Brevia

SHORT NOTES

Drops, surface tension and diapir models

ROBERTO FERREZ WEINBERG

Hans Ramberg Tectonic Laboratory, Institute of Earth Sciences, University of Uppsala, Box 555, S-751 22 Uppsala, Sweden

(Received 1 May 1992; accepted in revised form 7 September 1992)

Abstract—Dynamic similarity requires that laboratory models simulating geological diapirs must not be significantly affected by surface tension. This Short Note reviews the effects of surface tension and concludes that quantitative results of small model diapirs where the buoyancy force is close to the value of surface tension (Bond number, B_o , close to unity) may not be relevant to natural diapirs. To avoid the effects of surface tension ($B_o \gg 1$), physical models should be as large as possible, and use miscible fluids with low chemical diffusion rate and significant buoyancy.

INTRODUCTION

SEVERAL authors have used laboratory models to quantify processes related to natural diapirism in the mantle and crust (e.g. Ramberg 1967, Whitehead & Luther 1975, Talbot 1977, Marsh 1979, Kerr & Lister 1988, Rönnlund 1989, 1991, Griffiths & Campbell 1991, Neavel & Johnson 1991). According to scale-model theory, there must be geometric, kinematic and dynamic similarity between physical model and the natural analogue in order for the model evolution to simulate the natural prototype (Ramberg 1967). The large scale of geological diapirs renders the effects of surface tension negligible (Marsh 1979). Thus, in order to fulfil the condition of dynamic similarity, laboratory models should also be unaffected by surface tension. Talbot (1977) recognized that his and possibly many other models were affected by surface tension, and might therefore not be ideal for modelling large-scale geological structures. Griffiths et al. (1989) observed and discussed the effects of surface tension in their experiments, and Jackson et al. (1988) are among the few to consider the limitations on model size imposed by the surface tension of model materials. However, some recent laboratory models have studied the rise of drops with such low buoyancy forces (small radii and/or low density difference to the ambient fluid) that they run the risk of being swamped by the effects of surface tension (e.g. Marsh 1979, Neaval & Johnson 1991). This note illustrates this problem and draws some simple guidelines for laboratory experiments designed to explore geological diapirism.

SURFACE TENSION AND MARANGONI EFFECT

Intermolecular cohesive forces acting on any interface between two media cause a force per unit length of surface (or a free energy per unit area), called surface tension γ . At distances from the bounding surface less than the range of action of the cohesive forces (10^{-9} m for simple molecules, Batchelor 1967) these forces may become unbalanced and cause the liquid molecules to move inwards and shrink the interface area (for positive γ) or outwards and expand it (for negative γ). An expanding interface leads to a rapid and complete mixing of the fluids. If, however, the viscosity contrast is larger than 10, mixing between the two fluids is a lengthy process.

Static drops of one fluid inside another assume spherical shapes; a fluid cylinder breaks into small spheres; and small spheres aggregate to form large spheres. All these phenomena result from the minimization of surface free energy. Spheres have the minimum surface free energy because they have the smallest area per unit volume. Large spheres have smaller surface free energy per unit volume than small spheres. The same principle causes drops of high surface tension to keep spherical shapes when subjected to shearing whereas low surface tension drops will easily deform into ellipsoids (Bentley & Leal 1986). In addition, it causes a dampening of short wavelength instabilities in layer or line sources (Levich & Krylov 1969) so that the wavelength developed may be controlled by surface tension. However, it is interesting to note that a translating drop with neglegible inertia (Reynolds number, $Re \ll 1$) will also assume a spherical shape due to stress distribution around it, even when there is no surface tension affecting the system (Batchelor 1967).

Tate's law relates the weight of a drop of viscous fluid that detaches from a tip, to the surface tension (Adamson 1960). A modified Tate's law shows that the radius rof the falling (or lifting-off) drop increases with the surface tension, according to: $r=(3\gamma/4\Delta\rho g)^{0.5}$, where $\Delta\rho$ is the density difference between the drop and its sur-



Fig. 1. The immobile cap at the base of a rising viscous drop caused by the rearrangement of surfactants. The front of the drop remains mobile (redrawn from Griffith 1962).

rounding liquid and g is the gravitational acceleration. Tsuge (1986) presents a set of equations that relate the volume of bubbles detaching from a source nozzle $(V_{\rm b})$, to several controlling variables. In experiments where bubbles (or drops) slowly grow and detach from a nozzle (neglegible inertia), there are two relevant equations. The first equation calculates $V_{\rm b}$ for a statically growing bubble as a function of surface tension (gas flow rate into the bubble tends to zero; Tsuge 1986, equation 4), and the second calculates $V_{\rm b}$ for a bubble growing from a constant input of gas or liquid (constant flow rate into the bubble), where surface tension is disregarded (Tsuge 1986, equation 19). The larger of the two $V_{\rm b}$ values resulting from these equations will be the observed detachment volume. Thus, such experiments need to show clearly whether the detachment volume observed is the result of the control of γ , or of the control of the flow rate into the bubble.

