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Thermodynamic Study of Some Amino Acids, 2-Aminopropanoic Acid, 2-Amino-3-methylbutanoic Acid, 2-Amino-4-methylpentanoic Acid, and 2-Amino-3-phenylpropanoic Acid in Aqueous Saccharide Solutions at Different Temperatures: Volumetric and Ultrasonic Study

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Supporting Information

ABSTRACT: Knowledge of thermodynamic properties of amino acids in aqueous medium can provide valuable information about the stabilization mechanism of proteins. In this study, we report the density and speed of sound of some amino acids (2-aminopropanoic acid, 2-amino-3-methylbutanoic acid, 2-amino-4-methylpentanoic acid, and 2-amino-3-phenylpropanoic acid) in aqueous fructose, D-(+)-cellobiose, and D-melezitose solutions (0.1 mol·kg⁻¹) at (293.15, 298.15, 303.15, and 308.15) K. From these data, the apparent molar volume and apparent molar adiabatic compressibility have been evaluated. The partial molar volume, partial molar adiabatic compressibility, and their corresponding transfer functions were calculated. These results were used to interpret the type of interactions occurring in the ternary mixtures using the cosphere overlap model. The concentration and temperature dependence of solute—solute and solute—solvent interactions in the present study was also discussed.

■ INTRODUCTION

Because of the complex structure of proteins, direct studies of solute-solvent effects on these biological macromolecules are quite difficult.1 Therefore, to study the interactions of these macromolecules, some simple model compounds, amino acids, which are the basic structural units of proteins,^{1,2} should be taken. There is a difference in the side chains of these model compounds due to the size, shape, charge, hydrogen-bonding capacity, hydrophobicity, and chemical reactivity, due to which these side chains contribute to the structure and function of proteins, individually and collectively.³ Carbohydrates located at cell surfaces are very important as receptors with regards to the bioactive structure of hormones, enzymes, viruses, antibodies, and so forth.⁴ Therefore, in living systems, carbohydrate and protein interactions play an important role in various biochemical processes such as immunology, biosynthesis, pharmacology, and medicine.5

From the literature survey, we found that in recent years there are extensive studies of volumetric properties of amino acids in aqueous solution⁶⁻¹² and in mixed aqueous solutions,¹³⁻²⁵ and the protonation constant study of amino acids in mixed aqueous solution,²⁶⁻²⁹ but very few studies have been done in aqueous saccharide solutions.³⁰⁻³⁶

Amino acids (2-aminopropanoic acid, 2-amino-3-methylbutanoic acid, 2-amino-4-methylpentanoic acid, and 2-amino-3phenylpropanoic acid) are hydrophobic in nature. They have many biological characteristics. For example, in sickle cell disease, 2-amino-3-methylbutanoic acid is used as a substituent for the hydrophilic glutamic acid in hemoglobin because of its hydrophobicity; the hemoglobin does not fold correctly. 2-Amino-4methylpentanoic acid is a major component of the subunits in ferritin and other buffer proteins. 2-Amino-3-phenylpropanoic acid is used in the manufacturing of food and drink products. It is also sold as a nutritional supplement for its reputed analgesic and antidepressant effects. On the other hand carbohydrates due to their conformational flexibility are important for many biological processes, such as cell-cell recognition and molecular and cellular communication.³⁷ (2R,3S,4S,5R)-2,5-Bis(hydroxymethyl)oxolane-2,3,4-triol (fructose) is a fruit sugar and used in the formation of corn syrup. (2R,3S,4S,5R,6S)-2-(Hydroxymethyl)-6-[(2R,3S,4R,5R, 6*R*)-4,5,6-trihydroxy-2-(hydroxymethyl)oxan-3-yl]oxyoxane-3,4,5-triol (cellobiose) due to the presence of eight free alcoholic groups and three ether linkages has the tendency to form intermolecular as well as intramolecular hydrogen bonding. (2R,3R,4S,5S,6R)-2-[(2S,3S,4R,5R)-4-Hydroxy-2,5-bis(hydroxymethyl)-2-[(2R,3R,4S,5S,6R)-3,4,5-trihydroxy-6-(hydroxymethyl)oxan-2-yl]oxyoxolan-3-yl]oxy-6-(hydroxymethyl)oxane-3,4,5triol (melezitose) is a nonreducing trisaccharide. This is beneficial to insects as it reduces the stress of osmosis by reducing their own water potential. It is also a part of honeydew due to which it acts as an attractant for aunts and also as a food for bees. Therefore, the systematic study of amino acids and carbohydrate can provide valuable information about various biochemical processes. In this paper, we report the studies on the interactions of saccharides (mono, di-, and tri-) with amino acids (2-aminopropanoic acid, 2-amino-3-methylbutanoic acid, 2-amino-4methylpentanoic acid, and 2-amino-3-phenylpropanoic acid) by densimetry and ultrasonically at (293.15, 298.15, 303.15,

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and 308.15) K. In this study, apparent molar volume and apparent molar adiabatic compressibility at infinite dilution, their corresponding transfer functions, and partial molar expansions at infinite dilution are reported to discuss the types of interactions, that is, solute—solute or solute—solvent and also their nature (hydrophilic—hydrophilic, hydrophilic—hydrophobic, and hydrophobic—hydrophobic), occurring in the ternary system (amino acids + saccharides + water) and its effect with temperature.

MATERIALS AND METHODS

All amino acids (2-aminopropanoic acid, 2-amino-3-methylbutanoic acid, 2-amino-4-methylpentanoic acid, and 2-amino-3-phenylpropanoic acid, mass fraction > 0.99) and saccharides: D-(+)cellobiose, D-melezitose monohydrate (Hi Media, Mumbai, mass fraction > 0.99), and D-fructose (Loba Chemie Pvt. Ltd., Mumbai, mass fraction > 0.99) were used as such without further purifications. Before use these were dried under vacuum to constant weight and then stored over P2O5 in desiccators. Doubly distilled deionized water which has been freshly degassed was used for the preparation of the aqueous solutions. Stock solutions of saccharides $(0.1 \text{ mol} \cdot \text{kg}^{-1})$ were prepared by mass on the molality concentration scale. Solutions of 2-aminopropanoic acid and 2-amino-3-methylbutanoic acid in the concentration range (0.05 to 0.30) mol·kg⁻¹ and of 2-amino-4methylpentanoic acid and 2-amino-3-phenylpropanoic acid in the concentration range (0.03 to 0.14) mol \cdot kg⁻¹ were made by mass on the molality concentration scale with an accuracy of $\pm 1 \cdot 10^{-5}$. The weighings were done on an A&D Company, Limited electronic balance (Japan, model GR-202) with a precision of \pm 0.01 mg. The uncertainties in the solution molalities were in the range $\pm 2 \cdot 10^{-5}$ $mol \cdot kg^{-1}$. All solutions were prepared afresh before used.

Solution densities and speeds of sound were measured simultaneously and automatically, using an Anton Paar DSA 5000 instrument that was calibrated at (293.15, 298.15, 303.15, and 308.15) K with doubly distilled water and dry air. Both speed of sound and density are extremely sensitive to temperature, so it is controlled to $\pm 1 \cdot 10^{-2}$ K by built-in solid state thermostat. The sensitivity of the instrument corresponded to a precision in density and speed of sound measurements of $\pm 1 \cdot 10^{-6}$ g·cm⁻³ and $\pm 1 \cdot 10^{-2}$ m·s⁻¹, respectively. The reproducibility of density and speed of sound was found to be better than $\pm 5 \cdot 10^{-6}$ g·cm⁻³ and $\pm 5 \cdot 10^{-2}$ m·s⁻¹, respectively.

