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Magnetic Levitation in Analysis of Foods and Water

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ABSTRACT

This paper describes a method and a sensor that use magnetic levitation (MagLev) to characterize samples of food and water based on measurements of density. The sensor comprises two permanent NdFeB magnets positioned on top of each other in a configuration with like poles facing, and a container filled with a solution of paramagnetic ions. Measurements of density are obtained by suspending a diamagnetic object in the container filled with the paramagnetic fluid, placing the container between the magnets, and measuring the vertical position of the suspended object. We use MagLev to estimate the salinity of water, compare a variety of vegetable oils based on the ratio of polyunsaturated fat to monounsaturated fat, compare the content of fat in milk, in cheese, and in peanut butter, and determine the density of grains.

KEYWORDS: magnetic levitation (MagLev); density; analysis of food and water

INTRODUCTION

This paper describes a versatile technique based on magnetic levitation (MagLev)[1-13] for characterizing and distinguishing a variety of materials based on their density. We use MagLev to estimate the salinity of water, to distinguish different oils of plant origin based on their content of polyunsaturated and monounsaturated fat, to determine fat content in milk, cheese, and peanut butter, and to compare a variety of grains based on density.

All homogeneous matter has density. Changes in chemical composition or physical state (e.g., phase transition, crystallization, or purification) can result in changes in density. Density-based detectors of composition have the feature that they are universal (they respond to essentially all analytes), and do not require a chromophore (as do UV-Vis detectors).[14, 15] Density meters are used in research, industry, and healthcare to obtain information about the chemical composition of solid and liquid samples.[16] In solids, the density of polymers and minerals is commonly measured to assess crystallinity and purity, respectively.[1] The concentrations of solutes dissolved in fluids correlate with density as well; examples include measuring the content of sugar in soft drinks, the amount of alcohol in wine, the mole fraction of methanol in water, and the normality of sulfuric acid.[17] The chemical composition of bodily fluids also correlates with density. For instance, the density of urine can be used to assess dehydration and kidney function, and the density of blood correlates with hematocrit.[17, 18]

A variety of tools (e.g., floating bulb hydrometers[16], density gradient columns[19], pycnometers[16], oscillating-tube density meters[16], and suspended microchannel resonators[20, 21]) exist for measuring densities of solids and liquids and concentrations of solutes dissolved in them. These techniques involve tradeoffs between ease of operation, portability, and cost; for instance, density gradient columns and pycnometers offer high precision (10^{-4} g/cm^3) at the expense of portability and ease of operation, and require trained technicians to carry out accurate density measurements in centralized laboratories. Modern devices based on oscillating-tube technology[16] enable accurate measurements in a portable, automated, and high-throughput format, but cost several thousand dollars, and are applicable only to liquids with a limited range of viscosities. Suspended microchannel resonators (SMRs) enable the most sensitive measurements of density to date. SMRs are capable of measuring the densities of single cells and single particles (~50 nm – 3 μm in diameter) with a resolution of 10^{-4} g/cm^3, and detecting analytes binding to chemically functionalized microchannels and microspheres.[14, 20-23] Current designs of SMRs, however, are limited to analytes smaller than ~3 μm, require state-of-the-art facilities for fabrication, and carefully designed optics for detection.[14, 20-23] Many potential applications based on measurements of density would require (or benefit from) a method that is simple, inexpensive, portable, rapid, capable of measuring density values accurately using only microliter volumes of sample, and applicable to a wide variety of analytes (e.g., solids, liquids, gels, pastes, colloidal suspensions, and emulsions).

MagLev enables measurements of average density, and relative estimation of chemical composition based on differences in density.[9, 10, 12] The technique has six useful characteristics: i) it is applicable to a wide variety of analytes (solids, liquids, colloidal suspensions, gels, pastes), ii) it can be used with chemically heterogeneous and irregularly-shaped materials, iii) it is sensitive (capable of distinguishing densities of ± 0.02 to ± 0.0002 g/cm^3, depending on the experimental conditions; high precision is traded off against dynamic range), iv) it is compatible with samples with volumes ranging from 1 pL to 1 mL, v) it is simple (requiring only two NdFeB magnets and a vial containing a paramagnetic fluid), and vi) it is inexpensive, portable, and easy to use. MagLev is well suited for: i) making comparisons of samples based on density where density correlates with chemical composition, ii) monitoring chemical changes of a sample occurring over time, and iii) separating mixtures of materials into constituents (e.g., white from brown rice). This technique does not, however, provide information about the absolute chemical composition of a sample. MagLev should be particularly useful in situations where considerations of cost, simplicity, portability, and requirement for low sample volume or irregular sample shape outweigh the need for analyzing precise chemical composition of samples.

