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Containerless Measurements on Liquids at High Temperatures

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Abstract

The application of containerless techniques for measurements of the thermophysical properties of high temperature liquids is reviewed. Recent results obtained in the materials research laboratories at Intersonics are also presented.

Work to measure high temperature liquid properties is motivated by both the need for reliable property data for modeling of industrial processes involving molten materials and generation of data form basic modeling of materials behavior. The first two figures indicate the motivation for this work and present examples of variations in thermophysical property values from the literature. The variations may be attributed to changes in the specimen properties caused by chemical changes in the specimen or to and /or measurement errors.

The two methods used to achieve containerless conditions were aeroacoustic levitation and electromagnetic levitation [1]. Their qualities are presented. The accompanying slides show the layout of levitation equipment and present examples of levitated metallic and ceramic specimens.

Containerless techniques provide a high degree of control over specimen chemistry, nucleation and allow precise control of liquid composition to be achieved. Effects of minor additions can thus be measured in a systematic way. Operation in reduced gravity enables enhanced control of liquid motion which can allow measurement of liquid transport properties. Examples of nucleation control, the thermodynamics of oxide contamination removal, and control of the chromium content of liquid aluminum oxide by high temperature containerless processes are presented.

The feasibility of measuring temperature, emissivity, liquidus temperature, enthalpy, surface tension, density, viscosity, and thermal diffusivity are discussed in the final section of the paper. Temperature measurement is achieved by conventional pyrometry. Emissivity measurement presents an important issue in many processing applications, particularly when temperature-dependent properties are to be measured. The polarimetric technique is illustrated with an example of the relatively large change in emissivity which occurs during melting of niobium. The spectral emissivity of liquid inconel is shown to be almost constant over the temperature range from 1650 to 1950 K.

The proposed method for enthalpy measurements is drop calorimetry. This compliments

the contained techniques such as differential thermal analysis. The analysis of the effects of specimen emissivity are presented. Surface tension and density can be measured from the oscillation frequency of levitated drops [2] and by imaging the drop and determining its volume respectively.

Low gravity experiments to determine liquid viscosities and thermal diffusivities are described. A diagram of the apparatus used for ground-based experiment to evaluate a new method for measurements of thermal diffusivity are also shown.

In conclusion, containerless techniques combined with non-contact diagnostic techniques enable high temperature liquid property measurements under controlled conditions. Extensions of the techniques to low gravity will provide for more accurate measurements of transport properties.

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References:

1. J.K.R. Weber, S. Krishnan and P.C. Nordine, "The Use of Containerless Processing in Researching Reactive Materials" J. Metals, 43, 8-14 (1991).

2. B.J. Keene, "Surface Tension of Pure Metals", National Physical Laboratory Report DMM(A) 39, November, 1991, NPL, London, UK.

MOTIVATION

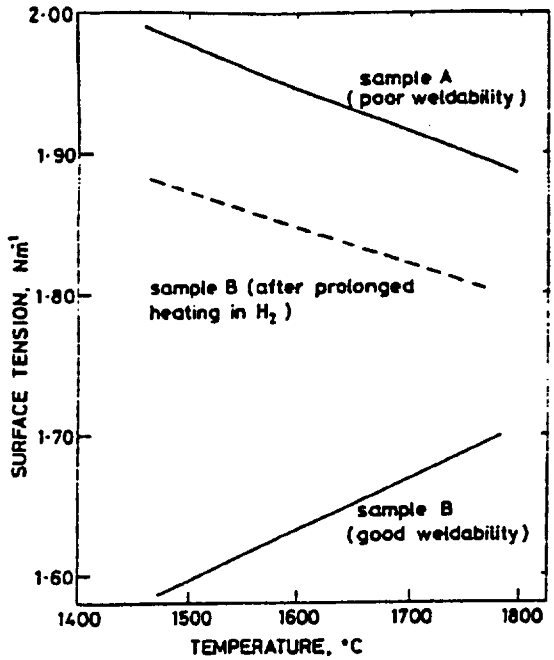
♦ **Many manufacturing methods involve HT liquid-phase processing.**