RESULTS AND DISCUSSION

The densities and speeds of sound of amino acids (2-aminopropanoic acid, 2-amino-3-methylbutanoic acid, 2-amino-4methylpentanoic acid, and 2-amino-3-phenylpropanoic acid) in aqueous saccharide (D-fructose, D-(+)-cellobiose, and D-melezitose monohydrate) solutions were determined using a DSA 5000 instrument at (293.15, 298.15, 303.15, and 308.15) K. The values of density and speed of sound are given in Table 1. From these values, the apparent molar volume and apparent molar adiabatic compressibility of amino acids have been calculated using the following equations:

$$V_{\phi} = (M / \rho) - \{(\rho - \rho_0) / m \rho \rho_0\}$$
(1)

$$K_{\phi,s} = (M\beta_s / \rho) - \{(\beta_{s,0}\rho - \beta_s\rho_0) / m\rho\rho_0\}$$
(2)

where *M* is the solute (amino acids) molar mass, ρ_0 , ρ , $\beta_{s,0}$, and β_s are the densities and coefficient of adiabatic compressibilities of

pure solvent and solution, respectively, and m is the solution molality. The coefficients of adiabatic compressibilities were calculated using the equation:

$$\beta_{\rm s} = 1/(u^2 \rho) \tag{3}$$

where *u* is the speed of sound value. The calculated values of V_{ϕ} and $K_{\phi,s}$ are given in Table S1 (given as Supporting Information, SI). V_{ϕ} values are increasing with molality of amino acids, suggesting that solute—solvent interactions are increasing with the increase in the concentration of amino acids.

The V_{ϕ} and $K_{\phi,s}$ data has been used to see the effect of temperature and saccharide concentration on solute—solvent interactions occurring in the ternary mixture of present study. For this purpose the linear regression analysis of V_{ϕ} and $K_{\phi,s}$ was carried out using the following equation:

$$Y_{\phi} = Y_{\phi}^{0} + S_{Q}m \tag{4}$$

where Y_{ϕ}^{0} (denotes V_{ϕ}^{0} or $K_{\phi,s}^{0}$) is the limiting value of partial molar property (equal to the infinite dilution partial molar property) and S_Q (S_Q denotes S_V or S_K) is the experimental or limiting slope. The observed values of limiting partial molar properties and their experimental slopes are given in Table S2 (given as SI), and V_{ϕ}^{0} values for amino acids in water are compared with the previously reported values.^{6,7,15,38-47} When the change on these properties of amino acids with the temperature is examined, it is generally observed that these partial molar properties increase with the increase in temperature and also with the change in the saccharides (except for 2-amino-4-methylpentanoic acid). The V_{ϕ}^{0} reflects the presence of solute-solvent interactions. It can be seen from the Table S2 that V_{ϕ}^{0} values are positive for all of the amino acids and increase with the increase in temperature, indicating that solute-solvent interactions are increasing with the increase in temperature. This may be caused by the reduced electrostriction of water due to (NH_3^+) and COO⁻) groups of the amino acids. The same observations have been shown by the other authors^{19,48,49} for amino acids in aqueous electrolyte solutions. The results of $V_{\phi}^{0}(\mathbf{R})$ reveals that the contribution of the side-chain R group is significant and positive for all of the amino acids and increase as the size of alkyl chain increases from 2-aminopropanoic acid to 2-amino-3-phenylpropanoic acid. The order of increase in V_{ϕ}^{0} value is 2-amino-3-phenylpropanoic acid > 2-amino-4-methylpentanoic acid > 2amino-3-methylbutanoic acid > 2-aminopropanoic acid It may be concluded that V_{0}^{0} values increase with increase in molar mass as reported earlier.^{19,50}

Also Table S2 reveals that S_V values for amino acids are decreasing with the change in the saccharide (except for 2-amino-3-phenylpropanoic acid) which suggests that the solute—solvent interactions are increasing from mono- to trisaccharide, whereas the $K_{\phi,s}^{0}$ values are negative for all of the amino acids in aqueous saccharide solutions. This indicates that the water molecules around amino acids are less compressible than the water molecules in the bulk solution.

The S_V values give information regarding solute—solute interactions. The trends of variation of S_V with temperature in all solvents are irregular. The S_V values are positive for all amino acids (except for 2-amino-3-phenylpropanoic acid which is positive in aqueous D-(+)-cellobiose and D-melezitose only) in aqueous fructose solutions at all temperatures, showing the presence of solute—solute interactions which are weaker than solute—solvent interactions. The negative values S_V in all other

Table 1. Density $(\rho/\text{kg}\cdot\text{m}^{-3})$ and Speed of Sound $(u/\text{m}\cdot\text{s}^{-1})$ of Amino Acids in Water and Saccharide–Water Solutions at Different Temperatures

