# THE EFFECT OF PARTICLE SIZE ON THE ELECTRICAL PROPERTIES OF POLY-VINYL ALCOHOL

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## ABSTRACT

*The present work deals with some factors affecting the electrical behaviour of polyvinyl alcohol (PVA) in the form of powder with different particle size (P) ranging from 63 up to 450 µm. The study of both dielectric constant (ε') and thermally stimulated depolarization current (TSDC) show remarkable particle size dependence. It was also found that compressing the sample at higher pressure, causes a decrease in the peak value of ε' versus T curve and has pronounced effect on both the shape and the value of TSDC. The activation energy was determined for different particle sizes using the initial rise method.*

## INTRODUCTION

The particle size of a polycrystalline material has a major effect on its electrical properties. This effect for ferroelectrics as well as ceramics, has been studied by many investigators[1-3]. Some works have been reported on compressed powders of different particle sizes in ferroelectric materials[4-6]. The dielectric constant of tryglycine sulphate (TGS) was found to decrease with decreasing the particle size, while the phase transition temperature shifted toward higher values. The role of particle size and grain size on the dielectric behaviour of peroviskite relaxor ferroelectric was also investigated[7]. Sharma et all[8] have studied the depolarization current in PVA, the activation energy and the relaxation time have been calculated under different polarization conditions. The formation of heterocharge was explained by dipole alignment and migration of charge over microscopic distances with trapping.

Masanobu Nagura[9] studied the molecular motion in a PVA crystal. It was established that the molecular motion in the crystal above $100^{0}C$ occurred in only the loose packing region.

To get an insight into the effect of particle size of polymer, we shall study the dielectric permittivity and thermally stimulated depolarization current for PVA.

## EXTPERIMENTAL

Polyvinyl alcohol in the form of powder was supplied by "BDH Chemical company, JAPAN". This powder was separated to different particle size by using the sevie analysis ranging from 63 to 450μm in mesh size. The same weight of powder with different particle sizes was comprressed under a hydrostatic pressure of 3.57 $Ton/cm^2$ to form tablets with a thickness of about (0.15 cm) and area (1.4 $cm^2$). The major surfaces of the samples were painted with a very conductive silver paint to ensure good contact. The dielectric constant, and thermally stimulated depolarization current, were measured during heating at a rate of about $1^{0}C/min$. The capacitance was measured by means of a precision capacitance bridge (Tesla, B.M. 400g, with sensitivity of $10^{-3}$pF). Poling of samples was done using a static electric field of about 2 kV/cm for a period of half an hour at constant temperature ($100^{\circ}C$). The sample was then cooled to room temperature in the presence of the applied electric field. The specimen was then short circuited after removing the field to eliminate surface charges, and the TSDC was observed using an Electrometer (Keithley 616, Digital Electrometer) by reheating the sample at a constant rate of about $1^{\circ}C/min$.

The samples used in this work with their particle sizes and pressure are represented Table 1.

Table (1)

| Sample No. | Particle Size (P) (μm) | Pressure (Ton/cm$^2$) |
|---|---|---|
| SM1 | 63 < P < 149 | 3.57 |
| ME1 | 149 < P < 315 | 3.57 |
| LA1 | 315 < P < 450 | 3.57 |
| SM2 | 63 < P < 149 | 1.78 |
| ME2 | 149 < P < 315 | 1.78 |

## RESULTS AND DISCUSSION

### 1. Dielectric measurements

The temperature dependence of the dielectric constant, ε' (T), for PVA samples was measured over a rang of temperatures from 30 up to 120°C at nearly a constant rate of 1°C/min. Figure (1) illustrates the dielectric behaivour for two series compessed at 3.57 Ton/cm$^2$ and 1.78 Ton/cm$^2$, with different particle sizes. Hence, the following points can be noticed :

(a) The dielectric permittivity depends on the particle size. This obviously appeared at low and high temperatures, where ε'(T) decreases with increasing the particle size, contrary to what is observed in the case of ferreoelectrics[6].

(b) The samples of particle size 149μm < P < 315 μm (medium) and of particle size 315 < P < 450 μm (large) show peak values ($\varepsilon'_{max}$) which are nearly the same, but the peak positions ($T_m$) depend on the particle size, where $T_m$ is about 80°C for medium and 90°C for large particle size. This will be more explained on discussing the rise of activation energy for larger particle size sample latter on.

