Synthesis and physico-chemical study of halogenated aromatic cardo polysulfonates

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Poly(R, R′, 4, 4′-cyclohexylidene diphenylene-2, 4-toluene disulfonate) (PS-3: R=R′=C1 and PS-4: R=CH₃ and R′=C1) have been synthesized by interfacial polycondensation technique by using water-chloroform (4:1 v/v) as an interphase, alkali (0.12 mol) as an acid acceptor and cetyl trimethyl ammonium bromide as an emulsifier. The reaction time and temperature are 3½ h and 0ºC, respectively. The structure of PS-3 and PS-4 are supported by IR and NMR spectral data. The intrinsic viscosities of the said polymers are determined in different solvents at three different temperatures 30, 35 and 40ºC. The calculated and experimental (floatation method) densities are found in good agreement with small fractional free volume. PS-3 and PS-4 possess excellent resistance to hydrolytic attack against water, acids, alkalis and salt, moderate tensile strength (15.6-17.0 N/mm²), excellent volume resistivity (1.1 to 3.9 × 10¹⁵ Ω cm), high electric strength (33.9-32.9 kV/mm) and low dielectric constant (1.36), good thermal stability (360ºC and 340ºC) and high Tg (143.5ºC and 134.3ºC). DSC and DTA endo/exothermic transitions have supported physico-chemical changes. No change in tensile strength is observed but volume resistivity has increased 1.5 to 54.2 times, while electric strength has increased 2.5 to 3.5 times upon introducing methyl and chlorine groups alone or in combination. The improvement in volume resistivity and dielectric constant is due to cancellation of partial charges present on the polymer molecules.

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Polymer chains containing inorganic atoms such as polysulfone and polysulfonates, have attracted attention as high performance polymers and used as engineering plastics¹. Thermally stable polymers containing a rigid backbone exhibit high softening temperature and poor solubility²-⁵ but introduction of cardo (Latin meaning a loop) groups⁶ (phthalide, phthalimidine, anthrone, fluorine and cyclohexyl) across the backbone chains result in amorphous polymers with enhanced solubility, thermal stability and thermo-mechanical properties. Thermally stable linkages such as ether, sulfide, sulfone and ketone lower the softening temperature and increase the solubility. Flexible linkages result in somewhat lowering of the end use temperature. Halogenated polymers are well-known for their excellent flame retardant property. The presence of cardo groups across the backbone chain not only enhances the solubility but also mechanical and thermal properties⁶. They find their applications as thermo-plastic molding parts, films, fibers, coatings, adhesive materials, fire proofing, lacquers and paints. The literature survey on aromatic polysulfonates reveal that the most of the work is confined with bisphenols and very little work has been reported on cardo bisphenols. Recently, we have reported a considerable work in the field of aromatic polysulfonates⁷-¹⁴ containing cyclohexyl as a cardo group. We have studied structure-property relationship among various polysulfonates of 1, 1′-bis(R, R′, 4-hydroxy phenyl)cyclohexane (R, R′=H, CH₃, Cl, Br) and 1, 3-benzene, 2, 4-toluene, 3, 3′-benzophenone, 4, 4′-diphenyl, 4, 4′-diphenylmethane, 4, 4′-diphenyl ether disulfonyl chlorides and observed small variation in thermal stability (340-380ºC). The substituents groups and the structure of the disulfonylechlorides have affected the tensile strength (1.1-38.4 N/mm²) and electrical properties such as volume resistivity (1.1 × 10¹⁴- 1 × 10¹⁵ Ωcm) and electric strength (7.4-27.1 kV/mm) to a considerable extent. All the polysulfonates possess excellent hydrolytic stability against acids, alkalis and salt solutions (10%) at room temperature. With a view to understand the effect of polar substituents on physical properties, the present work describes the synthesis and physico-chemical study of halogenated aromatic cardo polysulfonates (I):