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		<i>T</i> /K = 293.15		<i>T</i> /K = 298.15		<i>T</i> /K = 303.15		T/K = 308.15	
md+kg ⁻¹ kg/m ⁻¹ m/s ⁻¹ kg/m ⁻¹ m/s ⁻¹ kg/m ⁻¹ m/s ⁻¹ kg/m ⁻¹ m/s ⁻¹ 2.4minopropanok Addi n0 10199 mol-kg ⁻¹ Fructore 1.000549 1515.85 1.000570 1525.95 0.06699 1.000578 1.000541 1579.51 1.000540 1525.95 0.06681 1.000543 1533.20 1.000560 1525.55 1.000320 1534.22 0.1773 1.000400 1506.47 1.010354 1533.20 1.000660 1525.25 1.000320 1534.22 0.1776 1.010360 1506.17 1.010354 1532.21 1.010124 1534.32 1.000530 1534.32 0.00000 1.01187 1516.65 1.00072 1538.71 1.008578 1.01187 1538.71 1.00172 1538.71 1.008797 1533.59 0.00000 1.01187 1.012367 1.012377 1.012378 1.594.41 1.01197 1533.41 0.11187 1.012371 1.012353 1.011400 1538.45 1.000879 1538.45	т	$\rho \cdot 10^{-3}$	и	$\rho \cdot 10^{-3}$	и	$\rho \cdot 10^{-3}$	и	$\rho \cdot 10^{-3}$	и
Description Langeproparole Acid In 0.10199 mol·kg ⁻¹ Practose 0.0000 L00528 1490.00 L00654 1515.85 L000900 1523.58 0.06569 L006578 1496.77 L006573 1515.85 L000970 1535.84 0.05561 L007575 L006573 1513.32 L00553 1515.41 L00752 1535.42 0.1776 L010180 150.61 L000872 1530.85 L007149 1540.42 0.228104 L011806 1522.21 L0108472 1530.85 L007149 1540.42 0.20000 L011187 1493.51 L009944 1507.16 L00473 1518.87 L006794 1532.64 0.00000 L011187 1493.71 L011555 1511.05 L010702 1522.75 L008794 1532.64 0.10918 L0143.79 1501.47 L011297 1532.64 L000797 1532.64 0.10918 L01737 1501.47 L01218 L0132.64 L0192.75 L0083.45 L52.41 0.11927	$mol \cdot kg^{-1}$	kg∙m ⁻³	$m \cdot s^{-1}$	kg·m ⁻³	$m \cdot s^{-1}$	kg·m ⁻³	$m \cdot s^{-1}$	kg·m ⁻³	$m \cdot s^{-1}$
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0.05669 1.060573 1.494.01 1.005634 1.507.59 1.004141 1.519.54 1.002473 1.513.20 0.07736 1.009440 150.047 1.000535 151.53.21 1.006680 152.50 1.004570 153.82 0.17766 1.010805 150.161 1.003553 151.681 1.006897 153.085 1.007140 150.02 0.22104 1.011304 1.500.61 1.010384 153.821 1.008572 153.085 1.007140 153.02 0.00000 1.0113101 1.50547 1.010384 1.518.85 1.010473 1.518.75 1.006744 1.532.26 0.005758 1.011317 1.50477 1.012786 1.513.52 1.010472 1.52.275 1.008386 1.535.99 0.16933 1.014350 1.514.45 1.011072 1.52.275 1.008386 1.515.59 0.16934 1.01430 1.57.75 1.012786 1.535.73 1.013386 1.535.73 1.013386 1.535.73 1.013386 1.535.73 1.012336 1.535.75	0.00000	1.005258	1490.00	1.004028	1503.92	1.002569	1515.85	1.000900	1525.95
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0.28104 1.013104 151006 1.011806 1522.21 1.010284 153.4.52 1.008551 154.8.29 0.00000 1.011187 1.493.51 1.009944 1507.16 1.008473 1518.87 1.006794 1528.78 0.00000 1.011281 1.4947.71 1.011283 1.01072 1522.75 1.006794 1538.00 0.15993 1.014589 1501.47 1.012987 1514.55 1.011494 1532.46 1.00797 1.553.94 0.15993 1.016717 1507.92 1.014542 1520.60 1.013910 1532.26 1.012203 1541.95 0.239398 1.016717 1547.92 1.01452 1520.60 1.013910 1532.26 1.012203 1543.48 0.00000 1.017278 1.496.44 1.016107 1510.14 1.014530 1521.80 1.012836 1531.43 0.04444 1.020210 1503.78 1.018753 1.01773 1534.37 1.01773 1543.03 0.118219 1.022210 1503.78 1.002179 </td <td>0.22825</td> <td>1.011668</td> <td>1506.17</td> <td>1.010384</td> <td>1518.85</td> <td>1.008872</td> <td>1530.85</td> <td>1.007149</td> <td>1540.42</td>	0.22825	1.011668	1506.17	1.010384	1518.85	1.008872	1530.85	1.007149	1540.42
De.Aminopropancic Acidin 0.09815 mol·kg ⁻¹ n·(+)-Cellobiase 0.00000 1.011187 1493.51 1.009944 1507.16 1.008473 151.87 1.006794 1532.60 0.01918 1.012813 1.497.71 1.011255 1511.55 1.011474 1532.64 1.000979 1535.99 0.1593 1.015887 1504.97 1.014303 1517.75 1.012798 1532.44 1.011203 1543.95 0.19792 1.016171 1507.92 1.016825 1524.01 1.015304 1535.73 1.012836 1541.95 0.24961 1.018133 1514.66 1.018025 1527.11 1.016496 1538.78 1.01778 1547.76 0.00000 1.017278 1496.64 1.017271 1513.21 1.015778 152.40 1.015886 1558.22 0.04444 1.02101 1505.78 1.019854 1519.74 1.018448 1531.44 1.016755 1564.24 0.14444 1.02131 1506.47 1.019954 1519.74 1.0184.48 1531.47 1.01773	0.28104	1.013104	1510.06	1.011806	1522.21	1.010284	1534.32	1.008561	1543.29
$\begin{array}{c} 000000 & 1.011187 & 1.493,51 & 1.009944 & 1507.16 & 1.008473 & 1518.87 & 1.006794 & 1528.78 \\ 0.05788 & 1.012813 & 1497.71 & 1.011555 & 1511.05 & 1.010072 & 1522.75 & 1.008386 & 1535.90 \\ 0.10918 & 1.014259 & 1501.47 & 1.012987 & 1514.55 & 1.011494 & 1552.24 & 1.010979 & 1.515.99 \\ 0.15693 & 1.015587 & 1.504.97 & 1.014303 & 1.517.75 & 1.012798 & 1.52.48 & 1.011293 & 1.543.88 \\ 0.29398 & 1.019143 & 1.514.96 & 1.018025 & 1.524.10 & 1.01340 & 1.535.26 & 1.012303 & 1.541.95 \\ 0.24961 & 1.018133 & 1.514.96 & 1.018025 & 1.527.11 & 1.016496 & 1.538.78 & 1.014762 & 1.547.68 \\ \hline 0.00000 & 1.017278 & 1.496.44 & 1.016017 & 1.510.14 & 1.014530 & 1.521.80 & 1.01886 & 1.534.48 \\ 0.0498 & 1.018841 & 1.01773 & 1.514.96 & 1.01727 & 1.513.21 & 0.15778 & 1.524.90 & 1.016866 & 1.534.25 \\ 0.00000 & 1.017278 & 1.496.44 & 1.016017 & 1.510.14 & 1.014530 & 1.521.80 & 1.016866 & 1.534.26 \\ 0.04586 & 1.018861 & 1.01727 & 1.513.21 & 0.15778 & 1.524.90 & 1.016866 & 1.534.26 \\ 0.04586 & 1.018861 & 1.01727 & 1.513.21 & 0.107378 & 1.534.03 \\ 0.1444 & 1.020210 & 1.503.78 & 1.018881 & 1517.15 & 1.017379 & 1.536.00 & 1.016866 & 1.534.25 \\ 0.1444 & 1.022314 & 1.512.48 & 1.022349 & 1.522.26 & 1.019443 & 1.534.74 & 1.01773 & 1.543.03 \\ 0.32372 & 1.023768 & 1.512.48 & 1.023249 & 1.522.54 & 1.020823 & 1.536.74 & 1.017173 & 1.543.03 \\ 0.32372 & 1.023768 & 1.512.48 & 1.023249 & 1.525.54 & 1.020823 & 1.536.74 & 1.017173 & 1.543.94 \\ 0.28971 & 1.00578 & 1.499.57 & 1.005334 & 1509.36 & 1.008589 & 1.520.81 & 1.002179 & 1.535.94 \\ 0.09000 & 1.005243 & 1.499.57 & 1.006334 & 1.592.50 & 1.007439 & 1.533.81 & 1.002179 & 1.535.94 \\ 0.23991 & 1.000578 & 1.499.78 & 1.503.91 & 1.008485 & 1.520.18 & 1.00279 & 1.525.94 \\ 0.23995 & 1.01267 & 1.519.56 & 1.011631 & 1.533.48 & 1.01100 & 1.543.76 & 1.008215 & 1.529.40 \\ 0.49976 & 1.01234 & 1.510.99 & 1.00978 & 1.503.91 & 1.008405 & 1.522.18 & 1.006705 & 1.525.01 \\ 0.44067 & 1.01482 & 1.505.14 & 1.012377 & 1.518.41 & 1.010388 & 1.592.51 & 1.006755 & 1.525.97 \\ 0.23993 & 1.012697 & 1.519.56 & 1.011631 & 1.533.48 & 1.$			2-A1	minopropanoic Acid	in 0.09815 mol·kg	g^{-1} D-(+)-Cellobios	e		
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	0.00000	1.011187	1493.51	1.009944	1507.16	1.008473	1518.87	1.006794	1528.78