(c) Comparing the behaviour for samples with small particle size and medium particle size according to the pressure (Figure (2)) it is clear that for particle size 63 < P < 149μm, a slight hump of $\varepsilon'_{max}$ appeared around 50°C for the pressure (1.78 Ton/cm$^2$), while at a pressure of (3.57 Ton/cm$^2$) this hump

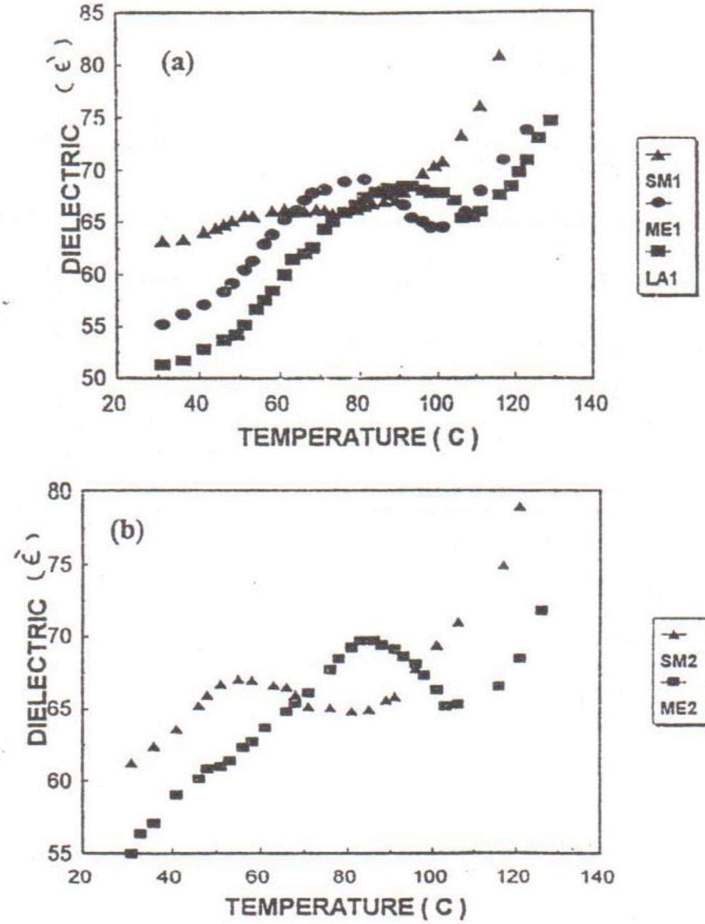


**Fig. 1 :** Temperature dependence of dielectric constant for PVA with different particle sizes at 3.57 ton/cm$^2$ (a), at 1.78 ton/cm$^2$(b)

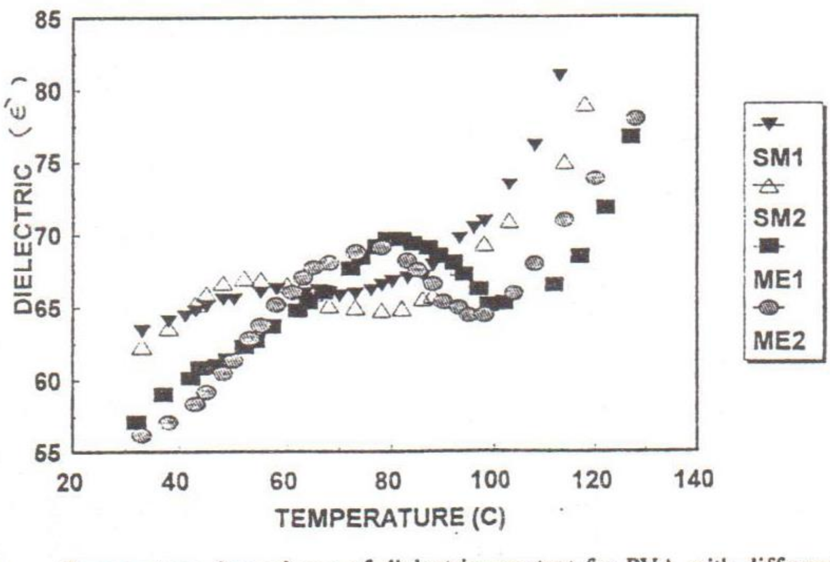


**Fig. 2 :** Temperature dependence of dielectric constant for PVA with different particle sizes at 3.57 ton/cm$^2$ (SM1, ME1), and 1.78 ton/cm$^2$ (SM2, ME2)

becomes broader and nearly vanishes, and for medium particle size the peak position is shifted towards lower temperature.

The decrease of dielectric constant with increasing particle size may be due to air gaps which reduce the density of samples by increasing the porosity; the latter is inversely proportional to the particle size. It was naturally found that the pressure is more effective on the smallest particle size, where the peak value appeared at low pressure. While increasing pressure to 3.57 Ton/cm$^2$ a plateau region occur instead of the peak, which may be attribted to two factors: the first, is the effect of pressure which cause more packing, the second is the increase of the surface energy between the particle of small size[10]. In fact, the surface of a given particle could have the form (1/4)nNe erg/cm$^2$, where e is the energy required to separate a particle from its nearest neighours n, and N is the number of particles per cm$^2$. If the diameter of the particle is $a_o$ then N is proportional to $1/a_o^2$ per cm. This means that the surface energy increases as the redius of the particle decreases. Both factors are combined to cause an apparent internal biasing force which restricts the motion of the chains so that no peaks appear.

Figure (3) show the behaviour of the sample during the second run, measured after 24 hours, the observed peak $\varepsilon'_{max}$ disappeared. This may be related to the degradation, which occured for PVA powder after heating in the first run up to 130°C.

## 2 - Thermally stimulated depolarization current (TSDC)

The TSDC for samples with different particle size under the mentioned conditions was measured, Figure (4) show that the smallest particle size gives higher values of TSDC than the larger particle size when compressed at 1.78 Ton/cm$^2$, and a deviation in TSDC values over all temperature range take place. Beside, the TSDC levels off in the temperature range from 50 to 100°C. Such an effect may be expected when the equilibrium polarization condition occur.

Unlike the above series, samples prepared at 3.57 Ton/cm$^2$, Figure (5) show similar TSDC at low temperature, while slight deviation begins at higher temperatures. This is probably associated with the thermal motion of the chains.

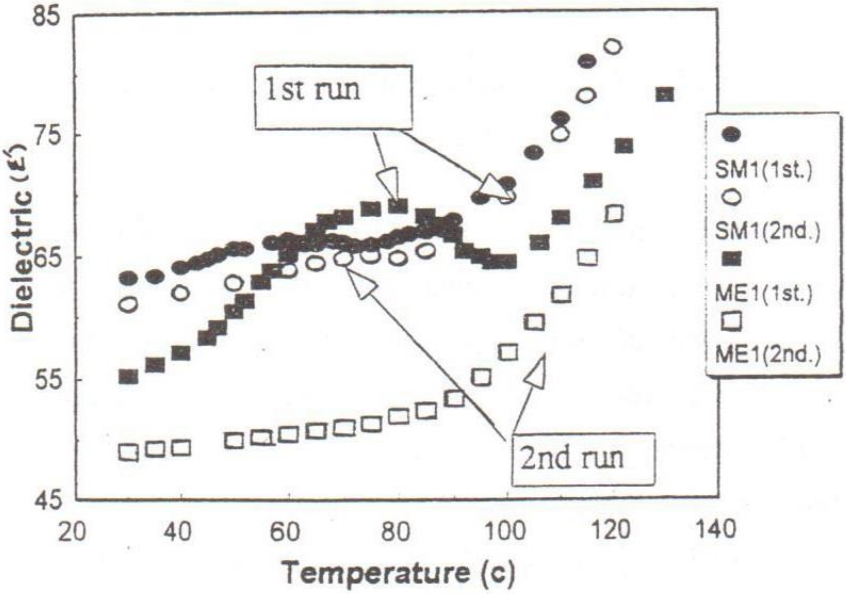


Fig. 3 : Effect of second cycle on the dielectric behaviour of PVA compressed at 3.57 ton/cm$^2$.

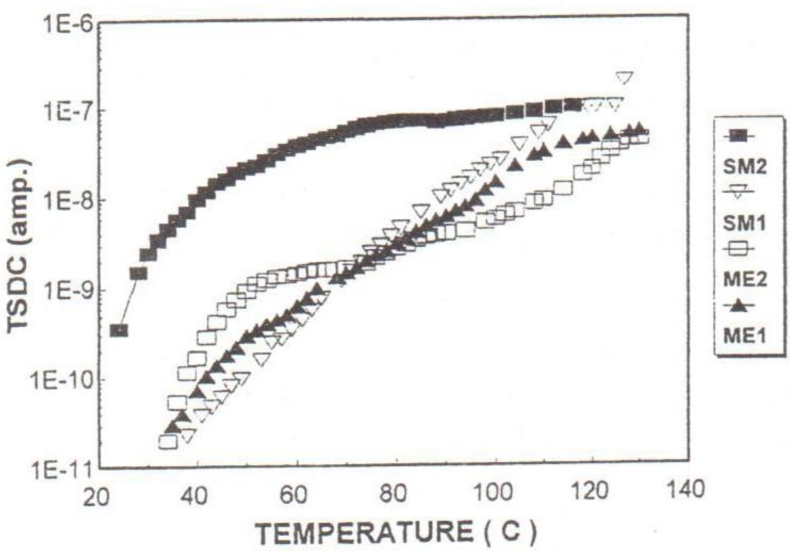


Fig. 4 : Temperature dependence of TSDC for PVA with different particle sizes at 3.57 and 1.78 ton/cm$^2$.