EXPERIMENTAL DESIGN

Choice of Analytes. We demonstrate the utility of MagLev with six different classes of analytes: water, oil, milk, cheese, grains, and peanut butter. We chose these analytes because they are common, practically relevant, and demonstrate unique capabilities of MagLev for analyzing a variety of substances (aqueous and organic liquids, colloidal suspensions, irregularly-shaped solids, and pastes).

Design of the Device. We measure densities of samples by levitating them in a paramagnetic solution placed between two NdFeB magnets (5 × 5 × 2.5 cm in length, width, and height, respectively) aligned parallel, 4.5 cm apart, with like poles facing one another (**Figure 1**). Diamagnetic samples levitate in this device when the gravitational force acting on the substance is balanced by the magnetic force (produced by the paramagnetic medium as a result of an applied magnetic field). The theory describing this balance is detailed elsewhere.[10]

Eq. 1 relates the density of the levitating sample ρ_s (kg/m^3) to its equilibrium levitation height h (m). In this equation, ρ_m (kg/m^3) is the density of the paramagnetic medium, g is the acceleration due to gravity, μ_0 (T·m·A^{-1}) is the permeability of free space, d (m) is the distance between the magnets, B_0 (Tesla) is the magnitude of the magnetic field at the surface of the magnets, and χ_m and χ_s (unitless) are the magnetic susceptibilities of the paramagnetic medium and the sample, respectively.

$$h = \frac{(\rho_s - \rho_m) g \mu_0 d^2}{(\chi_s - \chi_m) 4 B_0^{\ 2}} + \frac{d}{2} \qquad (1)$$

Sensitivity and Dynamic Range of MagLev. Analysis of samples by MagLev involves tradeoffs between the sensitivity of measurements and the dynamic range of densities that can be levitated within the same paramagnetic solution.[10] In the configuration of magnets described here, the magnetic susceptibility of the medium determines the ability of MagLev to resolve differences in density. This concept can be visualized by rearranging Eq. 1 into Eq. 2 to demonstrate that the resolution in levitation height (Δh) for levitating objects that differ in their densities by a fixed increment ($\Delta\rho_s$) can be tuned by adjusting χ_m (i.e., lowering χ_m will increase Δh).

$$\Delta h = \frac{g \mu_0 d^2}{(\chi_s - \chi_m) 4 B_0^{\ 2}} \Delta\rho_s \qquad (2)$$

Operating at maximum sensitivity (i.e., low concentration of paramagnetic ions in a solution that is already closely matched in density to the analyte), however, reduces the dynamic range of densities that can be levitated in the same medium.[10] Maximizing resolution, therefore, requires that the density of the medium be closely matched (within ± 0.001 – 0.005 g/cm^3) to the density of the analyte.

Choice of the Paramagnetic Solution. We use both aqueous and organic paramagnetic solutions for levitation. The paramagnetic solution must have the following characteristics: i) it must match the expected mean of the range of densities for the analyte (within 0.01 – 0.001

g/cm^3, depending on the experimental conditions) to enable an adequate balance of magnetic and gravitational forces required for levitation, and ii) it must be inert to the analyte (it should not swell, extract components from, mix with, or dissolve the analyte).

Levitating Organic and Water-Insoluble Samples. We use solutions of $MnCl_2$ or $GdCl_3$ in water for levitating water-insoluble samples. For levitating oils, we dissolve paramagnetic salts in a mixture of water and methanol because aqueous solutions are too dense for levitating oils. Typical solubilities of the major components of vegetable oils in methanol are 0.5–5 % (v/v).[24, 25] We expect that the miscibility of oils with methanol/water mixtures used in our experiments is below 0.5 % (v/v) and does not interfere with analysis on the time scale of the experiment (seconds).