0.10918 1.01425 1.014297 1.51425 1.011494 1.526.5 1.009777 1.535.99 0.15939 1.015587 1.504.97 1.014303 1.517.75 1.012798 1.532.94 1.01095 1.533.04 0.19792 1.016171 1507.92 1.015422 1.520.60 1.013910 1.532.76 1.012303 1.541.95 0.29398 1.019343 1.514.65 1.010825 1.527.11 1.016496 1.538.78 1.014762 1.547.76 0.20000 1.017271 1.946.44 1.010171 1.511.41 1.014350 1.521.80 1.012866 1.538.22 0.04486 1.010210 1.503.78 1.01294 1.512.74 1.014444 1.531.41 1.016757 1.536.74 1.019134 1.544.48 0.14441 1.023136 1.512.48 1.022349 1.525.54 1.020823 1.556.74 1.019134 1.546.14 0.23272 1.023768 1.512.48 1.022349 1.525.40 1.020233 1.551.94 1.020663 1.543.34 <tr< td=""><td>0.05758</td><td>1.012813</td><td>1497.71</td><td>1.011555</td><td>1511.05</td><td>1.010072</td><td>1522.75</td><td>1.008386</td><td>1532.60</td></tr<>	0.05758	1.012813	1497.71	1.011555	1511.05	1.010072	1522.75	1.008386	1532.60
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.10918	1.014259	1501.47	1.012987	1514.55	1.011494	1526.24	1.009797	1535.99
0.19792 1.01671 1507.5 1.01570 1532.26 1.013910 1532.26 1.01201 1532.26 0.24961 1.018133 1511.65 1.016825 1524.01 1.015304 1535.73 1.015388 1545.38 0.29930 1.017278 1496.44 1.016017 1510.14 1.016496 1538.78 1.0142836 1531.55 0.04966 1.017278 1496.44 1.016017 1513.21 1.015778 1524.79 1.014084 1531.45 0.04941 1.020210 1503.78 1.018881 1517.15 1.017778 1524.79 1.014084 1533.57 0.14444 1.021311 1566.47 1.019954 1519.74 1.018448 1533.57 1.017773 1543.03 0.23272 1.023768 1524.84 1.022349 1522.26 1.019463 1533.57 1.00773 1543.03 0.23871 1.0233749 1516.55 1.022344 1522.54 1.002875 1529.49 0.00000 1.0065243 1490.47 1.0031.43	0.15693	1.015587	1504.97	1.014303	1517.75	1.012798	1529.48	1.011095	1539.04
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	0.19792	1.016717	1507.92	1.015422	1520.60	1.013910	1532.26	1.012203	1541.95
0.29398 1.019343 1514.96 1.018025 1527.11 1.01496 1538.78 1.014762 1547.76 0.200000 1.017278 1496.44 1.016017 1510.14 1.014530 1521.80 1.012836 1531.55 0.04986 1.010561 1499.66 1.017271 1513.21 1.015778 1524.79 1.014084 1538.42 0.14444 1.022310 1503.78 1.019854 1519.74 1.018448 1531.14 1.016755 1540.70 0.18219 1.022362 1509.09 1.020978 1552.54 1.020823 1538.77 1.01773 1543.03 0.23372 1.023349 1516.55 1.023885 1529.50 1.022349 1540.60 1.020663 1549.92 2.Amino-3-methylbutanoic Acid in 0.10201 mol·kg ⁻¹ Fructose 0.00464 1.000578 1525.54 0.003304 1534.34 0.14704 1.009097 1505.75 1.007823 1518.16 1.0003703 1548.44 0.03464 1.000274 1503.36 1.000478 1533.48 <td>0.24961</td> <td>1.018133</td> <td>1511.65</td> <td>1.016825</td> <td>1524.01</td> <td>1.015304</td> <td>1535.73</td> <td>1.013588</td> <td>1545.38</td>	0.24961	1.018133	1511.65	1.016825	1524.01	1.015304	1535.73	1.013588	1545.38
- LAminopropanoic Acid in 0.09059 mol·kg ⁻¹ pMelezitose 0.00000 1.017278 1496.44 1.016017 1510.14 1.014530 1521.80 1.012836 1531.55 0.0486 1.018561 1499.66 1.017271 1513.21 1.017379 1528.60 1.016986 1538.22 0.1444 1.021311 1506.47 1.019954 1519.74 1.01448 1531.14 1.016755 1.640.70 0.18219 1.022362 1590.09 1.022349 1552.54 1.0202331 1536.74 1.019134 1546.14 0.23871 1.023768 1512.48 1.022349 1545.5 1.023885 1529.50 1.022349 155.54 1.020831 1.600 1.000678 1495.71 1.00334 1.002149 1516.16 1.000875 1525.94 0.05040 1.000578 1495.75 1.007438 1537.21 1.004949 1534.85 1.003149 1534.84 0.14704 1.009097 1505.75 1.007823 1518.90 1.006326 1529.66 1.001334 1534.84	0.29398	1.019343	1514.96	1.018025	1527.11	1.016496	1538.78	1.014762	1547.76
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			2-	Aminopropanoic Ac	cid in 0.09059 mol	kg ⁻¹ D-Melezitose			
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	0.00000	1.017278	1496.44	1.016017	1510.14	1.014530	1521.80	1.012836	1531.55
0.10491 1.020210 1503.78 1.018881 1517.15 1.017379 1528.60 1.015686 1538.22 0.14444 1.021311 1506.47 1.019954 1519.74 1.018448 1531.14 1.01775 1540.07 0.13219 1.022362 1509.09 1.020978 1522.26 1.019463 1533.57 1.017773 1543.03 0.23272 1.023549 1516.55 1.023849 1525.54 1.022839 1536.74 1.019134 1546.14 0.28971 1.025349 1516.55 1.023885 1529.50 1.022349 1516.16 1.002075 1525.94 0.00000 1.005233 1.490.47 1.004013 1504.38 1.002549 1510.61 1.00875 1520.81 1.002179 1530.43 0.09454 1.0007734 1500.30 1.006478 1513.72 1.004994 1524.85 1.00304 1533.48 0.14704 1.00097 1505.75 1.007823 1518.90 1.006326 1529.26 1.004615 1538.98	0.04586	1.018561	1499.66	1.017271	1513.21	1.015778	1524.79	1.014084	1534.48
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	0.10491	1.020210	1503.78	1.018881	1517.15	1.017379	1528.60	1.015686	1538.22
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	0.14444	1.021311	1506.47	1.019954	1519.74	1.018448	1531.14	1.016755	1540.70
0.23272 1.023768 1.512.48 1.022349 1.525.54 1.020823 1.536.74 1.019134 1.546.40 0.28971 1.025349 1.516.55 1.023885 1.529.50 1.022349 1540.60 1.020663 1.549.92 0.00000 1.005243 1490.47 1.004013 1.504.38 1.002549 1516.16 1.000875 1.525.94 0.00500 1.00578 1495.71 1.005334 1509.36 1.003859 1520.81 1.002179 1530.43 0.09454 1.007734 1500.30 1.006478 1513.72 1.00494 1524.85 1.003304 1538.98 0.19140 1.010234 1510.09 1.008947 1523.06 1.007439 1533.89 1.005703 1542.44 0.23291 1.011290 1514.18 1.009988 1527.12 1.008470 1537.18 1.006705 1546.44 0.29956 1.012371 1518.41 1.0108405 1520.18 1.008284 1552.929 0.004000 1.0111123 1.4995.34 1	0.18219	1.022362	1509.09	1.020978	1522.26	1.019463	1533.57	1.017773	1543.03
0.28971 1.025349 1516.55 1.023885 1529.50 1.022349 154.60 1.020663 1549.92 2-Amino-3-methylbutanoic Acid in 0.10201 mol·kg ⁻¹ Fructose 0.00000 1.005243 1490.47 1.004013 1504.38 1.002549 1516.16 1.000875 1525.94 0.09404 1.006578 1495.71 1.003334 1509.36 1.003859 1520.81 1.002179 1530.43 0.09454 1.007734 1500.57.5 1.007873 1513.72 1.004932 1533.89 1.005703 1542.84 0.14704 1.00907 1505.75 1.007854 1527.12 1.008470 1533.89 1.005703 1542.84 0.23291 1.011290 1514.18 1.009986 1527.12 1.008470 1537.18 1.008755 1525.94 0.23956 1.01123 1495.34 1.009878 1508.91 1.008405 1520.18 1.008755 1529.29 0.04997 1.012393 1500.24 1.01134 1513.68 1520.18 1.007755 15	0.23272	1.023768	1512.48	1.022349	1525.54	1.020823	1536.74	1.019134	1546.14
