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# "Soft Condensed Matter" Variations in Structural Properties at Sol to Gel Transition- An Ultrasonic Study of Elastic Properties at 10Mhz in Sodium Meta Silicate Gel

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***Abstract: The materials which are highly deformable with characteristic length scale much larger than \hose of constituent molecules are grouped in the class "soft condensed matter "Sodium metasilicate gel' is a highly porous medium with pore sizes in the order of 20 A$^o$ to 1000 A$^o$. By varying PH of the solution gel of different densities can be prepared The velocities are measured using well known Pulse Echo Overlap method [1] using MATEC MODEL 7700 Pulse modulator and Receiver system at 10MHz using X cut quartz transducer. The "Mc Skimin Δt Criterion" [2] is applied so that velocities are accurate up to 0.001%. The strength of inter atomic forces and mechanism of gelation are interpreted on the basis of experimental results. The gel transition which is a zero dimensional to One dimensional one is observed in these samples. Percolation model explains this transitions in the most acceptable way. By applying hydrostatic pressure up to 50 atmospheres explains the stability of the structure.***
***Key words: Soft condensed matter, pulse echo overlap, gelation, percolation, zero order to three dimensional transition***

## I. INTRODUCTION

The colloidal suspensions, ferrofluids, emulsions, Lyotropic liquid crystals, gels and forms includes the complex system called soft condensed matter They have mesoscopic length scales from 10 A$^0$ to 1000A$^0$. Because of their complexity and flexibility of their structure they are also called complex fluids. [3]. SCM has technological importance in industries dealing with catalysis, enhanced oil recovery, chromatography etc. [4,5]. Multicomponent nature is shown by colloidal systems because of their size and shape of particles. Long range interaction predominantly of screened coulomb repulsion are existing in these system. So the particles of the systems are often called soft spheres. By changing the particle density or concentration the screening length can be easily tuned. Van-der Waals interaction which is of short range interaction also exists besides repulsive potential in these systems. This results in a coulomb barrier of few $K_BT$ provides stability against flocculation in these soft condensed materials. The other colloidal system of interest is ferro fluids [6], electrohelogical fluids, [5]. The X ray and light scattering studies reveals a non-equilibrium structure for colloids, lamellar and sponge phase etc. as a function of shear rates. Dynamics of colloids are recently probed by Multiple scattering method. The difference between a solid and fluid lies in their contrasting response to the applied shear. Mechanical energy is stored by solids while as in the case of fluids dissipate mechanical energy and are viscous. Ultrasonic studies in visco-elastic media are carried out by Meeten and Shermen [7]

The percolation theory [8] can explains the properties of the random systems taking variations in the interconnections present. According to this model there happens a sharp transition at which a long range order suddenly appears. These transition can be achieved either of three ways, by increasing density, concentration, occupation. In this work studies about the above transition. The transition from to sol to gel is very similar to glass transition. The sol-gel phase transition and molecular size contribution are first studied by Flory and Stock Mayer [9]. A sharp gel point is predicted by their work. Sols are solutions contains finite molecules while gel refers to system containing extended network. Sol- gel transition the system is no longer liquid are a conventional solid. Such systems are grouped in the definition soft condensed matter. A mechanical rigidity is appeared at sol to gel transition point. At the gel point gel macromolecules co-exist with remaining sol molecules. Because of the variation in the cluster forming ability, there exist a variation in density with varying $P^H$ of the solution. So ultrasonic a velocity and attenuations varies with density. At gel point one can expect a change in a slope in the density - velocity variation plot of the gel system.

In this work measures the longitudinal velocity and attenuation in the gel samples as a functions of density and temperature. The results are interpreted on the basis of percolation model.

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### *A. Experimental Technique.*

The longitudinal velocities inside the samples are measured using "PEO" method which is already mentioned above. [11] The "McSkimin Δt Criterion" is applied. "Nonaque" is used as bonding material for longitudinal measurements.