♦ **Improved understanding of processes = more cost effective manufacturing.**

♦ **Lack of HT data for liquids.**

♦ **Conflicting values in data.**

♦ **Containerless techniques have advanced greatly in the last decade and offer unique opportunities for HT measurement.**



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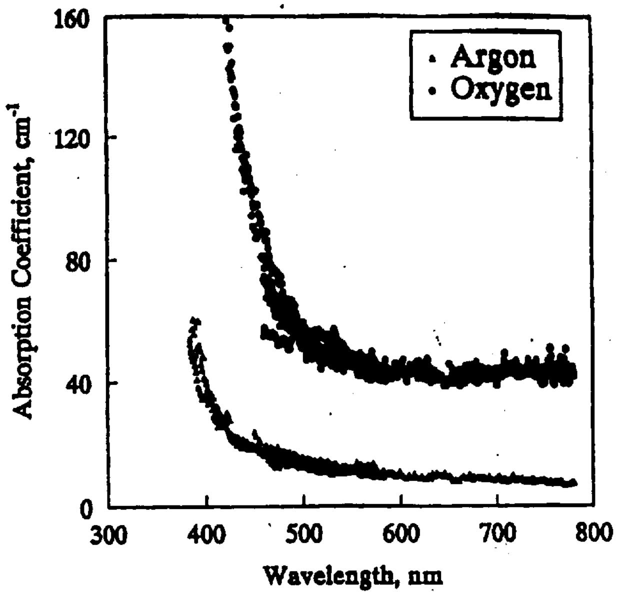
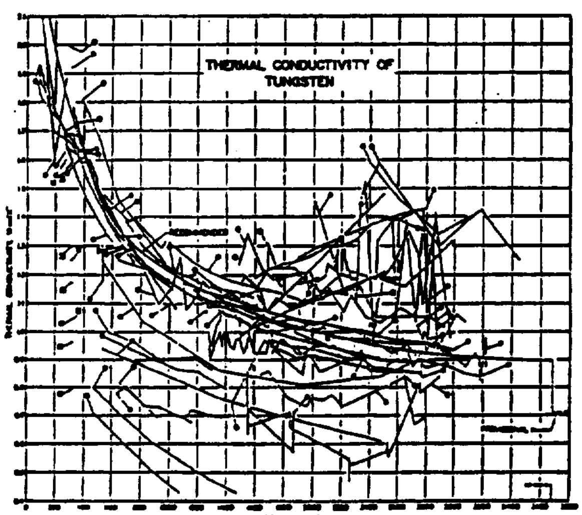


Research at Intersonics, supported by Air Force



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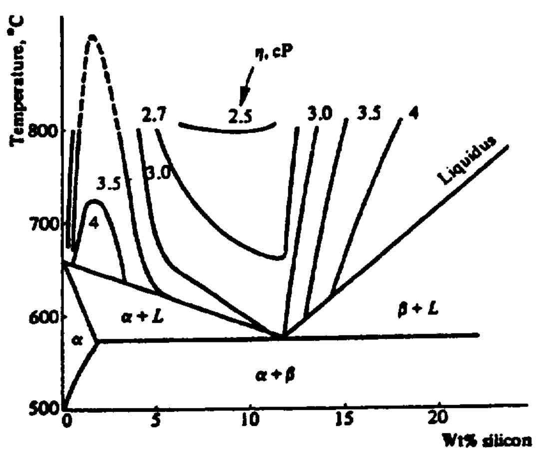