2-Amino-3-methylbutanoic Acid in 0.10201 mol·kg ⁻¹ Fructose 0.00000 1.005243 1490.47 1.004013 1504.38 1.002549 1516.16 1.000875 1525.94 0.05040 1.006578 1495.71 1.005334 1509.36 1.003859 1520.81 1.002179 1530.43 0.09454 1.007734 1500.30 1.006478 1513.72 1.004994 1524.85 1.003304 1534.34 0.14704 1.009097 1505.75 1.007823 1518.90 1.006326 1529.66 1.004615 1538.98 0.19140 1.010234 1510.09 1.008947 1533.48 1.0100 1543.76 1.008244 1552.01 0.23291 1.012967 1519.56 1.011631 1533.48 1.0100 1543.76 1.008243 1552.01 0.00000 1.011123 1495.34 1.009878 1508.91 1.008405 1520.18 1.006725 1529.29 0.004997 1.012393 1500.24 1.011134 1516.86 1.009654 1524.71 1.007964	0.28971	1.025349	1516.55	1.023885	1529.50	1.022349	1540.60	1.020663	1549.92
0.000001.0052431490.471.0040131504.381.0025491516.161.0008751525.940.050401.0065781495.711.0053341509.361.0038591520.811.0021791530.430.094541.0077341500.301.0064781513.721.0049941524.851.0033041534.340.147041.0090971555.751.0078231518.901.0063261529.661.0046151538.980.191401.0102341510.091.0089471523.061.0074391533.891.0057031544.840.232911.0112901514.181.0099861527.121.0084701537.181.0087551.546.440.299561.0129671519.561.0116311533.481.0101001543.761.0082841552.012-Amino-3-methylbutanoic Acid in 0.10029 mol·kg ⁻¹ p.(+)-Cellobiose0.000001.0111231495.341.0098781508.911.0084051520.181.0067251529.290.049971.0123931500.241.0111341513.681.0096541524.711.0079641533.780.099831.0136491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.012411533.461.0103321542.460.139891.0159931514.311.0169281533.181.0124901541.911.0125651550.790.285871.018246152			2-A	mino-3-methylbutar	noic Acid in 0.1020	$1 \text{ mol} \cdot \text{kg}^{-1}$ Fructos	e		
0.050401.0065781495.711.0053341509.361.0038591520.811.0021791530.430.094541.0077341500.301.0064781513.721.0049941524.851.0033041534.340.147041.0090971505.751.0078231518.901.0063261529.661.0046151538.980.191401.0102341510.091.0089471523.061.0074391533.891.0057031542.840.232911.0112901514.181.0099861527.121.0084701537.181.0067051546.440.299561.0129671519.561.0116311533.481.011001543.761.0082841552.012-Amino-3-methylbutanoic Acid in 0.10029 mol·kg ⁻¹ p.(+)-Cellobiose0.000001.0111231495.341.0098781508.911.0084051520.181.0067251529.290.049971.0123931500.241.0111341513.681.0096541524.711.0079641533.780.099831.0136491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.0120411533.461.0103321542.460.193891.0171251518.721.0158171531.181.0142901541.911.0125651550.790.285871.0182461523.151.0169281532.441.0153801545.931.0132741532.650.052871.0190231502	0.00000	1.005243	1490.47	1.004013	1504.38	1.002549	1516.16	1.000875	1525.94
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	0.05040	1.006578	1495.71	1.005334	1509.36	1.003859	1520.81	1.002179	1530.43
0.147041.0090971505.751.0078231518.901.0063261529.661.0046151538.980.191401.0102341510.091.0089471523.061.0074391533.891.0057031542.840.232911.0112901514.181.0099861527.121.0084701537.181.0067051546.440.299561.0129671519.561.0116311533.481.0101001543.761.0082841552.01	0.09454	1.007734	1500.30	1.006478	1513.72	1.004994	1524.85	1.003304	1534.34
0.191401.0102341510.091.0089471523.061.0074391533.891.0057031542.840.232911.0112901514.181.0099861527.121.0084701537.181.0067051546.440.299561.0129671519.561.0116311533.481.0101001543.761.0082841552.012-Amino-3-methylbutanoic Acid in 0.10029 mol·kg ⁻¹ p.(+)-Cellobiose0.000001.0111231495.341.0098781508.911.0084051520.181.0067251529.290.049971.0123931500.241.0111341513.681.0096541524.711.0079641533.780.099831.016491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.0120411533.461.0103321542.460.193891.0159931514.311.0146991527.221.0131821537.761.0114681546.670.239931.0171251518.721.0158171531.181.0142901541.911.0125651550.790.285871.0182461523.151.0169281535.441.0133 mol·kg ⁻¹ p.Melezitose1.0132741532.050.005031.0201021507.061.018161520.471.0173051531.561.0135781540.760.138711.0211251511.221.0162441527.551.0145451536.810.096931.0201021507.061.01816 <td< td=""><td>0.14704</td><td>1.009097</td><td>1505.75</td><td>1.007823</td><td>1518.90</td><td>1.006326</td><td>1529.66</td><td>1.004615</td><td>1538.98</td></td<>	0.14704	1.009097	1505.75	1.007823	1518.90	1.006326	1529.66	1.004615	1538.98
0.232911.0112901514.181.0099861527.121.0084701537.181.0067051546.440.299561.0129671519.561.0116311533.481.0101001543.761.0082841552.012-Amino-3-methylbutanoic Acid in 0.10029 mol·kg ⁻¹ p.(+)-Cellobiose0.000001.0111231495.341.0098781508.911.0084051520.181.0067251529.290.049971.0123931500.241.0111341513.681.0096541524.711.0079641533.780.099831.0136491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.0120411533.461.0103321542.460.193891.0159931514.311.0146991527.221.0131821537.761.0114681546.670.239931.0171251518.721.0158171531.181.012901541.911.0125651550.790.285871.0182461523.151.0169281535.441.0153801545.931.0136561554.662-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p.Melezitose2-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p.Melezitose0.000001.0177191497.341.0164521511.221.0149651522.701.0132741532.660.000001.0177191497.341.0164521511.221.0162441527.551.0143451536.81<	0.19140	1.010234	1510.09	1.008947	1523.06	1.007439	1533.89	1.005703	1542.84
0.29956 1.012967 1519.56 1.011631 1533.48 1.010100 1543.76 1.008284 1552.01 2-Amino-3-methylbutanoic Acid in 0.10029 mol·kg ⁻¹ D-(+)-Cellobiose 0.00000 1.011123 1495.34 1.009878 1508.91 1.008405 1520.18 1.006725 1529.29 0.04997 1.012393 1500.24 1.011134 1513.68 1.009654 1524.71 1.009188 1533.78 0.09983 1.013649 1505.14 1.012377 1518.41 1.010888 1529.21 1.009188 1538.24 0.14687 1.014825 1509.70 1.013542 1522.84 1.012041 1533.46 1.010332 1542.46 0.19389 1.015993 1514.31 1.014699 1527.22 1.013182 1537.76 1.011468 1546.67 0.23993 1.017125 1518.72 1.015817 1531.18 1.014290 1541.91 1.012565 1550.79 0.28587 1.018246 1523.15 1.016928 1531.22 1.014965 1522.70	0.23291	1.011290	1514.18	1.009986	1527.12	1.008470	1537.18	1.006705	1546.44
2-Amino-3-methylbutanoic Acid in 0.10029 mol·kg ⁻¹ p-(+)-Cellobioset 0.00000 1.011123 1495.34 1.009878 1508.91 1.008405 1520.18 1.006725 1529.29 0.04997 1.012393 1500.24 1.011134 1513.68 1.009654 1524.71 1.007964 1533.78 0.09983 1.013649 1505.14 1.012377 1518.41 1.010888 1529.21 1.009188 1538.24 0.14687 1.014825 1509.70 1.013542 1522.84 1.012041 1533.46 1.010332 1542.46 0.19389 1.015993 1514.31 1.014699 1527.22 1.013182 1537.76 1.011468 1546.67 0.23993 1.017125 1518.72 1.015817 1531.18 1.014290 1541.91 1.012565 1550.79 0.28587 1.018246 1523.15 1.016928 1535.44 1.0133mol·kg ⁻¹ p-Melezitose 2-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p-Melezitose 0.00000 1.017719 1497.34 1.016452 1511.22 1.014965 1522.70 1.013274 1532.65 0.05287 </td <td>0.29956</td> <td>1.012967</td> <td>1519.56</td> <td>1.011631</td> <td>1533.48</td> <td>1.010100</td> <td>1543.76</td> <td>1.008284</td> <td>1552.01</td>	0.29956	1.012967	1519.56	1.011631	1533.48	1.010100	1543.76	1.008284	1552.01