As a further evidence of this picture the activation energy was calculated using the initial rise method, Figs (6).

**Table (2)**
**Activation Energy**

| Particle Size (μm) | Activation energy (eV) | Pressure (Ton/cm$^2$) |
|---|---|---|
| 63 < P < 149 | 2.51 | 1.78 |
| 149 < P < 315 | 2.41 | 1.78 |
| 63 < P < 149 | 1.065 | 3.57 |
| 149 < P < 315 | 1.260 | 3.57 |
| 315 < P < 450 | 1.340 | 3.57 |

It can be seen Table 2, that the activation energy of PVA samples having the same particle size decrease as the pressure increases; since increased pressure may lower the defective layer which lead-in turn - to a drop in the activation energy[9]. One may attributed the remarkable effect of the particle size on the temperature dependence of TSDC to the difference in their activation energies.

## CONCLUSION

The dielectric permittuvity of PVA powder decreases with increasing the particle size and the position of the peak ($T_m$) shifts towards higher values.

The pressure is more effective on the smallest particle size 63<P<149μm, while increasing pressure to 3.57 Ton/cm$^2$ causes the annihilation of the peak in ε' versus T curve.

Thermally stimulated depolarization current can be enhanced by decreasing both particle size and pressure.

The measured activation energy depeneds on the particle size and the compressed pressure.

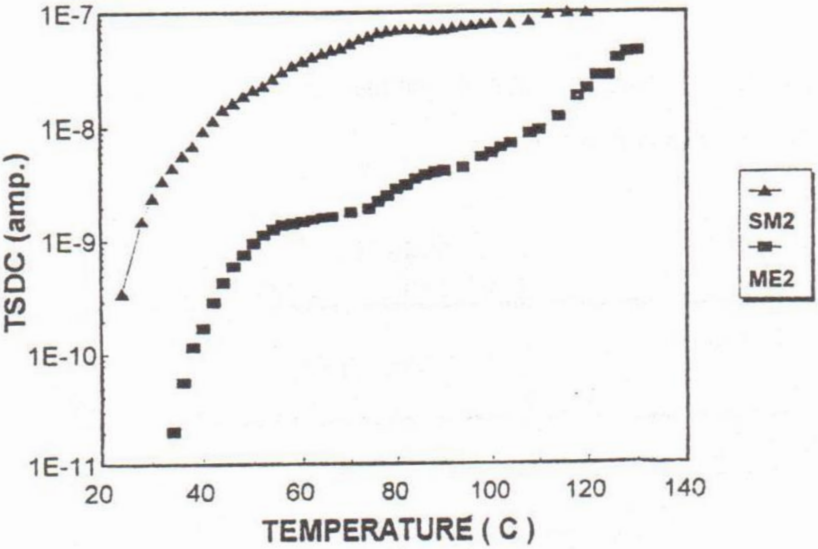


Fig. 5 : Temperature dependence of TSDC for PVA with different particle sizes at 1.78 ton/cm$^2$.

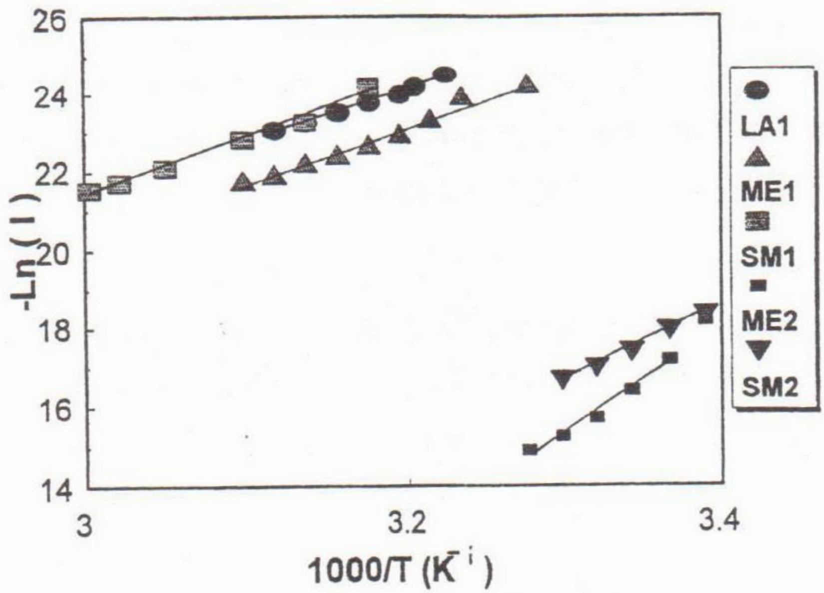


Fig. 6 : ln (I) vs 1000/T for PVA with different particle sizes, compressed at 3.57 and 1.78 ton/cm$^2$.

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