"Sodium metasilicate" gel has been prepared by dissolving sodium metasilicate powder in water. The following reaction will happen

$$Na_2Si_2O_3 + 3\ H_2O \rightarrow 2NaOH + Si(OH)_4$$

The PH of the solution is adjusted by adding sulphuric acid. If the $^{PH\ i}$s in the range 10 to 11 gelation takes place. $Si(OH)_4$ is tetrahedral molecule consists of a silicon bonded to four OH groups. Each $Si(OH)_4$ monomers can form as many as four reacted bonds. These bonds oxygen bridges to silicon atoms originated from other molecules. The condensation process progresses and more and more cross links forms and larger molecules appears. As result density of the system varies and at a particular density an infinitively extended macro molecules are formed which is the sol-gel transition point

For measuring ultrasonic velocity and attenuation a cell is designed and transducer is attached to one of it sides. The sides of the cell are parallel to$0.05^0$. The experimental gel samples are taken in this cell. These samples are rich in liquid. For temperature measurements this cell is placed inside a chamber and temperature is varied from $30^0$C to $40^0$C.The sample cell is placed in ice and temperature variation s from $0^0$C to $30^0$C are carried out.

## II. RESULTS AND DISCUSSION

The longitudinal velocity and attenuation is measured as a function of density and is shown in the table below.

| Density gm/cc | Longitudinal velocity m/s | Elastic constant C11 GPa | Longitudinal atteunuationin db/Length |
|---|---|---|---|
| 1.043 | 1473.28 | 0.2264 | 3.32 |
| 1.052 | 1489.39 | 0.2333 | 3.195 |
| 1.063 | 1502.44 | 0.2400 | 3.065 |
| 1.077 | 1520.38 | 0.2490 | 2.952 |
| 1.085 | 1561.44 | 0.2645 | 2.838 |
| 1.093 | 1589.32 | 0.2761 | 2.765 |
| 1.101 | 1602.14 | 0.2826 | 2.693 |
| 1.123 | 1636.23 | 0.3007 | 2.521 |
| 1.132 | 1665.54 | 0.3140 | 2.410 |

1.12 1.14

The ultrasonic velocity increases with increase in density and attains a maximum value 1665.54 and thereafter if PH is varied gelation is not taking place. The value of the density at this point is 1. 132.The Graph is divided into two parts PQ and QR and region above QR. In the low density region PQ, the proportion of clusters of larger sizes are very small. As density of the system increases clusters grow in size as predicted by percolation model.

$$Cij = \frac{1}{\sum bij} \sum \frac{dEij}{\Delta d} \frac{1}{V}$$

The first term in the equation $\frac{1}{\sum bij}$ can be expanded as a series AF $+BF^2$ $+CF^3$ +…. Where F is always less than one. Each term in the series is a measure of Probability of encountering clusters of particle sizes 1,2,3, etc. At low densities, the sum of the above series is very small so elastic constants are very small. For the existence of shear modulus clusters whose ends are not free are required. These type of clusters cannot be formed at low density limit. So shear modulus will be zero. As the density approaches the value 1.077 a point represented as Q in the graph there exists co-existence of sol gel molecules .so Q corresponds to the transition point. After the point Q systems with infinite clusters co-exist with finite sol molecules so the value of $\frac{1}{\sum bij}$ is greater than one so there exists a change of slope of the graph at point Q. If density is increased further the systems behaves as solid. The ultrasonic attenuation decreases because of formation of clusters of large size. So the sizes of pores filled with liquid decreases and scattering from this pore areas decreases leads to a corresponding decrease in attenuation.

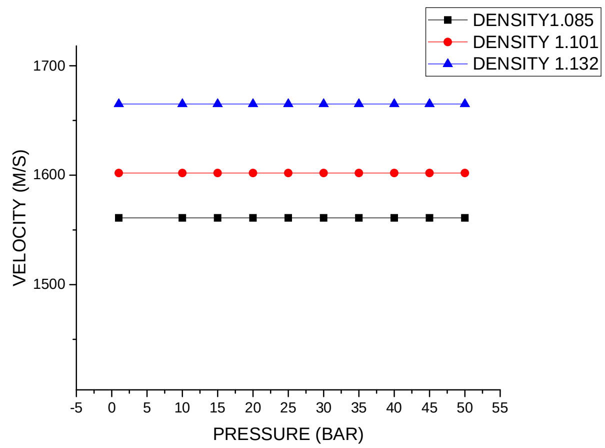
Velocity plot of cooling

Velocity plot of heating

The temperature variation of ultrasonic velocities is also measured. At a particular density when temperature changes from room temperature to 400C the variation in velocities are very small. This can be accounted as follows. when temperature increases due to thermal vibration of clusters the clusters with larger size breaks down so density of system decreases. So more gel molecules get converted into sol molecules. When temperature is decreased the sol molecules trapped between the clusters of larger size join with clusters of large size and more gel macromolecules are formed. So gelation increases more rapidly corresponding increase in ultrasound velocity

### *A. Pressure Variation Measurements*

The sample with the cell is enclosed in a pressure chamber and pressure is increased hydrostatically from 1 atmosphere to 50 atmospheres by a compressor connected to the equipment. The velocities are almost constant with increase of pressure. So gel is a stable semi solid which is get reflected by the measurement.