Levitating Aqueous and Water-Soluble Samples. We use organic solutions of a hydrophobic chelate of Gd^{3+} (gadolinium(III) diethylenetriamine triacetic acid didecyldiacetamide (Gd(DTAD)) for levitating aqueous samples or samples that readily dissolve in water. This complex is soluble (0.5–1 M) in many organic solvents including alcohols (methanol, ethanol, octanol), aromatic hydrocarbons (chlorobenzene, nitrobenzene, toluene, 3-fluorotoluene), polar aprotic solvents (acetone, dimethylformamide, dimethylsulfoxide, acetonitrile, tetrahydrofuran, diethylether), aliphatic hydrocarbons (pentane, hexanes), and halogenated hydrocarbons (dichloromethane, chloroform, iodomethane, diiodomethane). It is *not soluble* in water. The substantial (up to 1 M) solubility of Gd(DTAD) in a variety of organic solvents expands the capabilities of MagLev as an analytical technique and enables levitation of aqueous droplets within organic solvents. To prevent the dissolution of aqueous analytes in the organic phase, we pre-saturate the organic solutions with water.

We prepared Gd(DTAD) in a nearly quantitative yield using a two-step procedure from commercially available starting materials (see Supporting Information for details). We used UV-Vis spectroscopy to determine that the DTAD ligand binds to Gd^{3+} with 1:1 stoichiometry.[26] Based on literature precedent for similar complexes, we expect this complex to have a stability constant of at least of $\sim10^{15}$ M^{-1}.[27, 28, 29] We also expect the entire complex to be neutral with an 8-point coordination geometry of the ligand to Gd^{3+} and much lower Lewis acidity than $GdCl_3$.[27, 28]

Sources and Characterization of Samples. Aqueous samples containing NaCl were prepared by dissolving NaCl in water to obtain desired concentrations. Expressed human milk used in this study was voluntarily provided by a lactating female. Remaining samples were obtained from commercial sources and used without further modification. Supporting Information describes the details for nutritional content and the sources of samples used in this study.

Statistical Treatment of Data. We used a single stock for each of the analytes (e.g., a bottle of oil, a bag of rice, a jar of peanut butter). A single measurement of levitation height involved generating the sample by withdrawing a small batch of the analyte from the stock (e.g., a droplet of oil, a grain of rice, a dollop of peanut butter), placing it into a container filled with paramagnetic medium, positioning this container between the magnets, and measuring the levitation height of the sample using a ruler. For liquids, we measured the levitation height from the center of the droplet, and for irregularly-shaped solids we measured the levitation height from the approximate vertical midpoint of the sample. We levitated liquid samples (oils, aqueous solutions of NaCl, and milk) in triplicate, and used the maximum deviation from the mean to estimate the error in the measurement of levitation height. We performed seven measurement for each of the solid samples (i.e., grains and cheese) and pastes (i.e., peanut butter), and used the standard deviation from the mean to estimate the error. Photographs of samples represent typical results from a single measurement. Supporting Information tabulates the values of levitation height, their corresponding uncertainties, and describes in detail how we calculate densities (and the associated uncertainties) of the samples based on their levitation height.

RESULTS AND DISCUSSION

I. Analyzing Liquid Samples

Distinguishing Plant Oils Based on Density. We levitated 5 μL samples of 16 different oils of plant origin in 50 mM $GdCl_3$ dissolved in 62% methanol and 38% water (v/v) (**Figure 2A**).[30] To establish a correlation between density of oils and their levitation height, we measured densities of oils using a portable density meter based on harmonic oscillator technology (DMA 35N, Anton Paar). The densities of these oils had an inverse linear correlation with their levitation height (**Figure 2B**).

The major chemical constituent of oils is fat. Different oils, however, are composed of different kinds of fat (monounsaturated, polyunsaturated, saturated, and trans fat) that are present in different proportions (see Supporting Information for details on the chemical composition of oils).