Experimental Procedure
Materials

The chemicals used were of laboratory grade and were purified according to reported methods¹⁵. 1, 1′-
Bis(R, 4-hydroxy phenyl) cyclohexane (BC: R=H, and MeBC: R=CH₃) and 1, 1'-bis (R, R'-4-hydroxy phenyl) cyclohexane (CIB: R=R'=Cl and ClMeBC: R = CH₃ and R' = Cl)¹⁶-¹⁸ and toluene -2, 4-disulfonyl chloride (TDSC)¹⁹ were synthesized according to reported methods. BC, MeBC, CIB and ClMeBC were recrystallized three to four times from benzene and methanol-water systems, while TDSC was recrystallized from chloroform-n-hexane. The yields and m.p. of BC, MeBC, CIB, ClMeBC and TDSC were 81, 77, 70, 70 and 48%; and 186°C, 186°C, 180°C, 181°C and 52°C, respectively. The emulsifier cetyl trimethyl ammonium bromide (CTAB) (Sisco-chem) was used as received.

**Polymer synthesis**

Into a 250 mL three-necked flask equipped with a high-speed mechanical stirrer and a thermometer, a 0.005 mol CIB/ClMeBC and 0.012 mol sodium hydroxide were dissolved in 50 mL of distilled water. The solution was cooled to 0°C and 50 mg a cationic emulsifier cetyl trimethyl ammonium bromide was added and the solution was stirred vigorously for about 15 min. A solution of 0.005 mL TDSC in 12.5 mL chloroform was added dropwise over 10 min. The emulsion was vigorously stirred for 3½ h at 0°C. The polymer was isolated from a large excess of methanol, filtered, washed well with water and finally with methanol and dried at 50°C. The polymers were further purified at least three times from chloroform-methanol system. The yield was ~ 87-91%. The thin and thick transparent films were cast from 1% and 4% chloroform solutions, respectively and vacuum dried prior to their further characterization.

**Measurements**

The IR spectra (thin films) of PS-3 and PS-4 were scanned on a Bruker FTNMR (300 MHz) spectrometer by using CDCl₃ as a solvent and TMS an internal standard. The viscosity measurements at three different temperatures: 30°C, 35°C and 40°C were carried out with an Ubbelohde type suspended level viscometer and the intrinsic viscosities were determined by Huggins relationship. The density measurements were carried out by floatation method¹¹,¹³. The hydrolytic stability of polymer films was carried out at room temperature for varying period in water and 10% each of acids, alkalis and salt solutions. The electric strength (ASTM-D-149-92), volume resistivity (ASTM-D-257-92) and tensile strength measurements of 15 mm wide and 47-68 μm thick films were made at room temperature on a high voltage tester (Automatic Mumbai) by using 25/75 mm brass electrodes, Schering Bridge (Tettex Switzerland), Hewlet Packard high resistance meter at 500 V DC after 60 s and Instron Universal Testing Machine, Model No. 1186 at a testing speed of 50 mm/min, respectively. The TG-DTA (15ºC/min) and DSC (10ºC/min) measurements were made on a Mettler TS system in an N₂ atmosphere.

**Results and Discussion**

**IR and NMR spectral data**

Fig. 1 shows IR (thin films) spectra of PS-3 and PS-4. The characteristic IR absorption bands (cm⁻¹) of PS-
3 and PS-4 are 1166.9 (\(-\text{O-SO}_2\), \(\nu_a\)), 1406 (\(-\text{O-SO}_2\), \(\nu_s\)) and 758 and 748.5 (C-Cl str.) besides normal modes of vibrations of aliphatic and alicyclic groups. Fig. 2 shows NMR spectra of PS-3 and PS-4. The chemical shifts (ppm), multiplicity of various types of protons and coupling constant (\(J\)) are assigned as follow. For PS-3: 1.604-1.432[9H, d, \(\beta+\gamma\)-CH\(_2\)+-CH\(_3\)], 2.132[4H, s, \(\alpha\)-CH\(_2\)-], 6.864-6.802[2H, m, Ar-H(b), \(J=8.2\)], 7.134-7.074[2H, t, Ar-H(a), \(J=9.0\)], 7.549-7.523[1H, d, Ar-H(e), \(J=7.8\)], 7.917-7.890[1 H, d, Ar-H(d), \(J=8.1\)], 8.200-8.149[1H, t, Ar-H(c), \(J=7.8\)] and for PS-4: 1.576-1.453[6H, d, \(\beta+\gamma\)-CH\(_2\)+-CH\(_3\) (TDSC)], 2.216-2.004[10H, d, \(\alpha\)-CH\(_2\)+-CH\(_3\)(MeBC)], 7.054-6.649[4H, m, ArH(ClMeBC)], 7.557-7.531[1H, d, Ar-H(e), \(J=8.0\)], 7.977-7.951[1H, d, Ar-H(d), \(J=7.8\)], 8.372[1H, s, Ar-H (c)]. The signal due to residual chloroform and water are appeared at about 7.26 and 2.8, respectively. The integrated peak areas are found in accordance with different types of protons. Thus, IR and NMR spectral data supported the structures of PS-3 and PS-4.

**Viscosity measurements**

The solution viscosity is a direct measure of hydrodynamic volume of polymer molecules and furnishes knowledge on molecular interactions occurring in the solutions. The least square intrinsic viscosity \([\eta]\) and Huggins constant \((k')\) of both the polymers in different solvents: chloroform (CF), 1,2-dichloroethane (DCE) and tetrahydrofuran (THF) at three different temperatures: 30ºC, 35ºC and 40ºC are reported in Table 1. The observed thermodynamic goodness of the solvents is THF > DCE > CF. From Table 1, it is clear that PS-3 and PS-4 are moderately high molecular weights. \([\eta]\) has decreased with increasing temperature but the observed effect is small due to flexible nature of the polymer chains. No systematic trend in \(k'\) is observed due to polydisperse nature of PS-3 and PS-4. Thermally stable polymers containing rigid backbone exhibit high softening temperature and poor solubility and as a result they are difficult to process. Cardo polymers of low viscosity and improved solubility, high \(T_g\) and high thermal stability are reported in the literature.

**Density measurements**

The densities of PS-3 and PS-4 films were determined by a flotation method according to our recent publication and the average of six measurements is reported below.

![Fig. 2—NMR spectra (CDCl\(_3\)) of PS-3 and PS-4](image_url)

<table>
<thead>
<tr>
<th>Polymer</th>
<th>CF</th>
<th>(\eta)</th>
<th>(k')</th>
<th>(\gamma)</th>
<th>DCE</th>
<th>(\eta)</th>
<th>(k')</th>
<th>(\gamma)</th>
<th>THF</th>
<th>(\eta)</th>
<th>(k')</th>
<th>(\gamma)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PS-3</td>
<td>0.64</td>
<td>0.38</td>
<td>0.991</td>
<td></td>
<td>0.79</td>
<td>0.26</td>
<td>0.993</td>
<td></td>
<td>1.08</td>
<td>0.14</td>
<td>0.998</td>
<td></td>
</tr>
<tr>
<td>PS-4</td>
<td>0.48</td>
<td>0.77</td>
<td>0.997</td>
<td></td>
<td>0.66</td>
<td>0.28</td>
<td>0.983</td>
<td></td>
<td>0.75</td>
<td>0.25</td>
<td>0.997</td>
<td></td>
</tr>
<tr>
<td>PS-3</td>
<td>0.60</td>
<td>0.42</td>
<td>0.995</td>
<td></td>
<td>0.70</td>
<td>0.33</td>
<td>0.993</td>
<td></td>
<td>0.90</td>
<td>0.29</td>
<td>0.988</td>
<td></td>
</tr>
<tr>
<td>PS-4</td>
<td>0.48</td>
<td>0.62</td>
<td>0.949</td>
<td></td>
<td>0.59</td>
<td>0.45</td>
<td>0.991</td>
<td></td>
<td>0.73</td>
<td>0.23</td>
<td>0.983</td>
<td></td>
</tr>
<tr>
<td>PS-3</td>
<td>0.56</td>
<td>0.41</td>
<td>0.984</td>
<td></td>
<td>0.56</td>
<td>0.63</td>
<td>0.995</td>
<td></td>
<td>0.84</td>
<td>0.23</td>
<td>0.987</td>
<td></td>
</tr>
<tr>
<td>PS-4</td>
<td>0.44</td>
<td>0.61</td>
<td>0.979</td>
<td></td>
<td>0.55</td>
<td>0.46</td>
<td>0.979</td>
<td></td>
<td>0.62</td>
<td>0.41</td>
<td>0.994</td>
<td></td>
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</table>

![Table 1—The intrinsic viscosity, Huggins constant and correlation coefficient for PS-3 and PS-4 in three different solvents and temperatures](table_url)
measurements with standard deviation from the mean are reported in Table 2. Theoretical densities (ρ) of PS-3 and PS-4 were calculated according to Slonimskii et al.\textsuperscript{20}:

\[ \rho = \frac{KM}{N_A \Sigma \Delta V_i} \]

where \( K \) is the packing coefficient and for film samples \( K \) is 0.695, \( M \) is the polymer repeat unit molecular weight, \( N_A \) is the Avogadro’s number and \( \Sigma \Delta V_i \) is the intrinsic volume of the polymer repeat unit. The calculated densities are reported in Table 2. From Table 2, it is evident that the experimental and theoretical densities are in good agreement justifying the correct choice of the packing coefficient. A small difference between experimental and theoretical density is due to the fractional free volume, which is observed in most of the polymers. The introduction of polar groups (Cl and CH\(_3\)) in bisphenol moiety resulted change in density due to change in \( M \) and \( \Sigma \Delta V_i \). Halogenated polysulfonates (PS-3 and PS-4) have greater density than that of unhalogenated polysulfonates (PS-1\textsuperscript{10} and PS-2\textsuperscript{7}) due to high packing density.

Mechanical and electrical properties

For end use applications, the knowledge of thermo-mechanical and electrical properties of polymers is very important to a designer. Tensile data of polymers are useful for the purpose of engineering design and understanding the characteristic of the materials. The tensile strength of PS-3 and PS-4 along with unhalogenated polysulfonates (PS-1\textsuperscript{10} and PS-2\textsuperscript{7}) are reported in Table 3. From Table 3, it is clear that the tensile strength of PS-3 and PS-4 is almost the same as that of PS-1 and PS-2 indicating no much effect of introduction of polar groups on mechanical strength. Moreover moderate strength is due to moderate molecular weights as supported by viscosity data (Table 1). Mechanical properties of polymers depend upon several factors namely temperature, humidity, loading conditions, rate of loading, morphology, molecular architecture, molecular weight and molecular weight distribution, degree of cross-linking, extent of plasticization and fillers\textsuperscript{21,22}. Majority of the synthetic organic polymers are well-known for their electrical insulation characteristics. Volume resistivity and electric strength data are very useful for comparing relative insulation quality of material selection, to evaluate the effects of material composition and environment. They are useful to material scientists to design specific properties in combination. Electric properties of polymeric materials are affected by several factors namely humidity, impurities, degree of resin cure, temperature, nature of polymers, nature of fillers and additives, dimensions and geometry of electrodes and electrode material, sample thickness, time of voltage application and extent of ageing.

The electric strength, volume resistivity and dielectric constant (\( \varepsilon \)) data of PS-3 and PS-4 are reported in Table 3 along with PS-1\textsuperscript{10} and PS-2\textsuperscript{7} for comparison purposes. From Table 3, it is evident that the introduction of polar groups (CH\(_3\) and Cl) affected both volume resistivity and electric strength to a considerable extent but \( \varepsilon \) is slightly decreased due to presence of polar groups. The slight decrease in dielectric constant might be due to decrease in static charges on halogenated molecules. Volume resistivity has increased 1.5 to 54.2 times, while electric strength has increased 2.5 to 3.5 times due to presence of methyl and chlorine alone or in combination. The drastic improvement in volume resistivity of PS-4 as compared to PS-1\textsuperscript{10} is due to presence of electronegative chlorine and electropositive methyl groups, which mutually cancel the static charges on polymer molecules. In case of PS-3, volume resistivity improvement is about 1.5 times due to chlorine group alone. No much change in electric strength is observed due to presence of chlorine and methyl groups and chlorine groups alone. The low dielectric constant, excellent volume resistivity and high electric strength of polysulfonates under study signify the properties of good insulators, i.e., high

Table 2—Calculated and experimental (floatation) densities of PS-3 and PS-4

<table>
<thead>
<tr>
<th>Polymer</th>
<th>( M, , g )</th>
<th>( \Sigma \Delta V_i, , A^0 )</th>
<th>Density, g/cm(^3)</th>
<th>Fractional free volume change</th>
</tr>
</thead>
<tbody>
<tr>
<td>PS-1\textsuperscript{10}</td>
<td>484</td>
<td>405.24</td>
<td>1.3605(\pm)0.0017</td>
<td>1.3782</td>
</tr>
<tr>
<td>PS-2\textsuperscript{7}</td>
<td>512</td>
<td>438.14</td>
<td>1.3183(\pm)0.0003</td>
<td>1.3484</td>
</tr>
<tr>
<td>PS-3</td>
<td>622</td>
<td>464.40</td>
<td>1.4339(\pm)0.001</td>
<td>1.5455</td>
</tr>
<tr>
<td>PS-4</td>
<td>581</td>
<td>469.22</td>
<td>1.3439(\pm)0.0054</td>
<td>1.4288</td>
</tr>
</tbody>
</table>

Table 3—Mechanical and electrical properties of PS-3 and PS-4

<table>
<thead>
<tr>
<th>Polymer</th>
<th>Tensile strength ( N/mm^2 )</th>
<th>Volume resistivity ( \Omega , cm )</th>
<th>BDV ( kV/mm )</th>
<th>( \varepsilon ) ( 230V/50 , Hz ) atm. temp</th>
</tr>
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<tbody>
<tr>
<td>PS-1\textsuperscript{10}</td>
<td>16.5</td>
<td>(7.2 \times 10^{14})</td>
<td>9.6</td>
<td>-</td>
</tr>
<tr>
<td>PS-2\textsuperscript{7}</td>
<td>18.5</td>
<td>(4.2 \times 10^{15})</td>
<td>23.6</td>
<td>1.47</td>
</tr>
<tr>
<td>PS-3</td>
<td>15.6</td>
<td>(1.1 \times 10^{15})</td>
<td>33.9</td>
<td>1.36</td>
</tr>
<tr>
<td>PS-4</td>
<td>17.0</td>
<td>(3.9 \times 10^{16})</td>
<td>32.9</td>
<td>1.36</td>
</tr>
</tbody>
</table>
resistivity, low dielectric constant, high electric strength and good arc resistance\textsuperscript{22}.

The charge storage involves the alignment of dipoles in a material in the direction of applied electric field. The source of the dipoles is polar groups in the polymer chains, heterogeneity due to partial crystallinity, molecular asymmetry and polar additives in the material. The charge on capacitors polarizes the molecules and as a result the charge in the surface increases. The volume resistivity mainly depends on the presence of free charges (electrons or ions) and their mobility.

Polymers having polar groups in their chain would be expected to have high dielectric constant. Besides segmental motions, side chains and polar substituents are also moving in polymers. Low dielectric constant of PS-3 and PS-4 might be due to restricted conformational changes, which make dipolar motion of repeating unit smaller and behaves rigidly under thermal motion. Thus, PS-1 to PS-4 possess excellent volume resistivity, high electric strength and low dielectric constant as compared to some useful plastics\textsuperscript{21,22} signify their importance as good insulators.

### Hydrolytic stability

Polymeric materials absorb moisture in humid atmosphere and when immersed in water. The extent of water absorption mainly depend on the presence of hydrophilic polar groups, void volume, additives, humidity and temperature. Hydrolytic stability of polymeric materials depends on the nature and molecular structure of polymers. The tendency of water absorption by polar groups present in polymers increases the dissociation and hydrolysis and hence introduces ionic impurities, which affect electrical and mechanical properties to a greater extent. Hydrolytic stability of PS-3 and PS-4 towards H$_2$O, 10% each of acids (H$_2$SO$_4$, HCl, HNO$_3$ and CH$_3$COOH), bases (NaOH and KOH) and NaCl solutions was determined by change in weight method at room temperature after 24 h, one week and one month. The % weight change data are reported in Table 4. From Table 4, it is clear that PS-3 and PS-4 showed ±0.8% weight change indicating excellent hydrolytic stability. