## S. G. MASON—A RETROSPECTIVE

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Without exaggeration one can say that Professor Stanley G. Mason was one of the founders of the science of microrheology. His early interest in this field was aroused when trying to understand the complex behavior of papermaking suspensions, the obvious relevance of which became evident after joining the Pulp and Paper Research Institute of Canada (Paprican) in 1946. Besides his publications with Professor Otto Maass from his thesis work on critical phenomena, his early work deals almost exclusively with the hydrodynamic and colloidal aspects of pulp suspensions. He realized that in order to understand the complexity of such systems, it was necessary to study the properties of the individual pulp fibers that make up the suspension. In this respect the classical work of Jeffery (1922) on the motion of ellipsoidal particles in linear shear flows was extremely relevant. Mason compared a pulp fiber with a long slender spheroid and applied Jeffery's theory to the motion of single suspended fibers. Over the years a large number of papers by Mason and coworkers were devoted to the extensions of Jeffery's work. From this early work came the realization that the properties of suspensions can be understood from a knowledge of the behavior of the suspended particles, a realization which led to the science of microrheology, a term coined first by Mason himself.

Mason's early work on papermaking became classical among scientists working in the field of pulp and paper. It led, among other things, to an understanding of the electrokinetic properties of pulp fibers (Mason 1950b; Goring & Mason 1950; Biefer & Mason 1954), to a widely used method of measuring the specific surface of fibers (Mason 1950a) and to a test of the bursting strength of paper (Faichney & Mason 1952). Most importantly, it led to an understanding of the turbulent diffusion in paper machines (Mason *et al.* 1954) and the flocculation and flow behavior of papermaking suspensions (Mason 1954; Robertson & Mason 1956; Forgacs *et al.* 1958). The publications arising from his early period show the characteristics for which Mason became famous in the field of microrheology: a thorough mathematical analysis together with careful and elegant experimental observations. For these achievements alone Mason deserved a place among the world's great scientists. However, he gained even more fame for his subsequent work.

This early work on papermaking was instrumental in shaping the future developments in Mason's career. He realized from the beginning that the basic principles underlying particle motion in papermaking suspensions could be equally applied to a large number of other fields. As examples we can mention: (i) the work on turbulent diffusion in paper machines, which contained the germs of the modern statistical theory of turbulence and (ii) the motion of pulp fibers, which is similar to the motion of many nonspherical particles in other systems, such as red blood cells in blood flow, clay particles in papermaking coatings and logs floating on rivers. As a result of the many basic insights gained from papermaking research, Mason turned his attention more and more to model systems which could be rigorously described by hydrodynamic theory and observed by cinephotography. Initially he looked at rigid rods (Travelyan & Mason 1951), flexible rods (Forgacs & Mason 1959), droplets (Bartok & Mason 1958; Rumscheidt & Mason 1961a, b) and, of course, spheres (Bartok & Mason 1959). The work on hard spheres is of particular importance since it can be considered the simplest system for which exact results can be obtained. At the same time the motion of a sphere in a shear flow and the fluid flow around it (Cox et al. 1968) shows qualitatively similar behavior as more complex systems and serves as the starting point for understanding hydrodynamic particle interactions. The work on spheres is an extension of the early work by Einstein (1906, 1911), who calculated the viscosity of a dilute suspension of spheres. Part of this calculation consisted of calculating the flow field around a sphere in a linear flow field. Curiously,

Mason makes no reference to Einstein's early work. Einstein, however, made no comments about the form of the flow field, such as the existence of close streamlines orbiting the sphere.

The discovery of closed streamlines around spheres subjected to a simple shear flow had wide-ranging consequences. It implied that small particles could orbit a large sphere in shear flow and, by extension of these ideas, it led to the realization that all kinds of particles could orbit other particles, due solely to hydrodynamic interactions. Mason was particularly interested in the simple case of the interaction of two identical spheres in shear flow. His first paper on this topic goes back as far as 1952 (Manley & Mason 1952), while his last paper on this topic appeared in 1981 (Adler *et al.* 1981). Various interactions (besides hydrodynamic ones) are included in the analysis, such as the interactions between colloidal electrically charged spheres (van de Ven & Mason 1976, 1977), effects of external electric fields (Arp & Mason 1977a, b), effects of polyelectrolytes (Takamura *et al.* 1979), interactions of spheres connected by flexible rods (Takamura *et al.* 1981), which serve as a model for polymer-bridged doublets, and effects of Brownian motion (van de Ven *et al.* 1981). He also studied the interaction between two drops and their coalescence or (partial) engulfment (Torza & Mason 1970; Mason 1971). Not only were particle interactions studied in dilute systems, but in concentrated systems as well (Karnis *et al.* 1966).

Besides extending the theory of a sphere in a simple shear flow to two-sphere systems, many of Mason's publications deal with extensions of the theory to particles of different shapes, following the early work of Jeffery on ellipsoidal particles. We have already mentioned the work on rigid and flexible rods and drops. Other particles considered were threads and microcapsules. Many papers deal with macroscopic properties of suspensions consisting of such particles, most of which deal with suspensions of noninteracting spheroids. The properties studied were viscosity (Okagawa *et al.* 1974), rheo-optical properties (light scattering, turbidity) (Okagawa & Mason 1977; Cerda *et al.* 1981) and dielectric properties (Okagawa *et al.* 1978), most of them in both the presence and absence of electric fields. In several papers particle interactions were included (Arp & Mason 1977c; Ivanov *et al.* 1982).

The motion of closed streamlines also turned out to be a useful concept in explaining the phenomenon of aggregate or floc break-up. When closed orbits exist, it is difficult to break up aggregates. Even if a particle detaches from a large aggregate, it will not be convected away but stay associated with the floc, orbiting it on a closed trajectory. In the absence of closed streamlines, such as in extensional flow, break-up is much more effective (Kao & Mason 1975; Powell & Mason 1982).

In addition to the work in microrheology, Mason also became interested in the field of wetting and spreading of liquids on solid surfaces. Here again he was spurred by the relevance of this field to applications in papermaking (printing, sizing, absorbency etc). In this area he also showed that complicated phenomena (such as the wetting of a sheet of paper) can be broken down in simpler phenomena which can be studied in detail. Examples are the wetting of various model rough surfaces by several liquids (Huh *et al.* 1975; Huh & Mason 1977), the effect of sharp edges (Oliver *et al.* 1977) and contact angle hysteresis (Huh & Mason 1977; Bayramli *et al.* 1981). Again rigorous theories were developed which were tested with elegant wetting and spreading experiments, with liquids either advancing or receding on a horizontal disk (Mason 1978) or on a vertical probe (Okagawa & Mason 1978; Bayramli *et al.* 1981), a technique which Mason called "capillarography". He also took high-resolution pictures of advancing liquids in a scanning electron microscope, the first pictures of this kind ever produced by this technique (Oliver & Mason 1977).

Last but not least, we want to mention Mason's interest in biological systems, especially blood. Here, as well, he was able to explain several phenomena by studying model systems such as deformable drops in Poiseuille and pulsatile flow (Takano *et al.* 1968), which serve as models for erythrocytes and the irregular motion of single particles in concentrated dispersions, giving rise to apparent Brownian motion and dispersion (Goldsmith & Mason 1967).

In all these diverse fields of interest, papermaking, microrheology, wetting and spreading and biorheology, Mason produced a substantial number of films of dazzling beauty, combining scientific observations with esthetic value. His movies show fascinating motions of all kinds of particles, rotating, colliding, deforming, rupturing, embracing or engulfing. Since Mason rarely gave a seminar without showing a movie, it is no wonder that he was an extremely popular lecturer. Over his career, Mason supervised some 60 students and 20 postdoctoral fellows with whom he published about 270 papers, most of them after joining Paprican in 1946. Prior to this he obtained his B.Eng. (in 1936) and Ph.D. (in 1939) in chemistry at McGill University under the supervision of Professor Otto Maass. During the war he was a research engineer in the Department of National Defense. While at Paprican he held an honorary cross-appointment in the Department of Chemistry at McGill University, where he was appointed a full professor in 1966, Otto Maass Professor in 1979 and Emeritus Professor in 1985.

Among the many awards he received were the Kendall Award in Colloid Chemistry (1967), the Anselme Payen Award in Cellulose Chemistry (1969), the Bingham Medal in Rheology (1969), the Chemical Institute of Canada Medal (1973), the Dunlop Award (1975), the Howard N. Potts Medal (1980) and, one year before his death, the highest scientific distinction awarded in the Province of Quebec, the Prix Marie-Victorin (1986). When he retired in 1979, a festschrift in his honor was issued [J. Colloid Interface Sci. 71 (1979)], which included an appreciation of his work (Goldsmith & Goring 1979). More recently, an issue of Biorheology was also dedicated to him [Biorheology 26 (1989)], which included an overview of his accomplishments in this field (Goldsmith 1989).

In October 1989 the Society of Rheology held their annual meeting in Montreal, together with the 1st Annual Meeting of the Canadian Rheology Group. Since Mason spent almost all his life in Montreal, it was appropriate that the section on "Multiphase Systems and Biopolymers" be dedicated to him. Several papers in the session on "Microrheology" organized by Professor H. Brenner and myself appear in this issue. A well-deserved tribute to a man who devoted his lifetime to the science of microrheology. His death on 21 April 1987 is still fresh in our memories and his passing evoked a great sense of loss to his many friends, colleagues and students. We hope that his exemplary achievements will continue to be a source of inspiration for many.