All oils that we examined contained the same amount of total fat (14 g per 15 mL). The monounsaturated and polyunsaturated fats were the major constituents of the total fat content in these oils (11–13 g per 15 mL), while saturated fat (1–2.5 g per 15 mL) and trans fat (0–0.5 g per 15 mL) were the minor constituents. The oils also varied substantially in their content of monounsaturated and polyunsaturated fats. The levitation height *h* of oils containing mostly *poly*unsaturated fat (e.g., hempseed and walnut oils) was lower than those containing mostly *mono*unsaturated fat (e.g., olive oil and sunflower oil). **Figure 2A** shows that the levitation height *h* correlates directly with the amount of *mono*unsaturated fat and inversely with the amount of *poly*unsaturated fat. The data presented in **Figure 2** suggest that levitation height (and density) of oils can be used to estimate the extent of unsaturation of fat in these oils (e.g., the amount of monounsaturated vs. polyunsaturated fat).

Estimating Salinity of Water. About 97% of Earth's water is saline. Water of moderate to high salinity (above 50–150 mM [NaCl]), however, has limited uses and is unsuitable for drinking or agriculture.[31] By measuring the density of aqueous solutions containing predominantly sodium chloride it is possible to estimate the salinity of water. We levitated aqueous solutions containing different concentrations of NaCl in 50 mM Gd(DTAD) in mixtures of 3-fluorotoluene and toluene (95:5, v/v), and correlated the concentration of NaCl of these samples with their density and levitation height (**Figure 3**). For this demonstration, we chose to examine the range of 0–200 mM [NaCl]; this range is relevant for determining the palatability of drinking water (0–30 mM NaCl) and suitability for irrigation (< 150 mM).[31]

II. Using MagLev to Compare Content of Fat in Milk, Cheese, and Peanut Butter.

Content of Fat in Milk. Milk is a colloidal emulsion of fat globules in a water-based fluid. Milk of animal origin contains significant amounts of fat, protein, and carbohydrates; the caloric content and exact chemical composition of milk varies widely between different species. Fat is an important constituent of milk that determines the nutritional value of dairy products. We demonstrate the ability to estimate the content of fat in milk by magnetic levitation. We compared levitation heights of individual droplets of milk suspended in 40 mM Gd(DTAD) in 84:16 2-fluorotoluene/chlorobenzene mixture (**Figure 4A**). The levitation height of milk correlated qualitatively with the content of fat within a sample. For example, skim and reduced-fat milk are denser (and, therefore, levitate closer to the bottom magnet) than whole milk because of the lower content of fat.

Content of Fat in Cheese. We levitated samples of low-moisture mozzarella cheese ("string cheese", containing 5 g vs. 2.5 g of total fat per 28 g) in 1.0 M $MnCl_2$. Samples of cheese with higher content of fat levitated above (further away from the bottom magnet) those with reduced fat.

Content of Fat in Peanut Butter. We also levitated samples of peanut butter in 1.0 M aqueous $MnCl_2$. We were able to distinguish between different formulations of peanut butter based on density, and we presume, on fat and carbohydrate content (**Figure 4B**). The two kinds of peanut butter (Skippy's creamy and Skippy's creamy reduced fat) we examined contained similar numbers of calories, but differed in their fat (16 g vs. 12 g per 30 mL, respectively) and carbohydrate content (7 g vs. 15 g per 30 mL, respectively). The peanut butter with higher fat/lower carbohydrate content is less dense, and levitates further away from the bottom magnet, than peanut butter with lower fat/higher sugar content.

III. Analysis of Grains.

Grains. Grains constitute an important source of calories worldwide. Grains are composed primarily of carbohydrates, protein, fat, and water; the ratio of these components determines the density of grains. We levitated five distinct kinds of grains (rice, barley, kamut, millet, and amaranth) and compared their densities using MagLev (**Figure 5A**). We also compared several kinds of rice (brown, white, purple, and black) based on density (**Figure 5A**). We levitated individual grains (at least seven different individual samples) in an aqueous solution of 0.475 M $GdCl_3$ + 4.5 M $CaCl_2$ and recorded their levitation height using a ruler with mm-scale markings.