## III. CONCLUSION

The sol-gel transition is well studied. The results highlight the percolation model developed. The s ultrasonic studies reveal the structural modification happening at sol –gel transition point. The results also predict a sharp gel point and in well agreement with theory.

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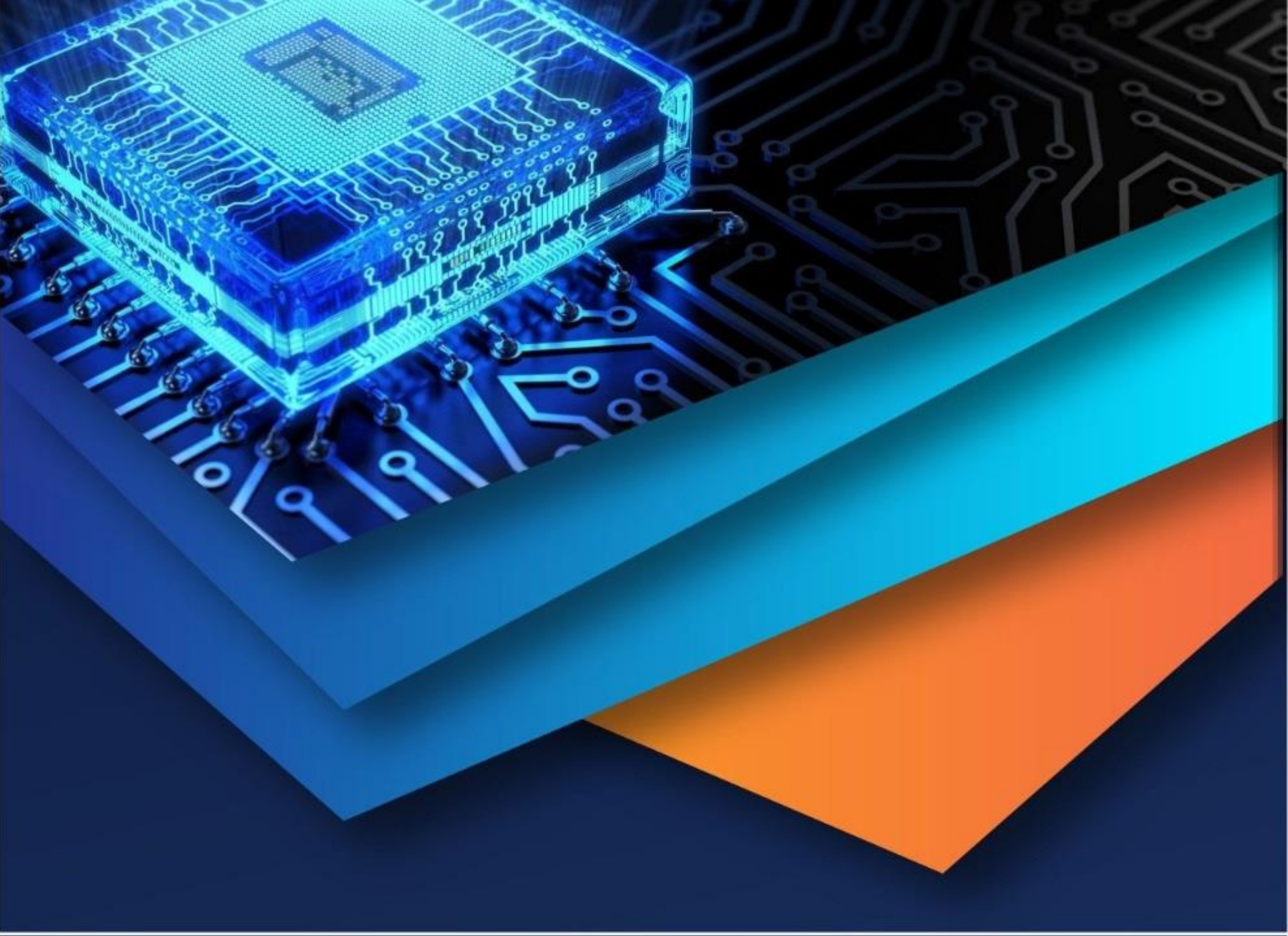


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