Gradients in surface tension may arise along an interface at rest due to variations in chemical concentration or temperature. Such gradients will drive motion along a liquid interface (Marangoni effect; Levich & Krylov 1969) resulting in emulsification or local breaking up of the interface initially at rest. Marangoni effect caused by surface tension gradients due to temperature variations is particularly interesting for experiments which involve drops rising through liquids of variable temperature distribution (Harper *et al.* 1967), or hot drops rising through cooler ambient fluid (Head & Hellums 1966). Two non-dimensional numbers relate surface tension to the forces which are important during translation of drops. The Bond number, B_o , expresses the ratio of buoyant forces to surface tension, and the Capillary number, C_a , expresses the ratio of the viscous forces to surface tension (Weast & Astle 1981):

$$B_{\rm o} = \Delta \rho r^2 \mathbf{g} / \gamma \tag{1}$$

$$C_{\rm a} = \eta_{\rm s} v / \gamma, \tag{2}$$

where η_s is the dynamic viscosity of the sphere and ν is the velocity. For values of B_o and $C_a < 1$, surface tension dominates the buoyancy or the viscous forces, respectively. The radius at which the B_o or $C_a = 1$ is called the critical radius, r_c (Bond & Newton 1928). Surface tension is a function of the area, whereas buoyancy is a function of the volume; thus decreasing sphere radii or $\Delta \rho$ causes a relative increase in the influence of surface tension. Surface tension values of some materials commonly used to model diapirs are found in Miner & Dalton (1953), Weast & Astle (1981), Weijermars (1986) and Jackson *et al.* (1988).

THE RISE OF SMALL VISCOUS DROPS

While measuring the velocity of slowly rising viscous drops, Bond & Newton (1928) were the first to observe that viscous spheres smaller than a critical radius (r_c) have the velocity of solid spheres. They found r_c to be a

Fig. 2. (a)–(d) Model simulating the ascent of composite diapirs where surface tension causes solid behaviour of the two small drops on the right-hand side. The buoyant drops are formed by two miscible fluids, Hyvis 3 ($\eta = 7-8$ Pa.s and $\rho = 870$ kg m⁻³) and Shell's callibrating oil 11 ($\eta = 9$ Pa.s and $\rho = 930$ kg m⁻³), rising through immiscible denser Reppos glucose 40 ($\eta = 10^2$ Pa.s and $\rho = 1420$ kg m⁻³), bounded by a Plexiglass cylinder 10 cm in diameter and in height. The rigid behaviour of the two small drops on the right-hand side is demonstrated in two ways. First, the lack of internal circulation, shown by stable internal stratification, with the denser dark oil remaining at the lower half of the sphere throughout the surface. Drop 1, on the left-hand side, is larger than the critical radius r_c and behaves as a viscous sphere circulating internally. All the fluids used are Newtonian. Hyvis is an artificial polybutene oil produced by BP Chemicals Ltd, and the glucose used is produced by Reppos Glucos AB.



Fig. 2

function of the surface tension and to become critical when the ratio between buoyancy force and surface tension given by the Bond number is unity $(B_0 = 1)$. Frumkin & Levich (1947) attributed such behaviour to the presence of surfactants on the sphere's surface. Surfactants generally decrease the surface tension of a liquid. A motionless drop may have surfactants uniformly distributed at its surface and any motion sweeps the surfactant molecules towards the rear of the drop, creating a surface tension gradient $\Delta \gamma$. According to Frumkin & Levich, it is $\Delta \gamma$, rather than γ alone, that causes a drop to behave like a solid sphere. The $\Delta \gamma$ creates tangential forces along the surface (Marangoni effect) that retard surface motion and hence the velocity of the sphere (Levich & Krylov 1969). Experiments by Griffith (1962) have shown that surfactants collect in an immobile cap at the base of the drop, while the front remains mobile (Fig. 1). The cap opposes movement along the surface of the drop and affects its effective viscosity. The drop's effective viscosity therefore depends on the size of the cap, which is a function of the amount of surfactants, and on the radius of the sphere. According to Griffith, the apparent success of Bond & Newton's equation (1) is because very commonly $\Delta \gamma = \gamma$.