The orientation of grains during levitation reflects the density distribution within the grain (i.e., the less dense part of the grain points upwards). We confirmed this observation by slicing the grain in half along the short axis to generate two halves of the grain that levitate at different heights (see Supporting Information for details).

Differences in levitation height between different kinds of rice corresponded qualitatively to differences in chemical composition. For instance, brown rice and white rice contain similar amounts of protein and carbohydrate, but differ in their fat content (brown rice being higher in fat); this difference in chemical composition is apparent from the differences in their levitation heights (**Figure 5B**). We found no statistically significant difference between different kinds of white rice and different kinds of brown rice (**Figure 5B**). Because MagLev does not yield information about chemical composition, it cannot be used to make conclusive statements about correlating complex chemical composition of different grains with their density. MagLev, however, is useful for analyzing systematic differences in chemical composition between two related samples (e.g., removal of husk, bran, and germ from brown rice to obtain white rice, or hydration status of grains during a process).

IV. Concluding remarks.

We conclude that MagLev is convenient method for measuring densities of food and water. We demonstrate the ability to levitate droplets of liquid (e.g., oil, milk, and aqueous solutions of salt) and irregularly-shaped solids and pastes (e.g., grains, cheese, and peanut butter), and correlate the levitation height (and density) of these materials with systematic variations in chemical composition (e.g., content of fat in milk, extent of unsaturation in vegetable oil, salinity of water). Potential applications of MagLev may include evaluating the suitability of water for drinking or irrigation, assessing the content of fat in food and beverages, or monitoring processing of grains (e.g., removing husk or drying).

MagLev offers a simple, inexpensive, and easy-to-use method for measuring densities of liquids and solids. MagLev has five useful attributes: i) it is applicable to a wide variety of analytes (solids, aqueous and organic solutions, colloidal suspensions, gels, and pastes), ii) it is compatible with objects that have irregular shapes and a broad range of volumes (1 pL to 1 mL), iii) it can be used with chemically heterogeneous samples, iv) it is rapid (density measurements can be performed within seconds to minutes), and v) it is accurate (± 0.02 – 0.0002 g/cm^3). MagLev also has several disadvantages: i) it requires a paramagnetic solution that may be incompatible with certain kinds of analytes (e.g., may cause swelling, extract components, or dissolve the analyte), ii) it involves a tradeoff between sensitivity and dynamic range, iii) it cannot measure densities of samples smaller than ~5 μm in diameter, and iv) it does not provide information about the precise chemical composition of a sample.

We believe MagLev will be broadly applicable as a density-based sensor of chemical composition. The technique is well-suited for general analysis of various samples based on density, monitoring changes in chemical or physical processes over time (e.g., extraction or dehydration), and separating heterogeneous mixtures into components (e.g., mixture of seeds that have different densities). MagLev may be particularly useful for analyzing samples based on density when the need for small sample volume, portability, simplicity, and low-cost are of primary importance.

AKNOWLEDGMENTS

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SUPPORTING INFORMATION AVAILABLE

Materials, methods, and details for the synthesis and characterization of Gd(DTAD); characterization of samples of food used in this study; details on using MagLev for calculating densities of samples and their uncertainties. This information is available free of charge via the Internet at http://pubs.acs.org.

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(30) The concentration of Gd^{3+} and composition of the medium were determined empirically to match approximately the density of the oils, to obtain the desirable dynamic range, and maximize the resolution between levitation heights of different kinds of oils. All samples of oils were then compared using this medium.

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Figure 1. Illustrations of MagLev device and levitating analytes. A) Schematic representation of the experimental set-up for MagLev. B) Photograph of a glass bead levitating in a cuvette filled with Gd^{3+} solution within a device used for MagLev. C) Photographs of various levitating samples: (i) a droplet of walnut oil levitating in 50 mM $GdCl_3$ in 62% methanol and 38% water (v/v); (ii) a droplet of water levitating in 50 mM Gd(DTAD) dissolved in 95:5 3-fluorotoluene/toluene (v/v); (iii) a droplet of bovine whole milk levitating in 40 mM Gd(DTAD) dissolved in 84:16 2-fluorotoluene/chlorobenzene; (iv) a piece of mozzarella cheese ("string cheese") levitating in 1.0 M $MnCl_2$; (v) a blob of peanut butter levitating in 1.0 M aqueous $MnCl_2$; (vi) a grain of brown rice levitating in aqueous 0.475 M $GdCl_3$ + 4.5 M $CaCl_2$. The appearance of spherical levitating droplets (i-iii) is distorted by the cylindrical shape of the vial. This distortion facilitates identification of the center of the drop.

