 12 gallons of thin liquor were treated with 85 grams of activated carbon and 85 grams of filter aid. The mixture was heated to 75° C., held there 30 minutes, filtered through filter aid, then concentrated in the glass evaporator to specific gravity 1.22. The mid-sirup was brought to pH 7.0 with calcium hydroxide, and treated with 85 grams of carbon, 85 grams of bentonite, and 85 grams of filter aid. After being held at 55° for 30 minutes, the mixture was filtered and concentrated to a sirup of 70–75% dry substance. Table II summarizes the sirup refining experiments and presents the characteristics of the resulting sirups.

Malt-converted granular wheat flour refined in this manner yielded a clear light-golden sirup (No. 18) of pleasant taste, containing about 0.6% protein and 0.7% ash (D.S.B.). Granular wheat flour converted with filtered fungal amylase liquor yielded a slightly darker sirup (No. 13, 14, 17, 19) with a stronger flavor, containing less than 1% protein and ash; whole corn converted in this manner yielded a similar sirup (No. 16) but with slightly more color. Mold bran fungal amylase liquor on extracted granular wheat flour yielded a dark, cloudy sirup (No. 20) of pleasant flavor, with high protein (4%) and ash (1%).

Bentonite appears to adsorb wheat protein strongly but has little affinity to corn protein. Used in the amount of 1% which appeared to be the optimum quantity required on a dry substance basis, the bentonite, together with carbon and filter aid, reduced the protein content about 65% and thus generally enabled the production of a clear, low protein sirup. The carbon was used in amounts of from 2 to 3%; larger amounts were uneconomical and relatively ineffective. The carbon alone effects a considerable reduction of the protein content, averaging about 47%, but without the bentonite treatment this reduction is not sufficient to yield a clear sirup. Sirup 12a, refined without bentonite, contained 2.40% protein (D.S.B.), whereas sirup 12b, refined from the same liquor but with bentonite, contained 1.57% protein (D.S.B.).

SUMMARY

It is possible to prepare, from granular wheat flour, a bland sirup that is clear and light in color. However, for the process to be practical, the granular wheat flour must be extracted with water to remove soluble protein before conversion. This step improves the rate of filtration of the converted mash and also lowers the protein content of the thin liquor. In general, converted wheat mashes filter slowly because of the glutinous nature of the wheat protein, and it has been determined that extraction of the flour is required in order for the rate to be practical.

The experimental work with malt and amylolytic liquor as conversion agents shows that these materials are comparable in their ability to convert starch to reducing sugars. The finished sirups produced with these converting agents were equally good with respect to protein content, color, and clarity.

The application of bentonite in the refining of sirup prepared from granular wheat flour was found to be beneficial in that the protein content of the sirup was lowered appreciably through its use.

ACKNOWLEDGMENT

The authors wish to express their appreciation to E. H. LeMense for measuring amylolytic activities and to F. R. Earle for many of the sugar determinations which were necessary in this investigation.

LITERATURE CITED

(1) Anonymous, *Food Materials and Equip.*, 6, No. 1, 1 (1946).
(2) Boyer, J. W., and Underkofler, L. A., *Chem. & Met. Eng.*, **52**, No. 12, 110 (1945).
(3) Gordon, W. O., *Food Inds.*, **15**, No. 6, 60 (1943).
(4) Olson, W. J., Evans, R., and Dickson, A. D., *Cereal Chem.*, **21**, 533 (1944).
(5) Schoene, L., Fulmer, E. I., and Underkofler, L. A., IND. ENG. CHEM., **32**, 544 (1940).
(6) Shaffer, P. A., and Hartmann, A. F., *J. Biol. Chem.*, **45**, 365 (1921).
(7) Van Lanen, J. M., and Le Mense, E. H., *J. Bact.*, **51**, 595 (1946).

RECEIVED April 22, 1947.