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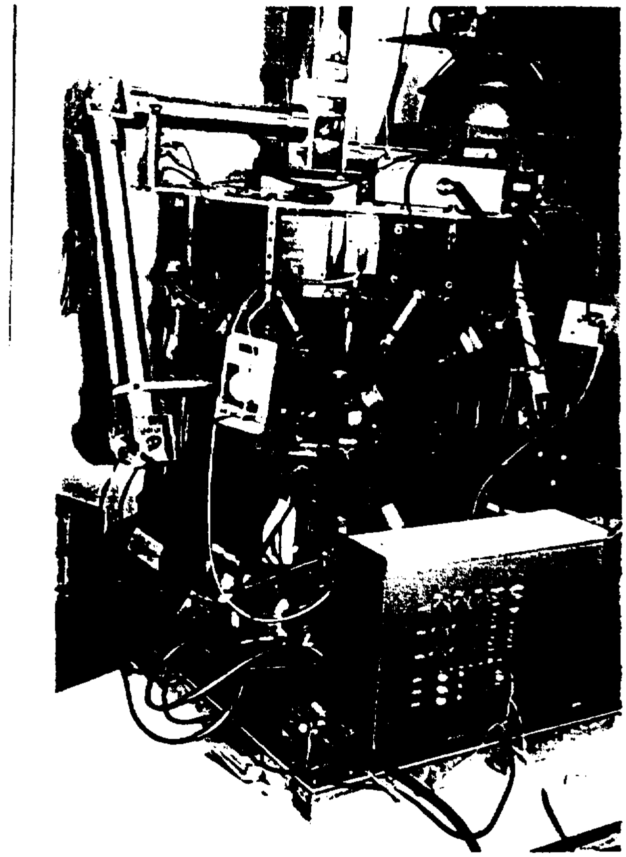
TABLE I

Comparison of Qualities of Electromagnetic and Aero-Acoustic Levitation

Electromagnetic Levitation	Aero-Acoustic Levitation
Metals and alloys	Metals, alloys, semiconductors and ceramics
Simultaneous positioning and heating	Separate positioning and heating
Operation in gas or vacuum	Requires gaseous atmosphere
Induction stirring	Aerodynamic stirring on ground
Field coils occlude specimen	Wide access to specimen



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QUALITIES OF CONTAINERLESS PROCESSING

- ♦ Eliminates contamination
- ♦ Eliminates nucleation
- ♦ Enables equilibration and purification

Low Gravity Processing

- ♦ Transport property measurements, D., α, η
- ♦ Control of segregation
- ♦ Control of gas evolution

Critical cooling rates, R_c for Calcia-Gallia-Silica Compositions

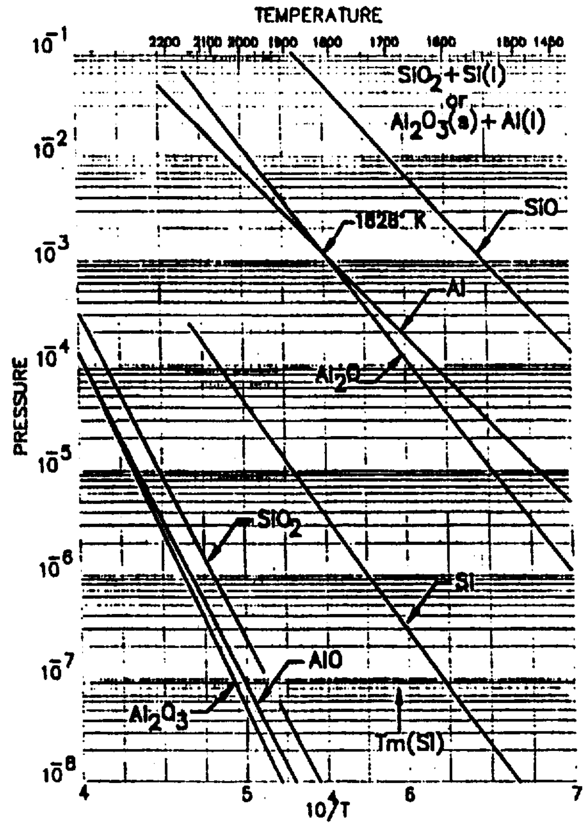
Specimen	Pendant Drop Expt. R_c, °C/s	Containerless Experiments R_c, °C/s	mass loss
CGS	300-350		
CGS-Pt1	450-550	< 10	1.8%
CGS-Pt5	> 700	40-80	0.7%
CG	> 700	50-60	0.6%

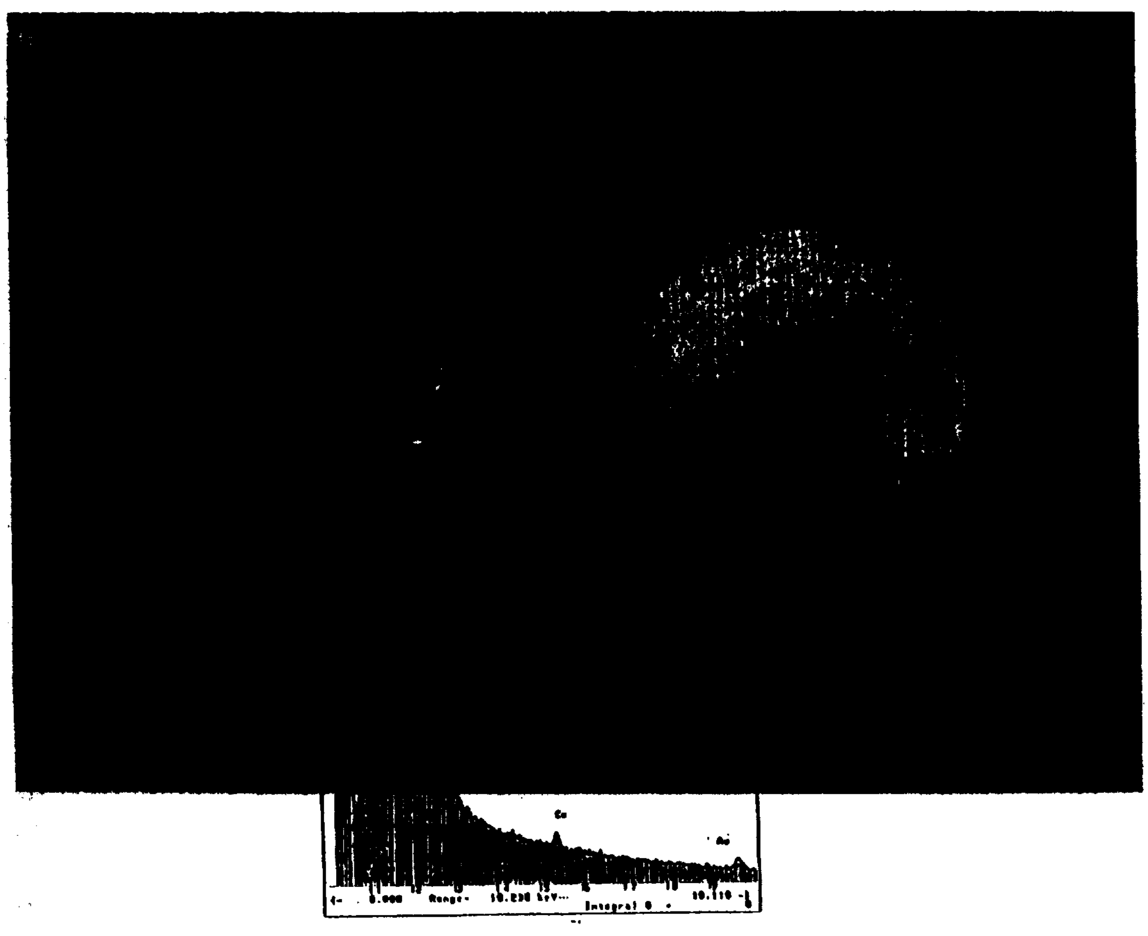
From: J.K.R. Weber, D.R. Merkley, C.D. Anderson, P.C. Nordine, C.S. Ray and D.E. Day, J. Am. Ceram. Soc., to be published.

$R = (O/M)vapor/(O/M)metal$

Where $R = \frac{\text{Total oxygen in gas}}{\text{Total metal in gas}}$

Brewer and Rosenblatt
Trans. Met. Soc. AIME 224, 1268 (1962).





Containerless Measurements and Feasibility Studies

1 Temperature - pyrometry

2 Emissivity - polarimetry

3 Liquidus temperature

4 Entahlpy - drop calorimetry

5 Surface tension - drop oscillation

6 Density/expansivity - drop imaging

7 Viscosity - drop oscillation

8 Thermal diffusivity/conductivity

$$\frac{1}{T} - \frac{1}{T_a} = \frac{\lambda \ln \epsilon_\lambda}{C_2}$$

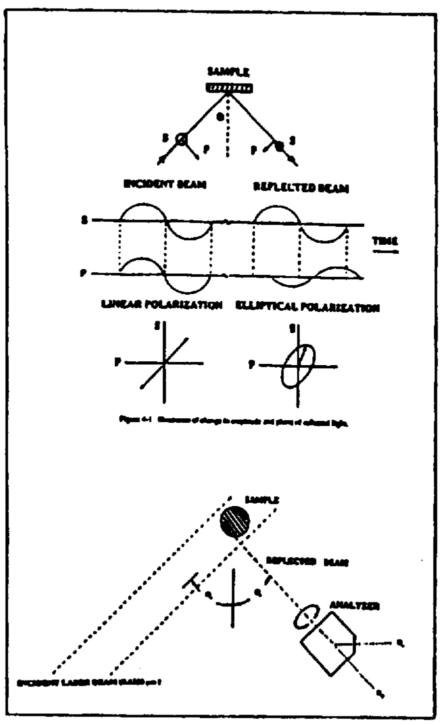
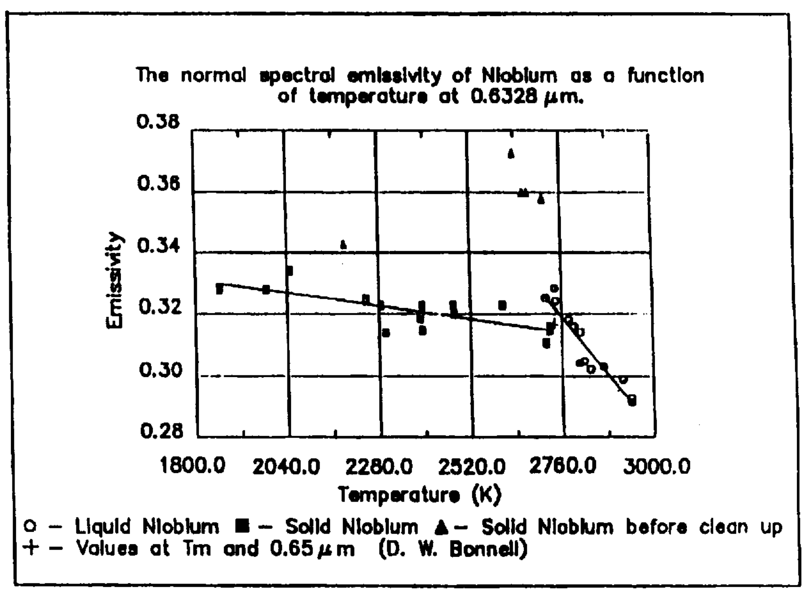
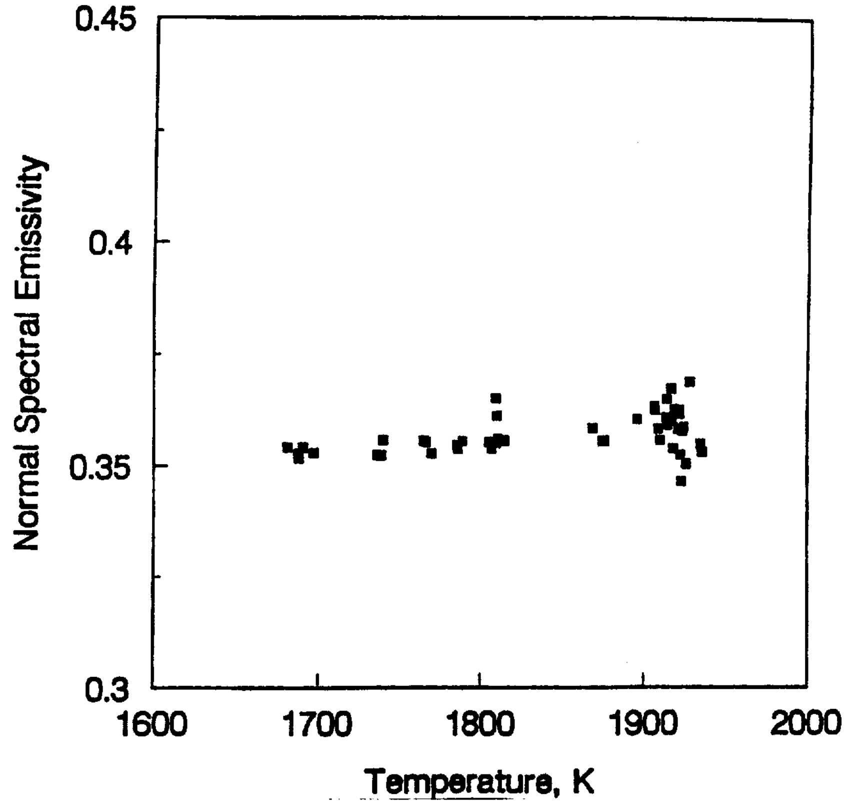