Figure 1.

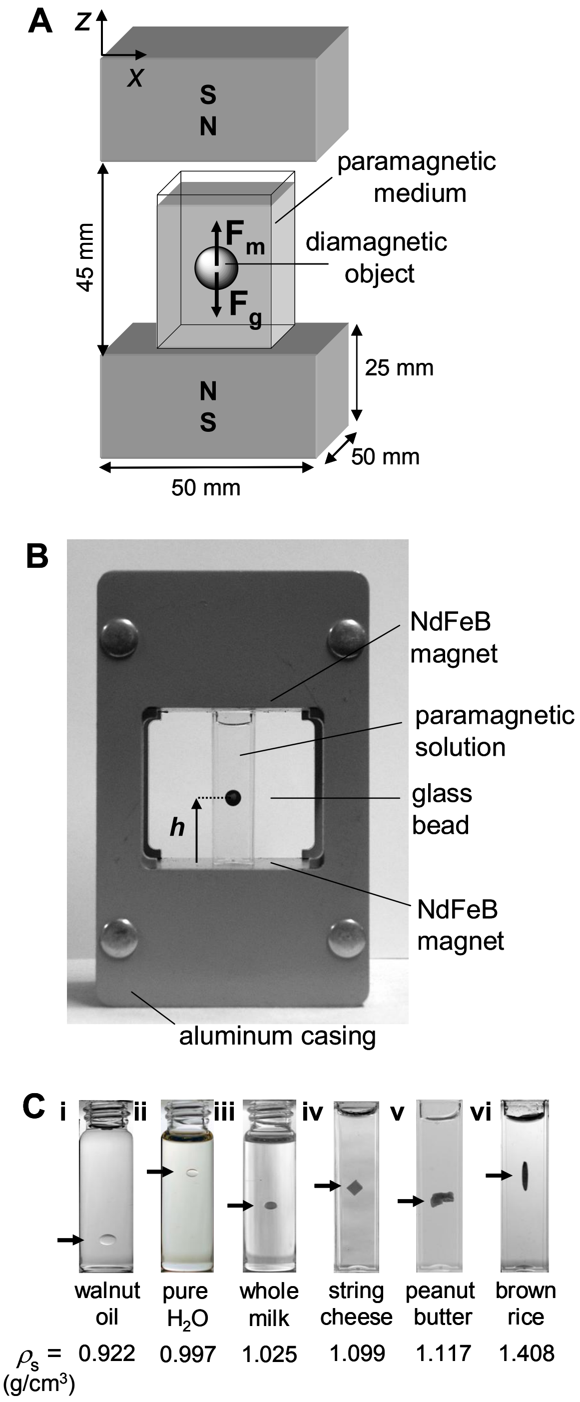

A
z
x
S
N
paramagnetic medium
F$_m$
diamagnetic object
F$_g$
45 mm
25 mm
N
S
50 mm
50 mm
B
NdFeB magnet
paramagnetic solution
glass bead
h
NdFeB magnet
aluminum casing
C
i
ii
iii
iv
v
vi
walnut oil
pure H_2O
whole milk
string cheese
peanut butter
brown rice
ρ_s = (g/cm^3)
0.922
0.997
1.025
1.099
1.117
1.408

Figure 2. Analysis of oils using MagLev. A) Plot correlating the amount of polyunsaturated (●) and monounsaturated (○) fat (14 g per 15 mL) with levitation height (●) of the corresponding oil droplets. B) Plot correlating the density of oils with their levitation height. Vertical error bars correspond to the maximum variation of individual measurement from the mean based on three independent measurements.

Figure 2.