## REFERENCES

- ADLER, P. M., TAKAMURA, K., GOLDSMITH, H. L. & MASON, S. G. 1981 Particle motions in sheared suspensions. XXX. Rotations of rigid and flexible dumbbells (theoretical). J. Colloid Interface Sci. 83, 516-530.
- ARP, P. A. & MASON, S. G. 1977a Particle behavior in shear and electric fields. VIII. Interactions of pairs of conducting spheres (theoretical). Colloid Polym. Sci. 225, 556–584.
- ARP, P. A. & MASON, S. G. 1977b Particle behavior in shear and electric fields. IX. Interactions of pairs of conducting spheres (experimental). Colloid Polym. Sci. 225, 980–993.
- ARP, P. A. & MASON, S. G. 1977c Interactions between two rods in shear flow. J. Colloid Interface Sci. 59, 378-380.
- BARTOK, W. & MASON, S. G. 1958 Particle motions in sheared suspensions. VII. Internal circulation in fluid droplets (theoretical). J. Colloid Sci. 13, 293–307.
- BARTOK, W. & MASON, S. G. 1959 Particle motions in sheared suspensions. VIII. Singlets and doublets of fluid spheres. J. Colloid Sci. 14, 13-26.
- BAYRAMLI, E., VAN DE VEN, T. G. M. & MASON, S. G. 1981 Tensiometric studies on wetting. I. Some effects of surface roughness (theoretical). Can. J. Chem. 59, 1954–1961.
- BIEFER, G. J. & MASON, S. G. 1954 Electroosmosis and streaming in natural and synthetic fibers. J. Colloid Sci. 9, 20–35.
- CERDA, C. M., FOISTER, R. T. & MASON, S. G. 1981 Rheo-optical transients in sheared suspensions. II. Polydisperse spheroids. J. chem. Soc. Faraday Trans. 1, 77, 2949–2967.
- Cox, R. G., ZIA, I. Y. Z. & MASON, S. G. 1968 Particle motions in sheared suspensions. XXV. Streamlines around cylinders and spheres. J. Colloid Interface Sci. 27, 7-18.
- EINSTEIN, A. 1906/1911 Investigations on the Theory of Brownian Movement. Dutton, New York (English translation 1926).
- FAICHNEY, L. M. & MASON, S. G. 1952 The bursting strength of paper. Recommended test method. *Pulp Pap. Can.* 53, 123–126.
- FORGACS, O. L. & MASON, S. G. 1959 Particle motion in sheared suspensions. X. Orbits of flexible thread-like particles. J. Colloid Sci. 14, 473–491.
- FORGACS, O. L., ROBERTSON, A. A. & MASON, S. G. 1958 The hydrodynamic behavior of papermaking fibres. *Pulp Pap. Can.* 59, 117-128.