0.000001.0111231495.341.0098781508.911.0084051520.181.0067251529.290.049971.0123931500.241.0111341513.681.0096541524.711.0079641533.780.099831.0136491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.0120411533.461.0103321542.460.193891.0159931514.311.0146991527.221.0131821537.761.0114681546.670.239931.0171251518.721.0158171531.181.0142901541.911.0125651550.790.285871.0182461523.151.0169281535.441.0153801545.931.0136561554.66C-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p-Melezitose0.000001.0177191497.341.0164521511.221.0149651522.701.0132741532.050.052871.0190231502.641.0177441516.271.0162441527.551.0145451536.810.096931.021021507.061.0188161520.471.0173051531.561.0155981540.760.138711.0211251511.221.0198281524.271.0183051535.341.0165891544.480.186331.022821515.951.0209711528.741.0194441539.601.0177141548.720.251001.0238431522.54 </td <td></td> <td></td> <td>2-Amino</td> <td>o-3-methylbutanoic</td> <td>Acid in 0.10029 mc</td> <td>$\mathrm{pl} \cdot \mathrm{kg}^{-1} \mathrm{D} \cdot (+) \cdot \mathrm{Cello}$</td> <td>biose</td> <td></td> <td></td>			2-Amino	o-3-methylbutanoic	Acid in 0.10029 mc	$\mathrm{pl} \cdot \mathrm{kg}^{-1} \mathrm{D} \cdot (+) \cdot \mathrm{Cello}$	biose		
0.049971.0123931500.241.0111341513.681.0096541524.711.0079641533.780.099831.0136491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.0120411533.461.0103321542.460.193891.0159931514.311.0146991527.221.0131821537.761.0114681546.670.239931.0171251518.721.0158171531.181.0142901541.911.0125651550.790.285871.0182461523.151.0169281535.441.0153801545.931.0136561554.66Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p-Melezitose0.000001.0177191497.341.0164521511.221.0149651522.701.0132741532.050.052871.0190231502.641.0177441516.271.0162441527.551.0145451536.810.096931.0201021507.061.0188161520.471.0173051531.561.0155981540.760.138711.0211251511.221.0198281524.271.0183051535.341.0165891544.480.186331.0222821515.951.0209711528.741.0194441539.601.0177141548.720.251001.0238431522.541.0225141535.041.0209701545.581.0192281554.42	0.00000	1.011123	1495.34	1.009878	1508.91	1.008405	1520.18	1.006725	1529.29
0.099831.0136491505.141.0123771518.411.0108881529.211.0091881538.240.146871.0148251509.701.0135421522.841.0120411533.461.0103321542.460.193891.0159931514.311.0146991527.221.0131821537.761.0114681546.670.239931.0171251518.721.0158171531.181.0142901541.911.0125651550.790.285871.0182461523.151.0169281535.441.0153801545.931.0136561554.66	0.04997	1.012393	1500.24	1.011134	1513.68	1.009654	1524.71	1.007964	1533.78
0.14687 1.014825 1509.70 1.013542 1522.84 1.012041 1533.46 1.010332 1542.46 0.19389 1.015993 1514.31 1.014699 1527.22 1.013182 1537.76 1.011468 1546.67 0.23993 1.017125 1518.72 1.015817 1531.18 1.014290 1541.91 1.012565 1550.79 0.28587 1.018246 1523.15 1.016928 1535.44 1.015380 1545.93 1.013656 1554.66	0.09983	1.013649	1505.14	1.012377	1518.41	1.010888	1529.21	1.009188	1538.24
0.19389 1.015993 1514.31 1.014699 1527.22 1.013182 1537.76 1.011468 1546.67 0.23993 1.017125 1518.72 1.015817 1531.18 1.014290 1541.91 1.012565 1550.79 0.28587 1.018246 1523.15 1.016928 1535.44 1.015380 1545.93 1.013656 1554.66 2-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p-Melezitose 0.00000 1.017719 1497.34 1.016452 1511.22 1.014965 1522.70 1.013274 1532.05 0.05287 1.019023 1502.64 1.017744 1516.27 1.016244 1527.55 1.014545 1536.81 0.09693 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54	0.14687	1.014825	1509.70	1.013542	1522.84	1.012041	1533.46	1.010332	1542.46
0.23993 1.017125 1518.72 1.015817 1531.18 1.014290 1541.91 1.012565 1550.79 0.28587 1.018246 1523.15 1.016928 1535.44 1.015380 1545.93 1.013656 1554.66 2-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ D-Melezitose 0.00000 1.017719 1497.34 1.016452 1511.22 1.014965 1522.70 1.013274 1532.05 0.05287 1.019023 1502.64 1.017744 1516.27 1.016244 1527.55 1.014545 1536.81 0.09693 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.19389	1.015993	1514.31	1.014699	1527.22	1.013182	1537.76	1.011468	1546.67
0.28587 1.018246 1523.15 1.016928 1535.44 1.015380 1545.93 1.013656 1554.66 2-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ D-Melezitose 0.00000 1.017719 1497.34 1.016452 1511.22 1.014965 1522.70 1.013274 1532.05 0.05287 1.019023 1502.64 1.017744 1516.27 1.016244 1527.55 1.014545 1536.81 0.09693 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.23993	1.017125	1518.72	1.015817	1531.18	1.014290	1541.91	1.012565	1550.79
2-Amino-3-methylbutanoic Acid in 0.10133 mol·kg ⁻¹ p-Melezitose 0.00000 1.017719 1497.34 1.016452 1511.22 1.014965 1522.70 1.013274 1532.05 0.05287 1.019023 1502.64 1.017744 1516.27 1.016244 1527.55 1.014545 1536.81 0.09693 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.28587	1.018246	1523.15	1.016928	1535.44	1.015380	1545.93	1.013656	1554.66
0.00000 1.017719 1497.34 1.016452 1511.22 1.014965 1522.70 1.013274 1532.05 0.05287 1.019023 1502.64 1.017744 1516.27 1.016244 1527.55 1.014545 1536.81 0.09693 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42			2-Am	ino-3-methylbutano	ic Acid in 0.10133	mol∙kg ^{−1} D-Melezit	ose		
0.05287 1.019023 1502.64 1.017/44 1516.27 1.016244 1527.55 1.014545 1536.81 0.09693 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.00000	1.017719	1497.34	1.016452	1511.22	1.014965	1522.70	1.013274	1532.05
0.09993 1.020102 1507.06 1.018816 1520.47 1.017305 1531.56 1.015598 1540.76 0.13871 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016589 1544.48 0.18633 1.02282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.05287	1.019023	1502.64	1.017/44	1516.27	1.016244	1527.55	1.014545	1536.81
0.130/1 1.021125 1511.22 1.019828 1524.27 1.018305 1535.34 1.016889 1544.48 0.18633 1.022282 1515.95 1.020971 1528.74 1.019444 1539.60 1.017714 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.09093	1.020102	150/.00	1.010820	1520.47	1.01/305	1531.50	1.015598	1540.76
0.10000 1.022602 1515.55 1.0207/1 1526.74 1.017444 1539.00 1.017/14 1548.72 0.25100 1.023843 1522.54 1.022514 1535.04 1.020970 1545.58 1.019228 1554.42	0.136/1	1.021123	1511.22	1.019020	1524.27	1.010303	1535.54	1.010309	1544.48
	0.25100	1.023843	1522.54	1.022514	1525.04	1.02.0970	1545.58	1.019228	1554.42

Table 1. Continued

	<i>T</i> /K = 293.15		<i>T</i> /K = 298.15		<i>T</i> /K = 303.15		T/K = 308.15	
т	$\rho \cdot 10^{-3}$	и	$\rho \cdot 10^{-3}$	и	$\rho \cdot 10^{-3}$	и	$\rho \cdot 10^{-3}$	и
$mol \cdot kg^{-1}$	kg·m ⁻³	m·s ⁻¹	kg•m ⁻³	$m \cdot s^{-1}$	kg·m ⁻³	$m \cdot s^{-1}$	kg•m ⁻³	m·s ⁻¹
0.27847	1.024512	1525.38	1.023175	1537.76	1.021625	1548.20	1.019869	1556.89
		2-Ar	nino-4-methvlpenta	noic Acid in 0.1032	$27 \mathrm{mol} \cdot \mathrm{kg}^{-1}$ Fructo	se		
0.00000	1.005339	1490.44	1.004110	1504.36	1.002649	1516.19	1.000978	1526.07
0.03514	1.006170	1494.76	1.004930	1508.64	1.003455	1520.14	1.001773	1529.86
0.04800	1.006471	1496.34	1.005226	1510.20	1.003747	1521.57	1.002061	1531.23
0.09415	1.007540	1501.98	1.006278	1515.82	1.004782	1526.73	1.003083	1535.62
0.09500	1.007559	1502.10	1.006296	1515.97	1.004800	1526.90	1.003101	1535.99
0.11577	1.008037	1504.08	1.006760	1518.53	1.005259	1528.35	1.003557	1537.62
0.14367	1.008669	1508.04	1.007377	1521.33	1.005868	1531.97	1.004160	1540.68
		2-Amino	-4-methylpentanoic	Acid in 0.10060 m	ol·kg ⁻¹ D-(+)-Cell	obiose		
0.00000	1.011139	1493.85	1.009895	1507.64	1.008424	1519.49	1.006744	1529.54
0.02884	1.011795	1497.34	1.010543	1511.09	1.009062	1522.74	1.007376	1532.58
0.04655	1.012198	1499.48	1.010942	1513.19	1.009455	1524.72	1.007764	1534.43
0.07528	1.012850	1502.94	1.011591	1516.60	1.010096	1527.92	1.008396	1537.40
0.09865	1.013380	1505.75	1.012119	1519.37	1.010617	1530.48	1.008910	1539.71
0.11448	1.013741	1507.65	1.012477	1521.28	1.010969	1532.25	1.009256	1541.34
0.14266	1.014376	1510.96	1.013117	1524.58	1.011603	1535.45	1.009878	1544.37
		2-Ami	no-4-methylpentanc	oic Acid in 0.09647	$mol \cdot kg^{-1}$ D-melezi	tose		
0.00000	1.016618	1495.94	1.015363	1509.64	1.013879	1521.46	1.012189	1531.21
0.03092	1.017311	1499.77	1.016042	1513.24	1.014548	1524.85	1.012851	1534.45
0.04754	1.017684	1501.82	1.016408	1515.16	1.014908	1526.66	1.013206	1536.18
0.07602	1.018320	1505.34	1.017034	1518.45	1.015525	1529.74	1.013816	1539.15
0.08835	1.018596	1506.80	1.017305	1519.84	1.015793	1530.97	1.014080	1540.41
0.10910	1.019058	1509.42	1.017760	1522.25	1.016241	1533.33	1.014520	1542.63
0.13503	1.019638	1512.50	1.018329	1525.19	1.016803	1536.10	1.015077	1545.20
			2-Amino-3-pł	nenylpropanoic Ac	id in Water			
0.00000	0.998219	1482.98	0.997050	1497.20	0.995645	1509.46	0.994020	1519.91
0.01916	0.999090	1485.30	0.997908	1499.39	0.996494	1511.52	0.994857	1521.85
0.04614	1.000308	1488.56	0.999107	1502.47	0.997681	1514.42	0.996027	1524.58
0.06873	1.001319	1491.30	1.000103	1505.05	0.998668	1516.85	0.996999	1526.87
0.07529	1.001611	1492.08	1.000391	1505.78	0.998953	1517.54	0.997280	1527.52
0.08564	1.002070	1493.33	1.000844	1506.95	0.999401	1518.62	0.997722	1528.55
0.10466	1.002911	1495.63	1.001671	1509.09	1.000223	1520.65	0.998531	1530.46
		2-Ar	nino-3-phenylpropa	noic Acid in 0.1073	$33 \mathrm{mol} \cdot \mathrm{kg}^{-1}$ Fructo	se		
0.00000	1.005628	1490.00	1.004394	1503.88	1.002929	1515.86	1.001261	1526.05
0.02149	1.006563	1492.65	1.005318	1506.43	1.003842	1518.25	1.002161	1528.32
0.04951	1.007782	1496.09	1.006524	1509.74	1.005034	1521.35	1.003336	1531.26
0.06322	1.008375	1497.77	1.007112	1511.35	1.005616	1522.85	1.003912	1532.68
0.07560	1.008911	1499.28	1.007642	1512.80	1.006143	1524.21	1.004427	1533.97
0.08562	1.009349	1500.49	1.008076	1513.96	1.006570	1525.29	1.004845	1535.01
0.09960	1.009952	1502.19	1.008685	1515.58	1.007163	1526.79	1.005432	1536.44
		2-Amino	-3-phenylpropanoic	Acid in 0.09980 m	ol·kg ⁻¹ D-(+)-Cell	obiose		
0.00000	1.011065	1495.24	1.009822	1508.89	1.008350	1520.78	1.006669	1530.87
0.02209	1.012044	1498.19	1.010781	1511.54	1.009301	1523.21	1.007614	1532.34
0.04927	1.013229	1501.83	1.011946	1514.80	1.010454	1526.20	1.008756	1536.19
0.06809	1.014036	1504.36	1.012742	1517.06	1.011240	1528.28	1.009530	1538.24
0.07631	1.014382	1505.46	1.013087	1518.04	1.011582	1529.17	1.009868	1539.11
0.08647	1.014806	1506.81	1.013511	1519.24	1.011995	1530.30	1.010280	1540.20
0.10652	1.015643	1508.76	1.014338	1521.70	1.012813	1532.58	1.011083	1542.38

	T/K = 293.15		<i>T</i> /K = 298.15		<i>T</i> /K = 303.15		<i>T</i> /K = 308.15			
т	$\rho \cdot 10^{-3}$	и								
$mol \cdot kg^{-1}$	$kg \cdot m^{-3}$	$m \cdot s^{-1}$	$kg \cdot m^{-3}$	$m \cdot s^{-1}$	$kg \cdot m^{-3}$	$m \cdot s^{-1}$	kg∙m ⁻³	$m \cdot s^{-1}$		
2-Amino-3-phenylpropanoic Acid in 0.09999 mol·kg $^{-1}$ D-Melezitose										
0.00000	1.017351	1496.19	1.016093	1509.91	1.014606	1521.72	1.012915	1531.60		
0.02263	1.018338	1499.08	1.017062	1512.63	1.015558	1524.28	1.013845	1533.95		
0.04925	1.019478	1502.50	1.018184	1515.84	1.016663	1527.29	1.014931	1536.68		
0.06759	1.020247	1504.88	1.018946	1518.06	1.017416	1529.36	1.015675	1538.57		
0.07459	1.020540	1505.79	1.019235	1518.89	1.017699	1530.13	1.015957	1539.28		
0.08884	1.021149	1507.53	1.019821	1520.56	1.018276	1531.70	1.016533	1540.74		
0.10485	1.021783	1509.60	1.020468	1522.14	1.018915	1533.28	1.017174	1542.34		

Table 1. Continued

cases show that solute—solute interactions are weak. Moreover, S_V is influenced by a number of effects.⁵¹

The partial molar volume and partial molar adiabatic compressibility of transfer at infinite dilution (i.e., ΔV_{ϕ}^{0} and $\Delta K_{\phi,s}^{0}$) of amino acids from pure water to aqueous saccharide solutions were calculated using the following equation:

$$\Delta Y = Y_{\phi}^{0}(\text{in aqueous saccharide solutions}) - Y_{\phi}^{0}(\text{in water})$$
(5)

The calculated values of partial molar volume of transfer at infinite dilution are given in Table S3 (given as SI) and illustrated in Figures F1 and F2 (given as SI).

The partial molar volume at infinite dilution of a nonelectrolyte is a combination of two factors:⁵²

$$V_{\phi}^{0} = V_{\rm int} + V_{\rm S} \tag{6}$$

where $V_{\rm int}$ is the intrinsic molar volume of the nonhydrated solute and $V_{\rm S}$ is the contribution due to the interaction of the solute with water. $V_{\rm int}$ is made up of following type of contribution:^{53,54}

$$V_{\rm int} = V_{\rm vW} + V_{\rm void} \tag{7}$$

where $V_{\rm vW}$ is the van der Waals volume ⁵⁵ and $V_{\rm void}$ is the volume associated with the voids and empty space present therein. ⁵⁶ This equation is later modified to evaluate the contribution of a solute molecule to its partial molar volume at infinite dilution as:

$$V_{2,m}^{0} = V_{\rm vW} + V_{\rm void} - n\sigma_{\rm S} \tag{8}$$

where $\sigma_{\rm S}$ is the shrinkage in the volume caused by the interaction of hydrogen bonding sites present in the solute with water molecules and *n* is the number of hydrogen bonding sites in molecule. The $V_{2,\rm m}^{0}$ of amino acids can be written as:

$$V_{2,m}^{0} = V_{vW} + V_{void} - V_{shrinkage}$$
(9)

By assuming that $V_{\rm vW}$ and $V_{\rm void}$ having same magnitude in water and aqueous saccharide solutions, the negative ΔV_{ϕ}^{0} values of 2-amino-3-methylbutanoic acid and 2-amino-4-methylpentanoic acid in aqueous fructose solution and 2-amino-3-phenylpropanoic acid in aqueous D-(+)-cellobiose solution are due to the lower decrease in $V_{\rm shrinkage}$ values.