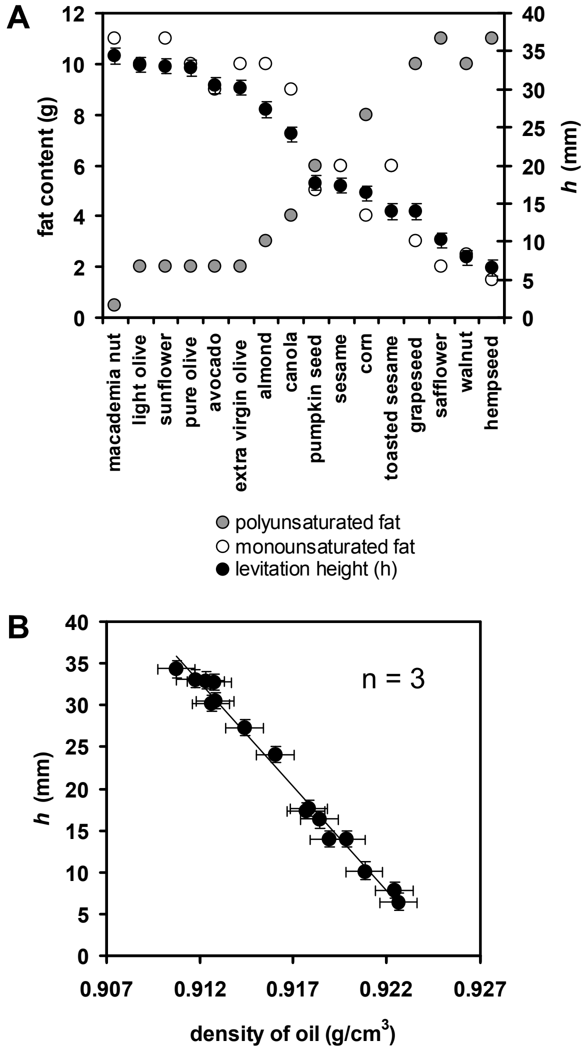

A
fat content (g)
12
10
8
6
4
2
0
40
35
30
25
20
15
10
5
0
h (mm)
macademia nut
light olive
sunflower
pure olive
avocado
extra virgin olive
almond
canola
pumpkin seed
sesame
corn
toasted sesame
grapeseed
safflower
walnut
hempseed
polyunsaturated fat
monounsaturated fat
levitation height (h)
B
h (mm)
40
35
30
25
20
15
10
5
0
n = 3
0.907
0.912
0.917
0.922
0.927
density of oil (g/cm3)

Figure 3. Plot correlating the levitation height of aqueous droplets with [NaCl]. Droplets were levitated in 50 mM Gd(DTAD) dissolved in 95:5 3-fluorotoluene/toluene (v/v). Data points represent average values from three independent measurements; error bars are represented by the size of the data points.

Figure 3.

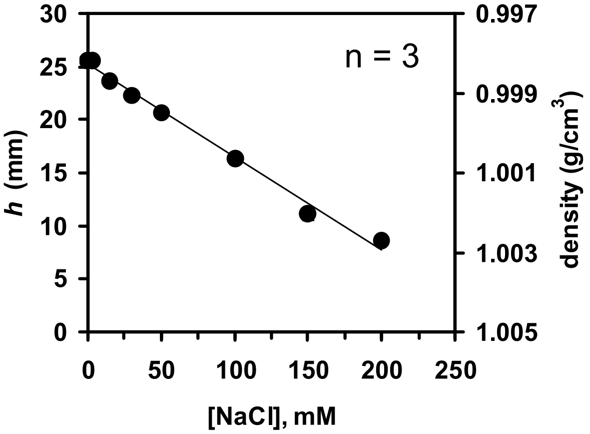

30
25
20
15
10
5
0
h (mm)
0.997
0.999
1.001
1.003
1.005
density (g/cm^3)
n = 3
0
50
100
150
200
250
[NaCl], mM

Figure 4. Comparison of food based on fat content. A) Photographs of milk droplets levitating in 40 mM Gd(DTAD) dissolved in 84:16 2-fluorotoluene/chlorobenzene. B) Photographs of "string cheese" samples with different fat content levitating in 1.0 M aqueous $MnCl_2$. C) Different kinds of peanut butter levitating in 1.0 M aqueous $MnCl_2$.