- GOLDSMITH, H. L. 1989 Stanley Mason: his contributions to the science of biorheology. *Biorheology* **26**, 99–105.
- GOLDSMITH, H. L. & GORING, D. A. I. 1979 Stanley G. Mason. An appreciation. J. Colloid Interface Sci. 71, 1–7.
- GOLDSMITH, H. L. & MASON, S. G. 1967 In *Rheology: Theory and Applications*, Vol. 4 (Edited by EIRICH, F. R.), Chap. 2, pp. 85–250. Academic Press, New York.
- GORING, D. A. I. & MASON, S. G. 1950 Electrokinetic properties of cellulose fibers. I, II and III. Can. J. Res. 28B, 307-322; 28B, 323-338; 28B, 339-344.
- HUH, C. & MASON, S. G. 1977 Effects of surface roughness on wetting (theoretical). J. Colloid Interface Sci. 60, 11-38.
- HUH, C., INOUE, M. & MASON, S. G. 1975 Uni-directional spreading of one liquid on the surface of another. Can. J. chem. Engng 53, 367-371.
- IVANOV, Y., VAN DE VEN, T. G. M. & MASON, S. G. 1982 Damped oscillations in the viscosity of suspensions of rigid rods. I. Monomodal suspensions. J. Rheol. 26, 213–230.
- JEFFERY, G. B. 1922 The motion of ellipsoidal particles immersed in a viscous fluid. Proc. R. Soc. Lond. A102, 161-179.
- KAO, S. V. & MASON, S. G. 1975 Dispersion of particles by shear. Nature 253, 619-621.
- KARNIS, A., GOLDSMITH, H. L. & MASON, S. G. 1966 The kinetics of flowing dispersions. I. Concentrated suspensions of rigid particles. J. Colloid Interface Sci. 22, 531-553.
- MANLEY, R. ST. J. & MASON, S. G. 1952 Particle motions in sheared suspensions. II. Collisions of uniform spheres. J. Colloid Sci. 7, 354–369.
- MASON, S. G. 1950a The specific surface of fibers. Its measurement and application. Tappi 33, 403-409.
- MASON, S. G. 1950b The electrokinetic properties of cellulose fibers. Tappi 33, 413-417.
- MASON, S. G. 1954 Fiber motions and flocculation. Tappi 37, 494-501.
- MASON, S. G. 1971 Film review "Coalescence of liquid drops". J. Colloid Interface Sci. 35, 517.
- MASON, S. G. 1978 Wetting and spreading. Some effects of surface roughness. In Wetting, Spreading and Adhesion (Edited by PADDAY, J. F.), p. 321. Academic Press, London.
- MASON, S. G., ROBERTSON, A. A., ALLEN, G. A. & WALKER, C. W. E. 1954 Turbulent diffusion in fourdrinier machines. *Pulp Pap. Can.* 55, 97–108.
- OKAGAWA, A. & MASON, S. G. 1977 Kinetics of flowing dispersions. X. Oscillations in optical properties of streaming suspensions of spheroids. Can. J. Chem. 55, 4243-4256.
- OKAGAWA, A. & MASON, S. G. 1978 Capillarography: a new surface probe. In Proc. 6th Fundamental Research Symp., Oxford, 1977, pp. 581-586. Tech. Section BPBIF, London.
- OKAGAWA, A., COX, R. G. & MASON, S. G. 1974 Particle behavior in shear and electric fields, VI. The microrheology of rigid spheroids. J. Colloid Interface Sci. 47, 536-567.
- OKAGAWA, A., COX, R. G. & MASON, S. G. 1978 Kinetics of flowing dispersions. XI. Dielectric constants of streaming suspensions of spheroids. J. chem. Soc. Faraday Trans. 174, 1242-1253.
- OLIVER, J. F. & MASON, S. G. 1977 Microspreading studies on rough surfaces by scanning electron microscopy. J. Colloid Interface Sci. 60, 480-487.
- OLIVER, J. F., HUH, C. & MASON, S. G. 1977 Resistance to spreading of liquids by sharp edges. J. Colloid Interface Sci. 59, 568-581.
- POWELL, R. L. & MASON, S. G. 1982 Dispersion by laminar flow. AIChE Jl 28, 286-293.
- ROBERTSON, A. A. & MASON, S. G. 1956 Pipe flow and fibre flocculation. *Pulp Pap. Can.* 57, 121–123.
- RUMSCHEIDT, F. D. & MASON, S. G. 1961a Particle motions in sheared suspensions. XI. Internal circulation in fluid droplets (experimental). J. Colloid Sci. 16, 210-237.
- RUMSCHEIDT, F. D. & MASON, S. G. 1961b Particle motions in sheared suspensions. XII. Deformation and burst of fluid drops in shear and hyperbolic flow. J. Colloid Sci. 16, 238-261.
- TAKAMURA, K., GOLDSMITH, H. L. & MASON, S. G. 1979 The microrheology of colloidal dispersions. IX. Effects of simple and polyelectrolytes on rotation of doublets of spheres. J. Colloid Interface Sci. 72, 385–400.
- TAKAMURA, K., ADLER, P. M., GOLDSMITH, H. L. & MASON, S. G. 1981 Particle motions in sheared suspensions. XXXI. Rotations of rigid and flexible dumbbells (experimental). J. Colloid Interface Sci. 83, 516-530.

- TAKANO, M., GOLDSMITH, H. L. & MASON, S. G. 1968 The flow of suspensions through tubes. IX. Particle interactions in pulsatile flow. J. Colloid Interface Sci. 27, 268–281.
- TORZA, S. & MASON, S. G. 1970 Three-phase interactions in shear and electric fields. J. Colloid Interface Sci. 33, 68-83.
- TRAVELYAN, B. J. & MASON, S. G. 1951 Particle motions in sheared suspensions. I. Rotations. J. Colloid Sci. 6, 354–367.
- VAN DE VEN, T. G. M. & MASON, S. G. 1976 The microrheology of colloidal dispersions. IV. Pairs of interacting spheres in shear flow. J. Colloid Interface Sci. 57, 505-516.
- VAN DE VEN, T. G. M. & MASON, S. G. 1977 The microrheology of colloidal dispersions. VII. Orthokinetic doublet formation of spheres. *Colloid Polym. Sci.* 255, 468–479.
- VAN DE VEN, T. G. M., TAKAMURA, K. & MASON, S. G. 1981 The microrheology of colloidal dispersions. X. Rotations of spheroids and high Péclet numbers. J. Colloid Interface Sci. 82, 373–383.