

Contributions of Distillate Components to Disperse Phase Structures in Model Spirit Solutions

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The dispersed phase of a Scotch distillate behaved differently from that of a model solution containing solely the esters that are normally present in such whiskeys. The presence of surface active distillate components, such as long-chain alcohols, aldehydes, and esters, had the major stabilizing influence on dispersed phase agglomerates. Increasing the hydrophilicity of the end group and the length of the aliphatic chain of individual congeners made positive contributions to surface activity. Possible surface active components in the test distillate were the long-chain aliphatic alcohols tetradecanol and hexadecanol and possibly ethyl esters of 18-carbon acids. The optimum ethanol concentrations for agglomerate stability were 1% in the presence of surface active agents and 3% in the absence of such components. Excess ethanol appeared to reduce the stabilizing effect of surface active compounds. Gas chromatography of new distillate headspaces over 4 h following dilution showed no significant changes in concentration of volatiles above the liquid phase. This suggests that changes in agglomerate diameter would have no effect on the assessment of spirit for the duration of a normal sensory panel. The formation of stable agglomerates may alter the release of aroma compounds in the mouth, influencing concentrations of compounds reaching the retronasal cavity and so influencing perceptions of flavor.

Keywords: Ethyl esters; whiskey; photon correlation spectroscopy; interfacial tension

INTRODUCTION

Relatively little is known about the physical chemistry of distilled beverages, such as whiskeys and brandies, which, until recently, were regarded as homogeneous. Recent studies have shown that, at ethanol concentrations typical of distilled spirits, ethanol exists as aggregates in aqueous solution (D'Angelo et al., 1994; Matsumoto et al., 1995). Ethanol aggregation occurs at mole fractions 0.05–0.08 (15–22% v/v), and the solubility of ethyl esters increased markedly with increasing ethanol concentration above this critical point (Conner et al., 1998). Dilution of the spirit from either maturation strength (50–70% ethanol) or bottling strength (40–43% ethanol) to 22–23% ethanol for sensory assessment (Perry, 1989; Piggott and Jardine, 1979) forms a supersaturated solution of the less hydrophilic esters, and the excess solute forms agglomerates (Conner et al., 1994a). Partitioning of distillate components on the basis of hydrophobicity takes place, notably with long-chain aliphatic esters, alcohols, and aldehydes forming the disperse phase. This reduces the free solution and consequently the headspace concentration of important aroma compounds (Conner et al., 1994b) and so influences the perception of aroma character.

When new distillates are assessed sensorially, headspaces have unattractive aroma characters, described

as *oily*, *soapy* and *grassy*. During maturation of Scotch malt distillates, there is an extraction of less volatile wood components into the spirit, which may be used to predict the development of mature spirit characters (Piggott et al., 1993). Associated with the development of mature spirit characters is a reduction in immature aroma character. However, little loss of distillate flavor components is observed during maturation; in fact, most concentrations increase through evaporation of water and ethanol through the cask wood. Fractionation of spirits showed that immature aroma characters are associated with polarity fractions containing medium- and long-chain ethyl esters (Piggott et al., 1992a,b).

Volatile components of distillates must also contribute to differences in aroma character, and contents vary in relation to such factors as raw material composition, yeast physiology, and still parameters. Short-chain aliphatic esters are generally considered to have *fruity* aroma characters, whereas esters with longer aliphatic chains, such as ethyl decanoate and dodecanoate, have *soapy* notes (Ribereau-Gayon, 1978). In this study, the first objective was to examine the diameter and stability of agglomerates in disperse phases formed on dilution of distillate for sensory analysis using photon correlation spectroscopy. A second objective was to determine how changes in agglomerate composition influenced interfacial tension, a key factor in disperse phase stability.