Figure 4.

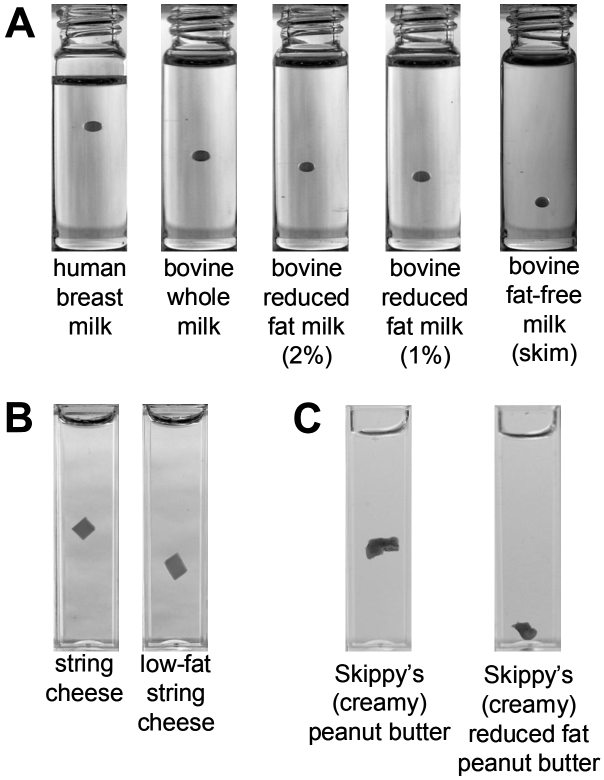
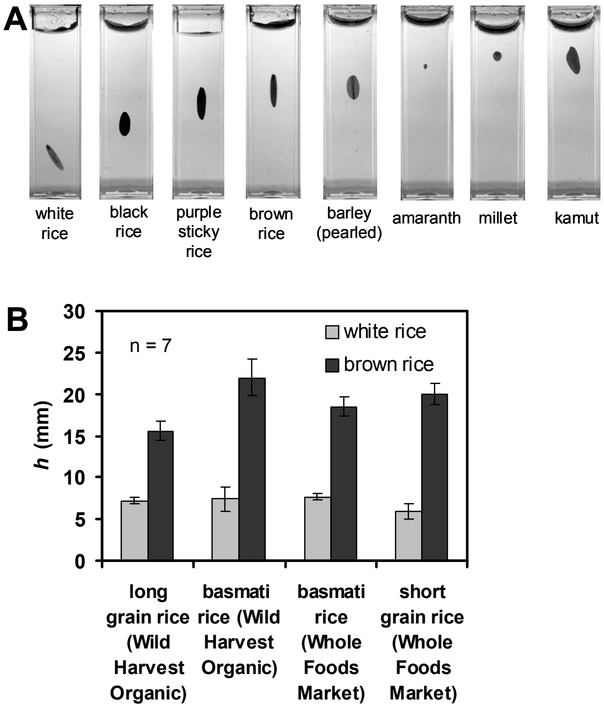

A
human breast milk
bovine whole milk
bovine reduced fat milk (2%)
bovine reduced fat milk (1%)
bovine fat-free milk (skim)
B
string cheese
low-fat string cheese
C
Skippy's (creamy) peanut butter
Skippy's (creamy) reduced fat peanut butter

Figure 5. Analysis of grains. A) Photographs of several kinds of grains levitating in 0.475 M $GdCl_3$ + 4.5 M $CaCl_2$. B) Bar graph comparing the levitation heights for different kinds of rice. All grains of rice were levitated in aqueous solution of 0.475 M $GdCl_3$ + 4.5 M $CaCl_2$. Error bars represent the standard deviation from seven independent measurements of different rice grains from the same rice stock.

Figure 5.


A
white rice
black rice
purple sticky rice
brown rice
barley (pearled)
amaranth
millet
kamut
B
n = 7
white rice
brown rice
h (mm)
0
5
10
15
20
25
30
long grain rice (Wild Harvest Organic)
basmati rice (Wild Harvest Organic)
basmati rice (Whole Foods Market)
short grain rice (Whole Foods Market)

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