Heat Transfer to Gases through Packed Tubes

EFFECT OF PARTICLE CHARACTERISTICS

MAX LEVA AND MILTON GRUMMER

Central Experiment Station, U. S. Bureau of Mines, Pittsburgh, Pa.

An earlier study on this subject has shown that heat transfer data through packed tubes could be correlated by the equation:

$$h = 0.813 \frac{k}{D_t} e^{-6\frac{D_p}{D_t}} \left(\frac{D_p G}{\mu}\right)^{0.9}$$

This equation does not show h in relation to packing characteristics such as uniformity, shape, thermal conductivity of the solid phase, and surface roughness. Extended work showed the equation applies to mixed packings provided the arithmetic-average diameter is chosen. It is satisfactory to use the nominal size as D_p for Raschig rings; an equivalent spherical diameter on a volume basis must be inserted in the equation for cylinders. The heat transfer coefficients increased for increasing thermal conductivity of the packing, and a relation was developed showing h as a function of k_p. Surface roughness influenced the magnitude of h slightly. The rougher particles gave somewhat higher heat transfer coefficients. A possible mechanism of heat flow through the core of packed tubes is discussed.

AN EARLIER investigation (*2*) undertaken to ascertain the relation between heat transfer coefficients for packed tubes and physical properties of the gas stream, as well as the physical dimensions of the apparatus, showed that the data obtained could be correlated by the equation:

$$h = 0.813 \frac{k}{D_t} e^{-6\frac{D_p}{D_t}} \left(\frac{D_p G}{\mu}\right)^{0.9} \quad (1)$$

A study of the effect of voids on the rate of heat transfer indicated that, for the range considered, no coordinated relation seemed to exist between voids and heat transfer coefficients.

The purpose of this earlier investigation was to obtain a fundamental equation adapted to simple packings such as smooth, uniform spheres. Consequently Equation 1 does not include factors related to the properties of the particles with the exception of particle diameter. The object of the work reported in this paper was to determine how Equation 1 must be modified to account for variations in the properties of packings—for example, nonuniformity in size, differences in shape, thermal conductivity, and degree of surface roughness. Such a modification of Equation 1 was of interest because it would preclude the extensive approximations necessary for industrial applications, as packing materials in practice are rarely smooth, uniform spheres.

The unit used for this study was described earlier (*2*). The same measurements were made, and equally careful precautions were observed to obtain reliable data. The variables which appeared to be simplest were studied first. The knowledge gained from these studies was then applied as the investigation gradually extended to more complex cases.

The general order of the work was as follows: (*a*) re-examination of Equation 1 using a smooth, spherical, uniform packing; (*b*) investigations with several smooth, spherical, nonuniform packings, and determination of the proper value for D_p; (*c*) extension of *b* to differently shaped particles, such as smooth cylinders and Raschig rings; (*d*) study of the effect of thermal conductivity of the packings, using various shapes; and (*e*) investigation of the effect of surface roughness on heat transfer.

The properties of the packing are given in Table I.

The compositions of packing materials Nos. 2, 3, 4, 15, 16, and 17 are presented in Table II.

RESULTS

The original data of this study are reported in Table III. Figures 1, 2, 3, and 5 show, on log-log paper, heat transfer coefficients plotted against the modified Reynolds numbers for the eighteen different packing materials investigated. The solid line on these graphs correlates the experimental findings, and the dotted line represents Equation 1.

Figure 1 shows the results obtained with the first four materials as reported in Table I. The observed data for packing No. 1 were in excellent agreement with the predicted values. For materials 2, 3, and 4, the heat transfer coefficients were measured and the observed value was substituted in Equation 1. The equation was then solved for the effective particle diameter D_p. The calculated values of D_p agreed closely with the average arithmetical diameters obtained on a volume basis. This value of D_p was chosen to evaluate the modified Reynolds number, and the dotted lines were drawn in Figure 1 on the basis of this diameter.