MATERIALS AND METHODS

Materials. A single batch of Longmorn new malt distillate was provided by the Chivas and Glenlivet group. Distillate was diluted to 63.4% v/v ethanol and then stored in a borosilicate glass bottle at ambient temperature until use. Models of immature whiskey used water purified by a MilliU10

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system (Millipore U.K. Ltd., Watford, U.K.) and ethanol of HPLC grade (Rathburn Ltd., Walkerburn, U.K.). Other solutes were >99% pure; ethyl esters, acetates, and alcohols were obtained from Sigma Chemical (Poole, U.K.); 2-ketones were from Fluka (Gillingham, U.K.) and aldehydes from Aldrich (Gillingham, U.K.).

Methods. Photon correlation spectroscopy (PCS) was employed for agglomerate size analysis utilizing a Malvern type 7027 spectrometer with a 40 mW He/Ne laser. All measurements were made at 25 °C with a 90° scattering angle. The aperture of the photomultiplier was set at 0.5, and the wavelength of the light source at 632.8 nm. Samples were filtered through 0.2 μ m cellulose nitrate filters prior to mixing and transferred to thermoregulated cuvettes, surrounded by xylene. Particle diameters and the polydispersity index were derived from the mean z-average diffusion coefficients calculated according to the method of cumulants analysis (Koppel, 1972).

Surface and interfacial tensions were measured using a torsion balance (White Electrical Instruments Ltd., Malvern Link, U.K.), which employs a platinum ring of 40 mm circumference. Accuracy was assessed by determining the surface tension of the water used or the interfacial tension of a water/octane mixture. Mean surface tension was 73 mN m⁻¹ (SD = 0.5 mN m⁻¹) at 20 °C. Mean interfacial tension was 51 mN m⁻¹ (SD = 1.0 mN m⁻¹) at 20 °C. Before interfacial tension was determined, samples (10 mL of aqueous ethanol with 10 mL of ester or alkane) were equilibrated at 25 °C for 1 h.

Headspace gas chromatography (Conner et al., 1994a,b) was used to determine activity coefficients, except for ethyl hexadecanoate. This was determined using a 7 μ m poly(methylsiloxane)-coated microextraction fiber (Supelco U.K., Poole, U.K.) exposed in the headspace for 5 min and then desorbed at 270 °C in the SPI injector of a Finnegan MAT ITS40 gas chromatograph linked mass spectrometer. A CP-Sil 52 CB column (0.32 mm $\times$ 30m; df = 0.5 μ m) (Chrompak Ltd., London, U.K.) was employed with helium gas at 1 mL min⁻¹ as carrier and a temperature gradient of 15 °C min⁻¹ to 240 °C after a 5 min desorption at 270 °C. Peak areas were calculated as the sum of responses to *m/z* 284 and 285.

Statistical analysis of data used Minitab v. 9.2.

RESULTS AND DISCUSSION

Size of Particles in Diluted Distillate. In preliminary experiments employing PCS it was apparent that with new distillate there was an increase in agglomerate size with time after dilution of spirit from 63.4% ethanol to 23% ethanol (Figure 1). Agglomerate size increased from 114 to 199 nm in the initial 4 h and by a further 28 nm over the following 20 h. We have previously shown (Conner et al., 1994a) that medium- and long-chain ethyl esters are most abundant in the disperse phase. The model for new distillate contained typical concentrations of medium- and long-chain ethyl esters and 3-methylbutanol found in whiskey (Table 1). On dilution, the new distillate model showed an agglomerate size similar to that of the newly made spirit, but after 24 h, agglomerates were significantly larger, at 398 nm, than with the new make spirit, 227 nm (*p* < 0.001).

Activities of the major ethyl esters were determined 0.5, 1, 2, 4, and 24 h after dilution. Values for 1 and 24 h are given in Tables 2 and 3. No significant changes with time were observed. Activities for ethyl octanoate, decanoate, and dodecanoate (Table 2) were calculated from on-column injections of headspace using previously published methods (Conner et al., 1994a). However, the activity of ethyl hexadecanoate was determined using a solid-phase microextraction fiber. Though these cannot be easily used to quantitate headspace components, activities (Table 3) were calculated from the peak areas

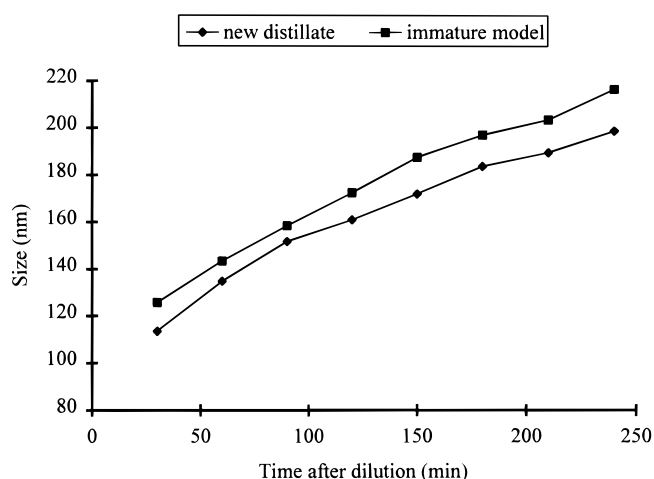


Figure 1. Changes in the particle size of the dispersed phase on dilution to 23% v/v ethanol of a new Scotch malt distillate and a model immature spirit containing the principal medium- and long-chain esters and isoamyl alcohol. Polydispersity index did not show any significant change with time. Mean values were 0.10 for the distillate and 0.13 for the model, with a standard deviation of 0.02.

Table 1. Composition of the Model of Immature Spirit Used in Particle Size Experiments^a

congener	concn (mg/L)	congener	concn (mg/L)
3-methylbutanol	200	ethyl dodecanoate	15
ethyl octanoate	9	ethyl tetradecanoate	5
ethyl decanoate	20	ethyl hexadecanoate	15

^a Concentrations are for the spirit diluted to 23% v/v ethanol.

Table 2. Activities of the Three Major Ethyl Esters in New Make and Immature Spirits Measured Using On-Column Headspace Injection (Conner et al., 1994a,b)

ethyl ester	new make		immature model	
	1 h	24 h	1 h	24 h
octanoate	0.02 (0.001)	0.02 (0.003)	0.03 (0.004)	0.03 (0.001)
decanoate	0.17 (0.04)	0.15 (0.02)	0.22 (0.02)	0.25 (0.03)
dodecanoate	0.27 (0.02)	0.24 (0.01)	0.34 (0.02)	0.36 (0.02)

Table 3. Determination of Ethyl Hexadecanoate Activity in New Make and Model Immature Spirits Using an SPME Fiber

sample	peak area (<i>m/z</i> 284 + 285/1000)	activity (SD)
new make spirit		
1 h	569.7, 496.1, 519.6, 523.4	0.429 (0.025)
24 h	499.2, 490.1, 578.6, 590.5	0.439 (0.043)
immature model		
1 h	667.1, 618.3, 652.6, 621.1	0.521 (0.020)
24 h	648.9, 649.7, 653.1, 671.1	0.534 (0.008)
pure solute	1306.5, 1086.8, 1172.3, 1346.9	

obtained from the sample headspace and the headspace above the pure solute (Grant and Higuchi, 1990). For reproducible measurements a short (5 min) equilibration time was required to prevent fiber saturation. The sum of ester activities was close to unity, indicating the solvent was saturated with the mixed components (Conner et al., 1994a). The lack of change in headspace concentrations indicated that changes in agglomerate diameter can be explained as coagulation of the dispersed phase. Dilution reduces ester solubility, resulting in supersaturation of the solution, and excess is excluded to form lyophobic colloidal aggregates. Although the initial emulsion was unstable, the disperse phase reached an equilibrium agglomerate size within