The effect of surface tension on small spheres is illustrated here by an experiment designed to study the patterns of internal circulation in drops formed by two fluids simulating composite diapirs. Figure 2 shows several steps in the slow rise of three drops through an ambient fluid ($\text{Re} \approx 10^{-6}$). The larger sphere, 1, circulates internally as it ascends, indicating transmission of viscous forces across and slip at the surface of the sphere. The patterns developed in drop 1 (Fig. 2) give an insight into the three-dimensional internal circulation of composite diapirs, and closely simulate the two-dimensional patterns shown in Weinberg (1992).

On the other hand, the two smaller spheres, 2 and 3 (Fig. 2), show neither movement of the surface (as depicted by the lack of disturbance of the oil film patterns on the surface of sphere 2 as it ascends) nor internal circulation. These two observations imply noslip along the surface of these small low-viscosity drops, indicating effectively rigid behaviour caused by surface tension. The exact volume of each fluid in the spheres in Fig. 2 and their average surface tensions or densities are all unknown. However, the critical radius for this experiment obviously lies between the radius of the large and the small spheres, i.e. between 2 and 3 mm. Three possible explanations may account for the observed rigidity of drops 2 and 3 in an experiment where no surfactants were intentionally added: (i) small quantities of surfactants were present in the original liquids; (ii) small impurities and dust particles may create a $\Delta \gamma$ (R. Griffiths personal communication 1991); or (iii) presence of surface tension gradients $\Delta \gamma$ due to heterogeneous distribution of the two internal fluids along the surface of the drops.

A LIMITING FACTOR TO THE SIZE OF LABORATORY MODELS

The increasing importance of surface tension, or surfactants, as the size of laboratory models decrease, imposes a size limit below which diapir models are no longer comparable to large-scale natural analogues (B_{0}) \leq 1). If in laboratory models $B_0 \leq 1$, then changes in parameters controlling the structures will be partly or totally obscured by the effects of surface tension. These effects will cause immiscible viscous drops to rise at the velocity of solid spheres (Fig. 2); cause the characteristic wavelength of instabilities developing on a rising immiscible cylinder to remain unchanged when the viscosity of the cylinder is varied; delay detachment of drops from their source nozzle to higher radii (volume) than those predicted by calculations which do not consider surface tension effects (as in Marsh 1979, Neavel & Johnson 1991); and obscure any small dependence of model diapir radii on the angle of inclination of the source (Marsh 1979).

Because of their low surface tension, miscible fluids with slow diffusion rate may be more appropriate for diapir models. However, miscible fluid models may be affected by changes in viscosity caused by inter-diffusion of species due to a gradient of chemical potential across the surface (R. Griffiths personal communication 1991). For example, Griffiths & Campbell (1991) noted, in models with drops formed by glycerol rising through glucose syrup, that water is diffused from the syrup into the glycerol, causing a very thin layer of syrup adjacent to the drop surface to dry and stiffen, with significant dynamical consequences for their models. They also found that the opposite occurs in drops of aqueous solution of K₂CO₃ rising through syrup, in which case water diffused into the syrup and produced a thin lowviscosity layer of syrup surrounding the drop. This outward diffusion of water did not influence the motion of their large drops (>1 cm), but would affect the rise velocity and internal circulation of small drops. For small drops with small Stokes velocity, chemical diffusion can also lead to entrainment of the surrounding fluid and mixing (discussed in Griffiths 1986).

It has been suggested that surface tension may be used to model the effects of a somewhat rigid shell formed around a rising magmatic diapir as it cools and solidifies (B. Marsh personal communication 1992). Although the rigid shell and surface tension may act in a similar way, their origins are different and whereas the effect of surface tension is a function of the drop's radius, the effect of the rigid shell is complex and depends on a series of factors, such as the radius of the diapir, the thickness and growth rate of the shell and its effective viscosity and viscosity contrast to the magma. Thus, using surface tension to simulate the rigid shell may be misleading until the effects of the latter on the rising diapir are clearly understood.

CONCLUSIONS

The effect of surface tension and surfactants must be recognized in laboratory models dealing with very small structures developing in immiscible fluids (mm size). This is a necessary condition if quantitative results are to be extended from models to natural geological structures such as magmatic diapirs. Surface tension tables may be found, for example, in Weast & Astle (1981), and several methods available for measuring surface tension are described in the literature (e.g. Adamson 1960, Shaw 1980). In order to achieve high B_o and C_a and avoid significant surface tension effects, models should use miscible fluids of slow chemical diffusion and large drop buoyancy (large radius and density difference). The effects of chemical diffusion on the viscosity of the miscible liquids should also be verified.

Acknowledgements—I would like to thank Drs Bruce Marsh, Ross Griffiths, Ross Kerr, Christopher Talbot and Stefan Bergman for critical reading and discussion; and Göran Rämme and Peter Rönnlund for suggestions and discussion. The assistance of Sharon Ford for language editing and Bertil Giös for developing the photographs are gratefully acknowledged.