lower decrease in $V_{\rm shrinkage}$ values. Further, ΔV_{ϕ}^{0} values can be explained on the basis of cosphere overlap model^{57,58} in terms of solute—cosolute interactions. According to this model, ionic-hydrophilic and hydrophilichydrophilic group interactions contribute positively, whereas hydrophilic-hydrophobic and hydrophobic-hydrophobic interactions contribute negatively to the ΔV_{ϕ}^{0} values. The negative ΔV_{ϕ}^{0} values of 2-amino-3-methylbutanoic acid, 2-amino-4methylpentanoic acid in aqueous fructose solution, and 2-amino-3-phenylpropanoic acid in aqueous D(+)-cellobiose solution indicate that the latter types of interactions are predominant over the former. From Table S3 and Figure F1 (SI), it shows that for 2-aminopropanoic acid and 2-amino-3-methylbutanoic acid in aqueous D-(+)-cellobiose and D-melezitose solutions comparable values of ΔV_{ϕ}^{0} are observed. Here one H-atom of 2-aminopropanoic acid is being replaced by the $-C(CH_3)_2$ group in 2-amino-3-methylbutanoic acid, resulting an increase of solventmediated solute-saccharide interactions, thereby reducing the electrostriction effect of the solvent. Also the magnitude of ΔV_{ϕ}^{0} decreases from 2-aminopropanoic acid, 2-amino-3-methylbutanoic acid, to 2-amino-4-methylpentanoic acid in aqueous fructose, D-(+)-cellobiose, and D-melezitose solutions (Figure F1, SI), which indicates the effect of the hydrophobic part. This is also due to the -CH3 group being replaced by -CH2CH- $(CH_3)_2$ while going from 2-aminopropanoic acid to 2-amino-4methylpentanoic acid, respectively. Hence hydrophilic-hydrophobic interactions are predominant than ionic-hydrophilic interactions, and the magnitude of ΔV_{ϕ}^{0} decreases. It can also be seen from Table S3 and Figure F3 (given as SI) that, while going from mono- to trisaccharide, the magnitude of ΔV_{ϕ}^{0} values increases. This shows that the ionic-hydrophilic and hydrophilic-hydrophilic group interactions are increasing with the change in the saccharide.

The $\Delta K_{\phi,s}^{0}$ values summarized in Table S3 of the SI show that $\Delta K_{\phi,s}^{0}$ values are positive except in case of 2-amino-4-methylpentanoic acid in aqueous fructose solutions at all temperatures. However both positive and negative values of $\Delta K_{\phi,s}^{0}$ are observed for 2-aminopropanoic acid and 2-amino-3-phenylpropanoic acid in aqueous D-(+)-cellobiose solutions. The $\Delta K_{\phi,s}^{0}$ values are increasing while going from mono- to trisaccharide. In the case of 2-amino-3-methylbutanoic acid, 2-amino-4-methylpentanoic acid, and 2-amino-3-phenylpropanoic acid (except at lower and higher temperatures), $\Delta K_{\phi,s}^{0}$ values increase almost linearly with the change in the saccharide. For 2-aminopropanoic acid (Figure F4 given as SI), there is a dip in $\Delta K_{\phi,s}^{0}$ values from aqueous fructose to D-(+)-cellobiose solution and then a sharp increase in aqueous D-melezitose solution indicating the presence

of hydrophobic-nonpolar interactions between the -OH group of the disaccharide and the $-CH_3$ group of the 2-aminopropanoic acid. These are also due to the disruption of hydration sphere of charged end centers of 2-aminopropanoic acid. As can be seen from Table S3, in aqueous fructose solution the magnitude of $\Delta K_{\phi,s}$ decreases from 2-aminopropanoic acid > 2-amino-3-methylbutanoic acid > 2-amino-3-phenylpropanoic acid > 2-amino-4-methylpentanoic acid. The magnitude of $\Delta K_{\phi,s}{}^0$ also decreases from 2-amino-3-methylbutanoic acid (although less in 2-aminopropanoic acid) to 2-amino-4-methylpentanoic acid in aqueous D-(+)-cellobiose solution. In the case of aqueous D-melezitose solution $\Delta K_{\phi,s}$ values are almost the same for all of the amino acids under study except for 2-amino-3-phenylpropanoic acid at lower temperatures. Positive $\Delta K_{\phi,s}^{0}$ values for the amino acids indicate the dominance of ionic/hydrophilic-hydrophilic interactions. As a result the cospheres of amino acids and saccharides overlap resulting in release of water to the bulk which is more compressible than the water present in the electrostricted region. The negative values of $\Delta K_{\phi,s}$ for 2-amino-3-methylbutanoic acid, 2-amino-4-methylpentanoic acid in aqueous fructose solutions, and 2-aminopropanoic acid and 2-amino-3-phenylpropanoic acid in aqueous D-(+)-cellobiose solutions, are due to the increase in hydrophobic side chain, which results in the disruption of hydration sphere of the charged centers of amino acids. Further, a positive contribution to $\Delta K_{\phi,s}^{0}$ gets reduced.

The V_{ϕ}^{0} values are related to the temperature as:

$$V_{\phi}^{0} = a + bT + cT^{2} \tag{10}$$

where *T* is the temperature in Kelvin and *a*, *b*, and *c* are constants.

The partial molar expansibility at infinite dilution, ϕ_E^0 , considered as a useful measure of solute—solvent interactions present in the solution is calculated by differentiating eq 10 with respect to temperature:

$$\phi_{\rm E}^{\ 0} = \left(\partial V_{\phi}^{\ 0} / \partial T\right)_{\rm P} = b + 2cT \tag{11}$$

The calculated values of ${\phi_{\rm E}}^0$ for the amino acids at different temperature are given in Table S4 (given as SI). The values of $\phi_{\rm E}^{0}$ decrease with the increase in temperature for all of the amino acids in aqueous saccharide solutions except for 2-aminopropanoic acid and 2-amino-4-methylpentanoic acid in aqueous fructose and 2-amino-3-phenylpropanoic acid in aqueous fructose and D-melezitose solutions. In case of 2-amino-3-methylbutanoic acid, the $\phi_{\rm E}{}^{0}$ values decrease with the increase in temperature in all aqueous saccharide solutions. The $\phi_{\rm E}^{0}$ values are positive in different saccharide solutions and at all temperatures in case of all of the amino acids (except for 2-aminopropanoic acid in aqueous D-melezitose solution) indicating the presence of solute-solute interactions. This reflects the release of electrostricted water from the loose solvation layers of the amino acids at higher temperatures and hence favoring amino acid-amino acid or amino acid-saccharide interactions. The values of partial molar expansibility give information regarding the size of the solute and its hydrophobocity.¹⁹ Table S4 shows that 2-amino-3-phenylpropanoic acid being the largest molecule among the amino acids studied (molecular wt. = 166.19188 g·mol⁻¹) has the highest $\phi_{\rm E}^{0}$ value (0.243 · 10⁻⁶ m³ · mol⁻¹ · K⁻¹) at T/K = 308.15 K.

CONCLUSION

Partial molar properties of some amino acids have been determined in water and saccharide—water solutions at different

temperatures. It was observed that these properties increase with the increase in temperature and also with the change in saccharide. All of these parameters support the existence of solute solvent interactions in the ternary mixture of present study. Amino acids with larger alkyl groups (2-amino-3-methylbutanoic acid, 2-amino-4-methylpentanoic acid, and 2-amino-3-phenylpropanoic acid) are undergoing hydrophobic hydration in the presence of saccharides as compared to 2-aminopropanoic acid.

ASSOCIATED CONTENT

Supporting Information. Apparent molar volume, apparent molar adiabatic compressibilities, apparent molar volume at infinite dilution, apparent molar adiabatic compressibilities at infinite dilution, their corresponding transfer functions, partial molar expansibilities at infinite dilution, and graphs of amino acids in saccharide—water solutions at different temperatures. This material is available free of charge via the Internet at http://pubs.acs.org.

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