Table 4. Measured Size and Polydispersity Index of the Dispersed Phase in a Supersaturated Solution of Different Esters in 23% v/v Ethanol and the Interfacial Tension (IT) of the Ester with 23% v/v Ethanol

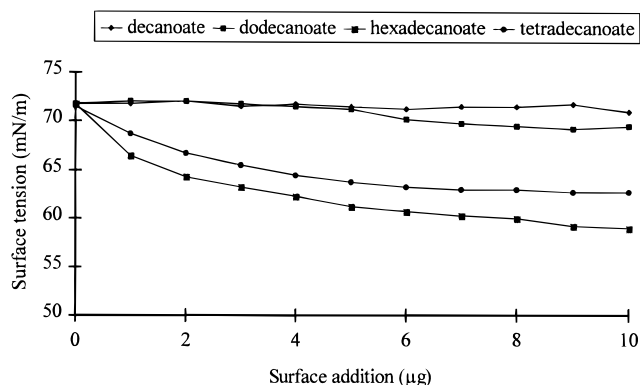
ethyl ester	size ^a (and polydispersity index) at			mean rate ^b	IT ^c
	15 min	2 h	24 h		
1, decanoate	364 (0.25)	703 (0.24)	870 (0.27)	3.32	10.5
2, dodecanoate	221 (0.09)	490 (0.15)	657 (0.16)	2.44	12
3, tetradecanoate	145 (0.07)	242 (0.05)	350 (0.09)	0.88	13.5
4, hexadecanoate	115 (0.10)	155 (0.08)	214 (0.08)	0.35	solid
1 + 2	269 (0.15)	771 (0.37)	840 (0.29)	4.66	11.0
1 + 4	144 (0.13)	204 (0.12)	435 (0.12)	0.48	9.5
2 + 4	131 (0.11)	180 (0.05)	424 (0.08)	0.45	10.0
1 + 2 + 4	122 (0.06)	180 (0.05)	405 (0.08)	0.53	9.0

^a Mean diameter in nanometers. ^b Linear rate over first 2 h. ^c With 23% v/v ethanol in mN m⁻¹.

24 h. The thermodynamic driving force of coagulation is minimization of the interfacial area of disperse phase in contact with water. In the absence of ethanol, rapid phase separation would be predicted. Ethanol is surface active and reduces interfacial tension between water and ester and so limits disperse phase coagulation. Distillate components must also contribute to this process as the equilibrium agglomerate size in new make spirit was significantly lower than in the model new distillate. It was important to identify such components.

Influence of Distillate Components on Equilibrium Agglomerate Diameter. A set of models was prepared at 65% ethanol, to give supersaturated solutions when diluted to 23% ethanol, of the following homologous series of ethyl esters: decanoate (100 $\mu\text{g mL}^{-1}$ at 23% ethanol); dodecanoate (50 $\mu\text{g mL}^{-1}$ at 23% ethanol); tetradecanoate (50 $\mu\text{g mL}^{-1}$ at 23% ethanol); and hexadecanoate (50 $\mu\text{g mL}^{-1}$ at 23% ethanol). Each was diluted to 23% ethanol, and agglomerate sizes were determined every 15 min for 2 h and after 24 h (Table 2). The mean linear rate of agglomerate diameter increased, and diameters at 2 and 24 h were inversely related to aliphatic chain length (adjusted R^2 values of 95.3, 95.2, and 96.4). No significant change with time was observed in polydispersity index for each system. Values for the interfacial tension of each ester with 23% ethanol, however, showed a direct relationship with length of ester aliphatic chain. Similar results have been obtained for alkanes (Zograf and Yalkowsky, 1974) and the differences attributed to the changes in surface group densities. Models were also prepared with mixtures of esters. The mixture of ethyl decanoate and dodecanoate showed a more rapid increase in agglomerate diameter than either ester alone. This can be attributed to the enhanced concentration of dispersed phase increasing the rate of particle collisions. However, in ester mixtures, ethyl hexadecanoate showed behavior similar to that of a surfactant, decreasing interfacial tension and yielding more stable agglomerates of smaller diameter. A decrease in the surface tension of water was observed on addition of tetradecanoate or hexadecanoate, but not with decanoate or dodecanoate (Figure 2).