REFERENCES

- Adamson, A. W. 1960. *Physical Chemistry of Surfaces*. Interscience, New York.
- Batchelor, G. K. 1967. An Introduction to Fluid Dynamics. Cambridge University Press, Cambridge.
- Bentley, B. J. & Leal, L. G. 1986. An experimental investigation of drop deformation and breakup in steady, two-dimensional linear flows. J. Fluid Mech. 167, 241–283.
- Bond, W. N. & Newton, D. A. 1928. Bubbles, drops and Stokes' Law. *Phil. Mag.* **5**, 794–800.
- Frumkin, A. N. & Levich, V. G. 1947. On the influence of surfaceactive substance on movement at the boundary in liquid media (in Russian). *Zhur. Fiz. Khim.* 21, 1183–1204.
- Griffith, R. M. 1962. The effect of surfactants on the terminal velocity of drops and bubbles. *Chem. Engng Sci.* 17, 1057–1070.
- Griffiths, R. W. 1986. The differing effects of compositional and

thermal buoyancies on the evolution of mantle diapirs. *Phys. Earth & Planet. Interiors* **43**, 261–273.

- Griffiths, R. W. & Campbell, I. H. 1991. Interaction of mantle plume heads with the Earth's surface and onset of small-scale convection. *J. geophys. Res.* **96**, 18,295–18,310.
- Griffiths, R. W., Gurnis, M. & Eitelbeg, G. 1989. Holographic measurements of surface topography in laboratory models of mantle hotspots. *Geophys. J.* 96, 477–495.
- Harper, J. F., Moore, D. W. & Pearson, J. R. A. 1967. The effect of the variation of surface tension with temperature on the motion of bubbles and drops. J. Fluid Mech. 27, 361–366.
- Head, H. N. & Hellums, J. D. 1966. Heat transport and temperature distributions in large single drops at low Reynolds numbers: a new experimental technique. A.I. Ch. E. J. 12, 553–559.
- Jackson, M. P. A., Talbot, C. J. & Cornelius, R. R. 1988. Centrifuge modelling of the effect of aggradation and progradation on syndepositional salt structures. Univ. Texas at Austin, Bur. Econ. Geol. NTIS, 173.
- Kerr, R. C. & Lister, J. R. 1988. Island arc and mid-ocean ridge volcanism, modelled by diapirism from linear source region. *Earth Planet. Sci. Lett.* 88, 143–152.
- Levich, V. G. & Krylov, V. S. 1969. Surface-tension-driven phenomena. Annu. Rev. Fluid Mech. 1, 293–316.
- Marsh, B. D. 1979. Island arc development: some observations, experiments and speculations. J. Geol. 87, 687–713.
- Miner, C. S. & Dalton, N. N. 1953. *Glycerol*. Reinhold, New York. Neavel, K. E. & Johnson, A. M. 1991. Entrainment in compositio-
- nally buoyant plumes. Tectonophysics 200, 1-15. Ramberg, H. 1967. Gravity, Deformation and the Earth's Crust in
- Theory, Experiments and Geological Application. Academic Press, London.
- Rönnlund, P. 1989. Viscosity ratio estimates from Rayleigh-Taylor instabilities. *Terra Nova* 1, 344–348.
- Rönnlund, P. 1991. Diapiric walls and stocks from layer and line sources. Unpublished Ph.D. thesis, Uppsala University.
- Shaw, D. J. 1980. Introduction to Colloid and Surface Chemistry (3rd edn). Butterworth, London.
- Talbot, C. J. 1977. Inclined and asymmetric upward moving gravity structures. *Tectonophysics* 42, 159–181.
- Tsuge, H. 1986. Hydrodynamics of bubble formation from submerged orifice. In: *Encyclopedia of Fluid Mechanics*, Vol. 3 (edited by Cheremisinoff, N. P.). Gulf Publishing Company, Houston, 191– 232.
- Weast, R. C. & Astle, M. J. 1981. Handbook of Chemistry and Physics. Handbook Series. CRC Press, Boca Raton.
- Weijermars, R. 1986. Finite strain of laminar flows can be visualized in SGM36-polymer. *Naturwiessenschaften* 73, 33–34.
- Weinberg, R. F. 1992. Internal circulation in a buoyant two-fluid Newtonian sphere: implications for composed magmatic diapirs. *Earth Planet. Sci. Lett.* **110**, 77–94.
- Whitehead, J. A. & Luther, D. S. 1975. Dynamics of laboratory diapirs and plume models. J. geophys. Res. 80, 705-717.