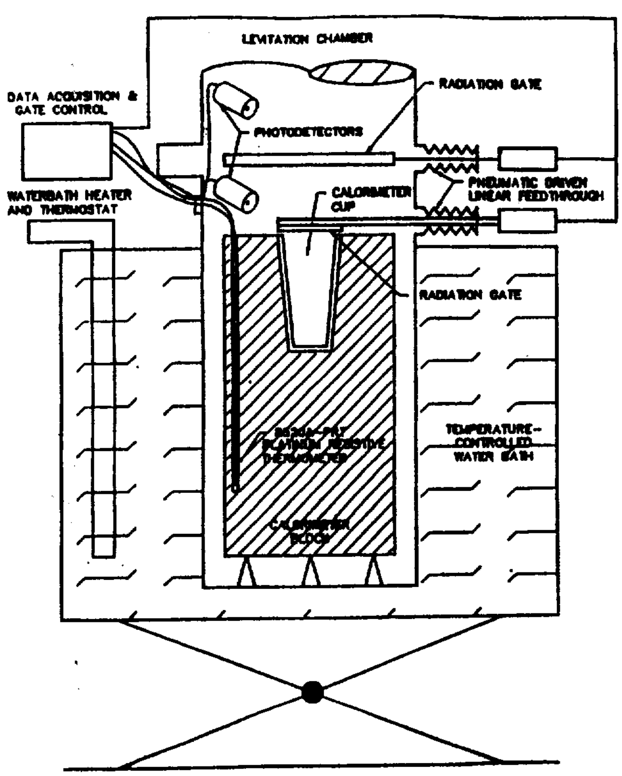
Illustration of Rotating Analyzer Ellipsometry



From: S. Krishnan, J.K.R. Weber, P.C. Nordine, R.A. Schiffman, R.H. Hauge and J.L. Margrave, High Temp. Sci., 30, 137 (1991).

Normal Spectral Emissivity of Inconel 718 at 633 nm





Calorimeter design. The instrument would be mounted beneath the electromagnetic levitation chamber with a thermostatically controlled water bath surrounding the calorimeter block.

13

Analysis of preliminary Niobium Results

Solid:

$$\epsilon_\lambda(T) = 0.3620 - 0.000017\ T$$

Liquid:

$$\epsilon_\lambda(T) = 0.7443 - 0.000154\ T$$

$$\frac{d\ln\epsilon}{dT} \text{ at } T_m = -4.78 \times 10^{-4}$$

$$\frac{C_p}{C_{pa}} = 0.83$$

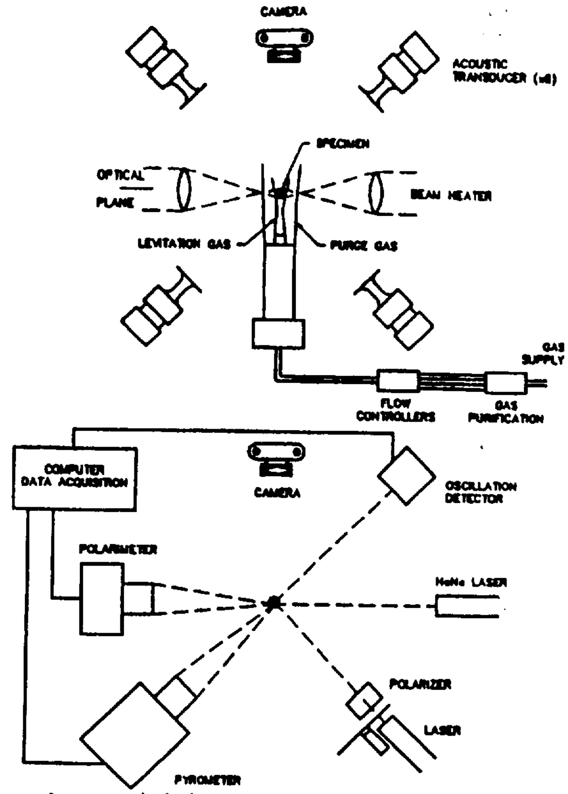
	C_p (J/mol.K) Bonnell	C_p (J/mol.K) Cezairliyan
Measurements	40.6	40.8
Corrected Values	33.7	33.8
Hultgren Est.	(33.5)	

SURFACE TENSION - RAYLEIGH EQUATION

$$\gamma = 3/8\ \pi\ m\ \omega_n^2$$

DENSITY & THERMAL EXPANSIVITY

$$\rho = m/v$$



Aero-acoustic levitator and ancillary equipment. Side view shows acoustic transducers, laser beam path and levitation nozzle. Lower view shows plan in the optical plane with relationships between instruments.

LOW GRAVITY EXPERIMENTS

MARANGONI CONVECTION

Marangoni number M_a is derived from the surface tension/temperature coefficient, $d\gamma/dT$; temperature gradient, ΔT; specimen radius, r; dynamic viscosity, ν; thermal diffusivity, α; and density, ρ.

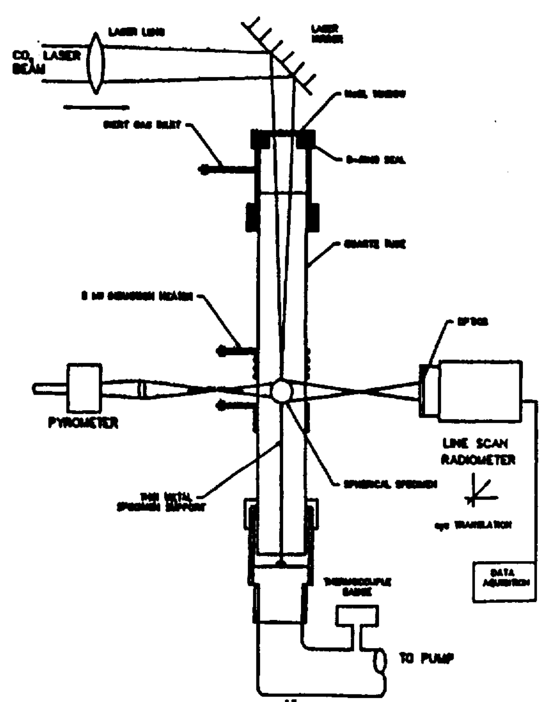
$$M_a = \frac{d\gamma}{dT}\frac{\Delta T\ r}{\rho\ \alpha\ \nu}$$

VISCOSITY

Lamb's relationship equates the time constant, τ, for damping of oscillations to viscosity, ν via the specimen radius, a, is used to calculate kinematic viscosity, ν, from the time dependence of liquid oscillation.

$$\nu = \frac{a^2}{5\tau}$$

THERMAL DIFFUSIVITY



Conclusions

Containerless melting combined with accurate NCTM provide the basis advancing TPMs on high temperature liquids.

Several properties could be measured in ground-based experiments with a well integrated facility.

Transport properties in liquids require minimal liquid motion and can best be conducted in reduced gravity.

A focussed program to advance key measurement techniques could provide much of the data required for industrial process modelling.

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