MACROSTATISTICAL HYDRODYNAMICS

Progress Report for the Period September 15, 1992-September 14, 1993

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MACROSTATISTICAL HYDRODYNAMICS

Summary

This research aims to correlate the macroscopic rheological behavior of suspensions with their statistical microstructure. This fundamental knowledge will benefit a host of technologies, including geothermal energy production, petroleum production and refining, and synfuels processing. The approach involves a novel combination of experiments, numerics, and theory. Experiments primarily involve tracking small balls as they fall slowly through otherwise quiescent suspensions of neutrally buoyant particles. Detailed trajectories of the balls, obtained either with new experimental techniques or by numerical simulation, are statistically interpreted in terms of the mean settling velocity and the dispersion about the mean. Determining the mean settling velocity of balls that are small relative to the suspended particles is a means of measuring the macroscopic zero-shear-rate viscosity without significantly disturbing the original microstructure; therefore, falling-ball rheometry is a powerful tool for use in studying the effects of microstructure on the macroscopic properties of suspensions. The dispersion about the mean yields information about the particle interactions. To date, the mean and dispersivity of a falling sphere's velocity has been determined as a function of the tracer sphere size and the suspended particle size, shape, and concentration. Currently, the pressure drop caused by the falling ball is being measured also. This will provide a much needed benchmark problem for numerical studies, as well as provide another measure of the macroscopic response of a suspension as a function of its microstructure. Also begun recently are two studies of boundary effects in two-phase fluids: the determination of the torque on a small ball spinning in a suspension (a sensitive measure of apparent slip at the boundary) and the determination of the velocity of a small ball rolling down the wall of a container holding a suspension. Numerical studies are also continuing, in which the three-dimensional dynamics of many interacting suspended particles are calculated using the boundary element method.

PROGRESS REPORT

Analysis of the collision-induced dispersion coefficient

In this period, a generalized Newtonian, one-dimensional, constitutive model and a "diffusion" equation for the particle concentration (Phillips, Armstrong, Brown, Graham, and Abbott, 1991) was tested against our data and shown to predict particle concentration and velocity fields in concentrated, monomodal suspensions quite well. In this model, the diffusion equation accounts for shear-induced particle migration through particle flux expressions derived by scaling arguments. The viscosity of the suspension is modeled as an increasing function of the volume fraction of particles. Particle concentrations for neutrally buoyant, monomodal suspensions of spheres undergoing flow between rotating concentric cylinders were measured using nuclear magnetic resonance imaging (Abbott, Tetlow, Graham, Altobelli, Fukushima, Mondy, and Stephens, 1991). The predictions agree very well with the measurements, thereby verifying the scaling analysis and the basic mechanics incorporated into the constitutive description.

The above model of the collision-induced, phenomenological, dispersion coefficient governing the shear-induced lateral migration of suspended spheres furnishes a value for the dispersion coefficient of about 2.4ϕ . For a solids volume fraction of $\phi=0.50$, this predicts that the dimensionless Fickian dispersion coefficient for such shear flows will be about unity. This is the same order of magnitude as the dimensionless dispersivity seen in the falling ball experiments described earlier, in which a ball (of the same size as the suspended spheres) settles through a quiescent suspension. This suggests that the same basic physics governs both phenomena.

Additional dispersivity data was taken this period in quiescent suspensions, concentrating on the detailed path taken by balls of various sizes falling through suspensions with a suspended-sphere volume fraction of 0.15. Several sizes of suspended spheres were used in order to vary the ratio of the falling-ball size to the suspended sphere size from 1 to 21. The purpose of this study was to elucidate the functional dependence of the dispersivity coefficient on the sizes of both the falling ball and suspended spheres. Results were compared to theory by Davis and Hill (1992) and to scaling arguments developed in conjunction with the model discussed above. We found that, in moderately concentrated suspensions (solid volume fraction of 0.15), the scaling of the vertical dispersivity with the relative sizes of the settling sphere and suspended spheres agrees with the dilute theory calculations by Davis and Hill. However, as the concentration increases the

dispersivity depends less and less on the relative sizes of the settling sphere and the suspended spheres, becoming nearly independent at a solids fraction of 0.50. In suspensions of rods, the dispersivity appears to also be invariant to the size of the settling sphere. The dispersivity for a single size settling sphere increases as the concentration of suspended particles increases. In suspensions of spheres or rods, the dispersivity increases linearly with concentration.

New experimental techniques

We proposed to concentrate on extending the technique of falling ball rheometry to more complex suspensions. To this end we developed a non-Newtonian liquid which can either match the index of refraction or density of our PMMA particles, thereby creating the capability to study suspensions with a non-Newtonian suspending fluid. We also perfected methods to align particles with aspect ratio greater than 1.0 (rods), thus preparing to study the dispersion in suspensions with transversely isotropic properties.

We also initiated a study of the appropriate slip/stick boundary condition for materials with microstructure through laboratory examination of a technique similar to falling-ball rheometry: rolling-ball viscometry. In rolling ball viscometry, a dense ball is allowed to roll/slide down an inclined surface (in this case, the inside of a cylinder containing suspension), and its rate of travel is compared to that in a fluid of known viscosity. In a suspension, we can ratio the time it takes for the ball to travel a known distance in the suspension to that in the suspending liquid alone and, from this ratio, estimate the apparent relative viscosity. This procedure is similar to estimating the apparent viscosity with falling ball viscometry; however, the immediate region of the suspension seen by the moving ball is not uniform but has structure determined by the proximity of the bounding wall.

Preliminary rolling-ball data was taken for a suspension with volume fraction solids equal to 0.30. Three sizes of rolling balls were used such that the ratio of the size of the rolling ball to the size of the suspended spheres ranged from 0.75 to 6.0. As in falling ball rheometry, the instantaneous velocity of the moving ball is not steady; however, a statistical analysis reveals that the *average* velocity of the ball, measured over a distance usually equivalent to 100 to 1000 suspended particle diameters, is reproducible. The *average* velocities of the three sizes of balls then translate into three measured relative viscosities. These viscosities all agreed within the 95% confidence limits of the data, indicating that the measured viscosity was independent of the size of the falling ball. Furthermore, the viscosity measured with rolling ball viscometry was statistically indistinguishable from

that measured with falling ball rheometry. This is surprising in light of the fact that the suspended particles cannot be distributed near the wall in the same manner that they are distributed in the bulk of the suspension, simply due to geometric constraints. Further data may allow differences now obscured by the statistical nature of the system to be distinguished. It appears that rolling ball viscometry will allow more insight into the behavior of concentrated suspensions and is a technique worth further study.

This period, we also created an apparatus necessary to study flowing suspensions. A homogeneous flow apparatus (HFA) (Graham, and Bird, 1984) was built, consisting of two moving, flat, parallel belts and a containing vessel made from polymethyl methacrylate and aluminum bracing plates. A motor drives the belts in a smooth, steady motion. The belts are coupled with a gear drive so that their motion is parallel but opposite in the center section of the HFA.

Boundary element modeling of suspensions

The ability to perform dynamic simulations of a ball falling through suspensions of dozens of particles was enhanced by decreasing the computational effort required at each time step. This was accomplished by using a variable-order-Gauss quadrature routine. The number of quadrature points is *locally* increased where needed to obtain an accurate solution, but in most elements the number of quadrature points is reduced without affecting the accuracy of the solution. The resulting number of equations is reduced from our original method, and the accuracy is enhanced. Furthermore, we obtained a six-fold speed-up in obtaining most solutions.

Continuation of dispersion experiments

In this year, we continued the analytical efforts to develop a constitutive description of flowing, concentrated suspensions, accounting for shear-induced "diffusion" (migration) of the suspended particles. We extended the description to include the effects of a shear-thinning suspending fluid. Nuclear magnetic resonance imaging (NMRI) data was taken on suspensions subjected to flow between rotating concentric cylinders. Results showed that the concentration and velocity profiles are very dependent on the rheology of the suspending liquid. The constitutive description captured this behavior and showed excellent agreement with the NMRI data for this one-dimensional flow. Our hopes are to develop the capability to describe two dimensional flows, and to gather the appropriate data with which to test the model.

We also initiated the study of flow-induced migration of nonspherical particles. NMRI provided concentration profiles of the test suspensions of fibers. Fibers of length 0.32 cm and diameters ranging from 0.015 cm to 0.13 cm were used. Somewhat surprisingly, there was no effect of particle shape on the steady-state concentration profiles produced in the flow between concentric rotating cylinders. Fibers of aspect ratio up to 20 migrated to the same final concentration distribution as did spheres. We plan on extending the constitutive description to include fibers as well as spherical particles.

Also this year, we will complete the falling-ball dispersion studies in suspensions of uniform spheres. We have developed a method to analyze short-time (correlated) dispersion data from the falling-ball experiments based on the expression derived for Brownian tracer particles by Uhlenbeck and Ornstein (1930). Their expression accounted for particle inertia. We have found that true inertial effects do not account for the short-time behavior of a ball falling through our concentrated suspensions. By tracking balls of the same size but different densities in a concentrated suspension, we found that the dispersions were statistically invariant if the time were rescaled with the relevant average settling velocity. This suggested that the observed, nonlinear, short-time variances in position were caused by the time needed for the settling ball to change its local environment. Because a large settling ball drags the nearby suspended spheres along with it, this time scale could be considerably longer than the time it took the sphere to settle one suspended-sphere diameter. The new model appears to describe well the short-time quadratic behavior and the long-time linear behavior of the variances, as well as the transition between. This allows much more of our data to yield unambiguous dispersion coefficients.

Plans for the year beginning September 15, 1993

The pressure drop across a ball falling in a quiescent suspension

This year we will extend the falling ball studies to measure the pressure drop across the ball. In a single-phase, Newtonian fluid this pressure is constant, independent of viscosity (at low Reynolds number), and can be described analytically (Pliskin and Brenner, 1963)*. In a suspension, the pressure drop may be dependent on the microstructure and, like the viscosity, may vary due to the discrete, noncontinuum nature of the material.

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* see also, Feldman and Brenner, 1968.

Preliminary measurements indicate that the pressure drop across a ball falling in a suspensions of 30 volume percent solid spheres is identical to that predicted in a single-phase Newtonian liquid. Like the viscosity measurements in a moderately concentrated suspension, the pressure drop behavior is independent of the relative sizes of the falling ball and suspended spheres (however, the absolute measure of pressure drop in both single- and two-phase fluids is directly proportional to the weight of the falling ball).

Boundary element simulations

Significant progress was made this year in enhancing numerical modeling capabilities. The boundary element method (BEM) was reformulated to implement on a computer with massively parallel architecture. Initial tests on four processors benchmarked and verified the new formulation. We now expect to be able to significantly increase the size of tractable problems (the number of particles modeled).

Other BEM simulations of fully three-dimensional trajectories of rods in linear and nonhomogeneous shear flows were undertaken to support the rod migration studies discussed above. We showed that there is little effect of walls in these type flows, even when the gap between walls is only slightly larger than the length of the suspended particles.

In this year, we hope to improve the speed of the computations, as well as increase the number of suspended particles possible to simulate, by continuing to explore the possibilities of solving these multi-particle problems on computers using parallel architectures. We also plan to complete the pressure drop measurements in falling-ball experiments, examining the effects of relative ball size to particle size in suspensions of various solid concentrations. We also envision the need to use newly created tools such as a spinning-ball rheometer to further explore the slip/stick issue. The appropriate equipment will be completed, calibrated, and initial studies of suspensions of uniform spheres will be performed. Finally, we expect to complete NMRI migration studies on non-Newtonian liquids, fibers of various sizes, and multidimensional flows (such as flow in journal bearings and through contractions).

We will also use the HFA to measure the fluctuations in the velocities of neutrally buoyant particles in concentrated suspensions subjected to simple shear flow. These measurements are related to our previous work to determine the dispersivity of falling balls, except here we focus on the self-diffusion of the particles. These measurements will provide essential information to allow proposed microstructural linkages to continuum models to be checked. In other experiments, we will revisit the rolling ball rheometer to study the stick/slip phenomena and microstructure near walls.

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Appendix A.

PUBLICATIONS ACKNOWLEDGING DOE SUPPORT (BY THE "MACROSTATISTICAL HYDRODYNAMICS" GROUP) (September 15, 1992-March 31, 1993)

I. Publications by H. Brenner, *et al.*

Journal Articles Published

Shapiro, M., and Brenner, H., Comments on "The reaction independence of mixing coefficients in fixed-bed reactors," *AIChE J.*, **38**, 1675-1676 (1992).

Brenner, H., and Shapiro, M., Reply to comments on "The reaction independence of mixing coefficients in fixed-bed reactors," *AIChE J.*, **38**, 1678-1679 (1992).

Haber, S., and Brenner, H., "Effect of entrained colloidal particles in enhancing the transport of adsorbable chemical contaminants," *J. Colloid Interface Sci.* **155**, 226-246 (1993).

Iosilevskii, G., and Brenner, H., "Taylor dispersion in systems containing a continuous distribution of reactive species," *Int. J. Non-Linear Mech.*, **28**, 69-86 (1993).

Edwards, D.A., Shapiro, M., and Brenner, H., "Flow, dispersion and reaction in two-dimensional model porous media," *Phys. Fluids A* **5**, 837-848 (1993).

Journal Articles Accepted for Publication

Iosilevskii, G., Mendoza-Blanco, A.E., and Brenner, H., "The slow axisymmetric rotation of a sphere in a transversely-isotropic fluid," *Quart. J. Mech. Appl. Math.* (in press, 1993).

Frankel, I., and Brenner, H., "Taylor dispersion of orientable Brownian particles in unbounded homogeneous shear flows," *J. Fluid Mech.* (in press, 1993).

Iosilevskii, G., Brenner, H., Moore, C.M.V., and Cooney, C.L., "Mass transport and chemical reaction in Taylor-vortex flows with entrained catalytic particles: Applications to a novel class of immobilized enzyme biochemical reactors," *Trans. Roy. Soc. Lond.* (in press, 1993).

Iosilevskii, G., and Brenner, H., "Stokes law for a sphere in a transversely-isotropic fluid. The axisymmetric case," *Stability Appl. Anal. Continuous Media* (in press, 1993).

Lue, A., and Brenner, H., "Phase flow and statistical structure of Galton board systems," *Phys. Rev. E.* (in press, 1993).

Mavrovouniotis, G.M., and Brenner, H., "A micromechanical investigation of interfacial transport processes: I. Interfacial conservation equations, *Phil. Trans. Roy. Soc. (Lond.)* (in press, 1993).

Mavrovouniotis, G.M., Brenner, H., Edwards, D.A., and Ting, L., "A micromechanical investigation of interfacial transport processes: II. Interfacial constitutive equations," *Phil. Trans. Roy. Soc. (Lond.)* (in press, 1993).

Journal Articles Submitted for Publication

Batycky, R.P., Edwards, D.A., and Brenner, H., "Thermal Taylor dispersion in an insulated circular cylinder. Part I. Theory," *Int. J. Heat Mass Transfer* (submitted, 1992).

Batycky, R.P., Edwards, D.A., and Brenner, H., "Thermal Taylor dispersion in an insulated circular cylinder. Part II. Applications," *Int. J. Heat Mass Transfer* (submitted, 1992).

Batycky, R.P., Edwards, D.A., and Brenner, H., "Internal energy transport in adiabatic systems. Thermal Taylor dispersion phenomena" *Phys. Fluids A* (submitted, 1992).

Batycky, R.P., Edwards, D.A., and Brenner, H., "Thermal Taylor dispersion phenomena in nonadiabatic systems," *J. Fluid Mech.* (submitted, 1992)

Iosilevskii, G., and Brenner, H., "Taylor dispersion in discrete reactive mixtures," (resubmitted, 1993).

Book (in press)

Brenner, Howard and Edwards, David A., *Macrotransport Processes*, Butterworth-Heinemann, 714 pp.+xxiv, Boston, Massachusetts (1993).

Chapters in Books

Abbott, J.R., Brenner, H., Graham, A.L. and Mondy, L.A., "Techniques for analyzing the behavior of concentrated suspensions," in *Particulate Two-Phase Flow* (M.C. Roco, Ed.), Chapt. 1, pp. 1-32, Butterworth-Heinemann, Boston, MA (1993).

Conference Proceedings

Brenner, H., Altobelli, S.A., Graham, A.L., Abbott, J.R. and Mondy, L.A., "Hydrodynamic particle migration in small-amplitude, oscillatory, circular Couette flow: The limits of reversibility," in *Theoretical and Applied Rheology* (Proceedings of the XIth International Congress on Rheology, Brussels, Belgium, Aug. 17-21, 1992), Vol. 2, pp. 585-587 (P. Moldenaers and R. Keunings, Eds.), Elsevier, New York (1992).

Davis, A.M.J., Kezirian, M.T., and Brenner, H., "On the motion through a viscous fluid of a spherical particle touching a plane wall: Slip boundary conditions," *Third International Symposium on Current problems in Rheology, Biorheology and Biomechanics*, Moscow, Russia (September 1-5, 1992).

Batycky, R.P., and Brenner, H., "On the need for fictitious initial conditions in nonconservative effective medium theories, Proceedings of the *Second International Conference on Discrete Element Methods (DEM)*, pp. 131-143. (J.R. Williams and G.G.W. Mustoe, Eds.), IESL (Intelligent Engineering Systems Laboratory) Publications, Massachusetts Institute of Technology, Cambridge, MA (March 18-19, 1993).

Theses

Arthur Lue, "Phase Flow and Statistical Structure of Galton Board Systems," for the degree of Master of Science in Chemical Engineering, MIT (1992).

Richard P. Batycky, "Thermal Taylor Dispersion Phenomena," for the degree of Master of Science in Chemical Engineering, MIT (1992).

James R. Abbott, "Dispersive Processes in Concentrated Suspensions," for the degree of Doctor of Philosophy in Chemical Engineering, MIT (1992).

II. Publications by A.L. Graham and L.A. Mondy, *et al* (March 1992-March 1993)

*S. Dingman, M. S. Ingber, L. A. Mondy, J. R. Abbott, and H. Brenner, "Particle Tracking in Three-Dimensional Stokes Flow," *J. Rheology*, **36**(3), 413-440 (1992).

D. L. Tullock, N. Phan-Thein, and A. L. Graham, "Boundary Element Simulations of Spheres Settling in Circular, Square and Triangular Conduits," *Rheol. Acta*, **31**, 139-150 (1992).

L. A. Mondy, A. S. Geller, and D. J. Rader, "Application of Boundary Element Techniques to the Motion of Aerosol Particles," in *Boundary Element Technology VII*, C. A. Brebbia and M. S. Ingber, Eds, (Elsevier, New York, 1992) pp 105-116.

L. A. Mondy and M. S. Ingber, "Direct Second Kind Boundary Integral Formulation for Stokes Flow Problems," *Theoretical and Applied Rheology (Proc. of XIth International Congress on Rheology, Brussels, Belgium, August 17-21, 1992) Volume 1*, P. Moldenaers and R. Keunings, Eds, (Elsevier, New York, 1992) pp 201-203.

V. Ilic, D. Tullock, N. Phan-Thien, and A. L. Graham, "Sedimenting Spheres in Square and Circular Conduits: Experiments and Numerical Predictions," in *Theoretical and Applied Rheology (Proc. of XIth International Congress on Rheology, Brussels, Belgium, August 17-21, 1992) Volume 1*, P. Moldenaers and R. Keunings, Eds, (Elsevier, New York, 1992) p 304.

T. Papathanasiou, M. S. Ingber, L. A. Mondy, and A. L. Graham, "Boundary Element Modeling of Three-Dimensional Multiparticle Composites," in *Theoretical and Applied Rheology (Proc. of XIth International Congress on Rheology, Brussels, Belgium, August 17-21, 1992) Volume 1*, P. Moldenaers and R. Keunings, Eds, (Elsevier, New York, 1992) p 307.

*H. Brenner, S. A. Altobelli, A. L. Graham, J. R. Abbott, and L. A. Mondy, "Hydrodynamic Particle Migration in Small-Amplitude, Oscillatory, Circular Couette Flow: The Limits of Reversibility," in *Theoretical and Applied Rheology (Proc. of XIth International Congress on Rheology, Brussels, Belgium, August 17-21, 1992) Volume 2*, P. Moldenaers and R. Keunings, Eds, (Elsevier, New York, 1992) pp 585-587.

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* Also included in preceding H. Brenner/DOE Publication List.

R. Mor, M. Gottlieb, A. L. Graham, and L. A. Mondy, "Viscosity and the Distribution of Orientations in Confined Suspensions of Rod-Like Particles," in *Theoretical and Applied Rheology (Proc. of XIth International Congress on Rheology, Brussels, Belgium, August 17-21, 1992) Volume 2*, P. Moldenaers and R. Keunings, Eds, (Elsevier, New York, 1992) pp 622-624.

D. C. Guell, L. A. Mondy, and A. L. Graham, "Aligned Short-Fiber-Reinforced Composite Materials," in *Theoretical and Applied Rheology (Proc. of XIth International Congress on Rheology, Brussels, Belgium, August 17-21, 1992) Volume 2*, P. Moldenaers and R. Keunings, Eds, (Elsevier, New York, 1992) p 878.

M. S. Ingber and L. A. Mondy, "Direct Second Kind Boundary Integral Formulation for Stokes Flow Problems," *Computational Mech.*, 11(1), 11-27 (1993).

V. Ilic, D. L. Tullock, N. Phan-Thein, and A. L. Graham, "Translation and Rotation of Spheres Settling in Square and Circular Conduits: Experiments and Numerical Predictions," *Int. J. Multiphase Flow*, 18(6), 1061-1075 (1992).

D. M. Husband, L. A. Mondy, E. Ganani, and A. L. Graham, "Particle Migration in Concentrated Suspensions of Bimodal Spheres: Pipe, Couette, and Open Surface Flows," manuscript submitted *Rheol. Acta* (1993).

T. D. Papathanasiou, M. S. Ingber, L. A. Mondy, and A. L. Graham, "The Effective Elastic Modulus of Three Dimensional Multiparticle Composites," manuscript submitted *Journal of Composite Materials* (1993).

Appendix B

SEMINARS AND TECHNICAL PRESENTATIONS OF THE "MACROSTATISTICAL HYDRODYNAMICS" GROUP ACKNOWLEDGING DOE SUPPORT (September 15, 1992- September 14, 1993)

Presentations by H. Brenner *et al.*

1992

(with J.R. Abbott, L.A. Mondy and A.L. Graham) 1992 AIChE Annual Meeting, Miami Beach, Florida (November).

(with G.M. Mavrovouniotis and D.A. Edwards) 1992 AIChE Annual Meeting, Miami Beach, Florida (November).

(with R.P. Batycky and D.A. Edwards) 1992 AIChE Annual Meeting, Miami Beach, Florida (November).

Department of Mechanical Engineering, Fluid Mechanics and Heat Transfer Group, Massachusetts Institute of Technology, Cambridge, Massachusetts (November).

Department of Chemical Engineering, University of Calgary, Calgary, Canada (November).

¹Faculty of Engineering, University of Calgary, Calgary, Canada (November).

1993

Department of Chemical Engineering, West Virginia University, Morgantown, West Virginia (February).

(with R.P. Batycky), 2nd International Conference on Discrete Element Methods, Cambridge, Massachusetts (March).

(with A.L. Graham and L.A. Mondy), Eleventh Symposium on Energy Engineering Sciences, DOE, Argonne National Laboratory, Argonne, Illinois (May).

(with R.P. Batycky), Joint ASCE-ASME-ASCE Summer Applied Mechanics Meeting, Symposium on Recent Advances in Structured Continua, University of Virginia, Charlottesville, Virginia (June).

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¹Distinguished Visitor Lecture Series

Appendix C

HONORS, AWARDS AND SIGNIFICANT PROFESSIONAL ACTIVITIES BY H. BRENNER (September 15, 1992-March 31, 1993)

- Consulting Editor, Chairman of the Advisory Board, The Butterworth-Heinemann Series on Chemical Engineering.
- Member, Editorial Advisory Boards: *International Journal of Multiphase Flow*, Pergamon Press; *Transport in Porous Media*, Kluwer Academic Publishers.
- Member, International Advisory Committee, The Caribbean Congress of Fluid Dynamics.
- Distinguished Visiting Lecturer, University of Calgary.
- Chairman of the session on "Drops and Two-Phase Flow, 18th International Congress of Theoretical and Applied Mechanics, Haifa, Israel (1992).
- Member, Scientific Committee, International Symposium on Advances in Structured Continua II, Moscow, Russia (August 22-26, 1993).
- Member, Scientific Committee, IUTAM Symposium on "Liquid-Particle Interaction in Suspension Flow," Grenoble France (July 5-9, 1993).

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