To study the effect of other components, PCS and interfacial and surface tension analyses were repeated for saturated solutions at 23% ethanol of an aliphatic acid ester, ethyl decanoate; a medium-chain alcohol ester, decyl acetate; a ketone, 2-dodecanone; a medium-chain alcohol, dodecanol; and a similar aldehyde, dodecanal (Table 5). The two esters yielded the largest

**Figure 2.** Changes in the surface tension of water on the surface addition of different esters dissolved in pentane.**Table 5. Measured Size and Polydispersity Index of the Dispersed Phase in a Supersaturated Solution of Different 12-Carbon Compounds in 23% v/v Ethanol and the Interfacial Tension (IT) of the Ester with 23% v/v Ethanol**

	size ^a (and polydispersity index) at			mean rate ^b	IT ^c
	15 min	2 h	24 h		
1, ethyl ester	364 (0.25)	703 (0.24)	870 (0.27)	3.32	10.5
2, 1-alcohol	209 (0.13)	251 (0.13)	382 (0.17)	0.39	solid
3, aldehyde	233 (0.12)	237 (0.17)	298 (0.14)	0.02	8.0
4, 2-ketone	267 (0.31)	397 (0.33)	404 (0.28)	1.53	8.0
5, acetate ester	357 (0.20)	694 (0.31)	846 (0.20)	3.27	9.5
1 + 2	276 (0.16)	404 (0.21)	457 (0.19)	1.26	6.5
1 + 3	119 (0.20)	135 (0.15)	157 (0.17)	0.16	5.0
1 + 4	175 (0.07)	280 (0.13)	394 (0.14)	0.82	9.0
1 + 5	314 (0.20)	760 (0.26)	880 (0.28)	4.16	10.0

^a Mean diameter in nanometers. ^b Linear rate over first 2 h. ^c With 23% ethanol in mN m⁻¹.

agglomerate diameter and the greatest rates of increase. The aliphatic alcohol and aldehyde showed the smallest agglomerate diameters. For dodecanol the rate of agglomerate diameter increase was similar to that of ethyl hexadecanoate in the previous experiment. The aldehyde showed no significant change in agglomerate diameter over 2 h and only a small increase over 24 h. As in Table 4, the polydispersity indices did not vary significantly with time. As both alcohol and aldehyde are solids at 25 °C, their influence on interfacial tension could not be determined. Both, however, exhibited the surface active effects in the presence of ethyl decanoate observed previously. These compounds enhanced agglomerate stability, reduced equilibrium diameter, and had marked effects on interfacial tension and the surface tension of water (Figure 3).

Interactions of Distillate Components with 23% Ethanol. Headspace gas chromatography was used to quantify the activity coefficients for homologous series of aliphatic acid ethyl esters, aliphatic alcohol acetate esters, and aliphatic alcohols, aldehydes, and ketones. A series of almost parallel lines was obtained when the natural log of activity coefficients was plotted against the number of methylene and methyl groups (Figure 4). The results for the two esters were not significantly different when the total number of carbons in the molecule was taken into account. The activity coefficient can be defined as the excess or nonideal mixing function of a solute with the solvent. This can be converted to the free energy contributions ($RT \log \gamma$) of the polar headgroup (intercept) and each methylene group (gradient). Free energy contributions for headgroups are summarized in Table 6 and include a

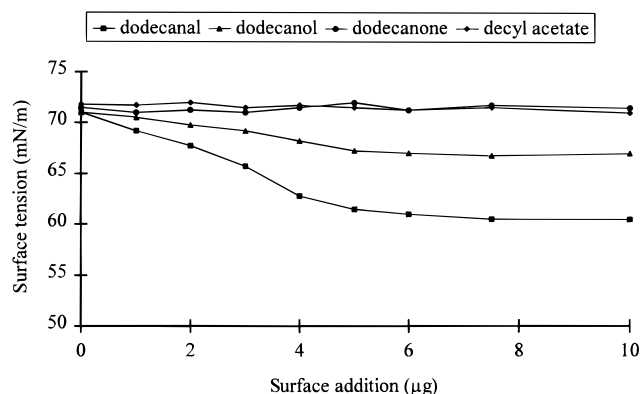


Figure 3. Changes in the surface tension of water on the surface addition of different 12-carbon compounds dissolved in pentane.

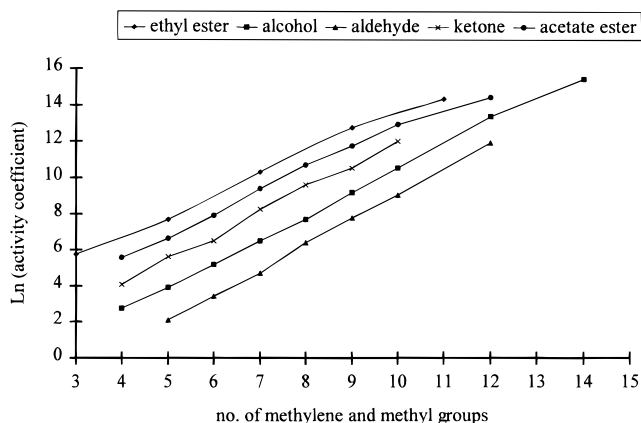


Figure 4. Activity coefficients of homologous series of aliphatic compounds containing different functional groups in ethanol.

Table 6. Free Energy Contributions of End and Methylene Groups for Different Compound Types Dissolved in 23% Ethanol

end group	end group contribution (J mol ⁻¹)	methylene group contribution (J mol ⁻¹)
H ··· CH=O	-8700	3470
H ··· CH ₂ OH	-3400	3200
H ··· COCH ₃	-2100	3200
H ··· OCOCH ₃	600	3200
H ··· COOC ₂ H ₅	5100	2800

correction for the terminal methyl group, calculated at 4770 J mol⁻¹ from alkane solubility (Davis, 1973a). Corrected values for free energy contribution were mostly negative, except for the aliphatic esters, suggesting that all other headgroups were hydrophilic. The most hydrophilic was the aldehyde group. Values for methylene group contributions were not significantly influenced by the end group and were similar to those from partition studies using alkane water systems (Davis, 1973b). With both esters there was a reduction in methylene group contribution when the aliphatic chain reached 14 carbons. Similar results have been observed previously for the aqueous solubility of alkanes (Davis et al., 1974) and can be attributed to folding of the alkyl chain to minimize surface area.

The presence of the surface active distillate components had major stabilizing influences on dispersed phase agglomerates. The hydrophilicity of the end group made a positive contribution to surface activity. This is well documented: increasing interaction with the aqueous phase and reducing solubility in the oil

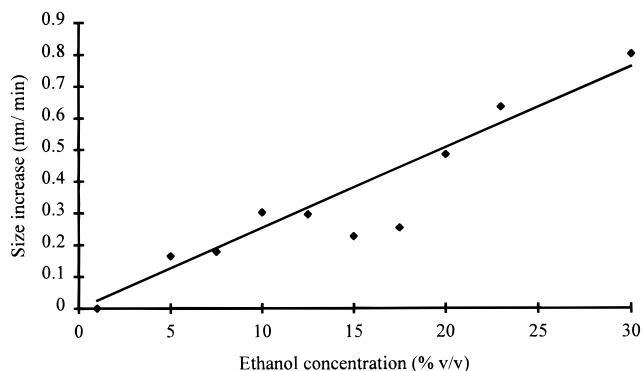


Figure 5. Effect of ethanol concentration on the rate of size increase for immature model. Concentrations of model components were not altered in line with changes in ethanol concentration. Polydispersity index decreased from 0.27 at 1% ethanol to 0.03 at 30% ethanol.

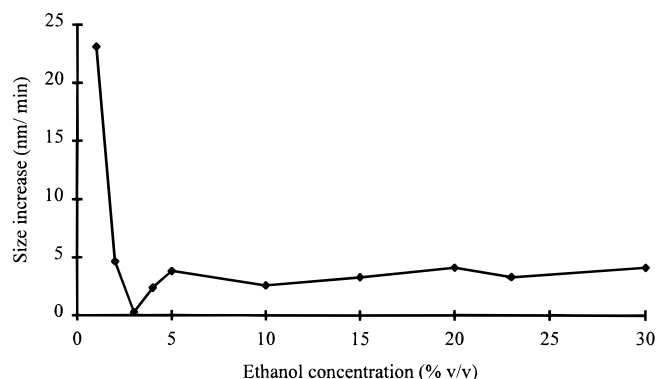


Figure 6. Effect of ethanol concentration on the rate of size increase for a supersaturated solution of ethyl decanoate. The concentration of ethyl decanoate was not altered in line with changes in ethanol concentration. Polydispersity index decreased from 0.45 at 1% ethanol to 0.15 at 10%, increasing thereafter to 0.38 at 30% ethanol.

phase favor the correct orientation of the molecule at the interface (Davis, 1973b). Methylene groups also have long been known to make a positive contribution to surface activity (Traube's rule), but for the series of ethyl esters the increasing aliphatic chain length appears to induce surface activity in the ester.

This suggests that the major surface active components in the Longmorn distillate are the long-chain aliphatic alcohols, tetradecanol and hexadecanol, and possibly ethyl esters of 16- and 18-carbon acids (Piggott et al., 1993).

Effect of Ethanol Concentration. The model of the new distillate and a supersaturated solution of ethyl decanoate were prepared at different alcoholic strengths (from 1 to 30% v/v ethanol) to assess the impact of changing ethanol concentration on disperse phase stability. The model distillate (Figure 5) contained surface active esters, whereas ethyl decanoate (Figure 6) showed no surface activity. With the new distillate model in 1% ethanol, agglomerate size was stable for 2 h. The agglomerate diameter increased roughly in parallel with increasing ethanol concentration, over the ranges 1–10 and 20–30% ethanol with a decrease in polydispersity index. For the saturated solution of ethyl decanoate (Figure 6) at ethanol concentrations <3%, unstable agglomerates with high polydispersity were formed with rapid phase separation. At 3%, the rate of agglomerate size change was at a minimum, but increased up to 5% followed by a plateau to 30%. This suggests there must

be an optimum solvation for the agglomerates by the ethanol: 1% in the presence of surface active agents; 3% in the absence of such components. However, ethanol appears to have only a limited effect on the increase in agglomerate size. Excess ethanol appears to reduce the stabilizing effect of surface active compounds.

Conclusions. The dispersed phase of a Scotch distillate behaved differently from that of a model containing solely the esters that are normally present in such whiskeys. Agglomerates were partially stabilized by the presence of surface active components in distillates. Effects of individual compounds appeared to relate to the hydrophilicity of the head and the length of the alkyl chain. However, gas chromatographic analysis of new distillate headspaces over 4 h following dilution showed no significant changes in concentration of volatiles above the liquid phase. This suggests that changes in agglomerate diameter would have no effect upon sensory assessments of spirit over a 2–4-h period. However, changes in agglomerate size and stability have been shown to alter the release of aroma compounds in the mouth on ingestion by consumers (Harrison and Hills, 1997). This would influence the concentrations of compounds reaching the retronasal cavity and, consequently, the perceptions of flavor.

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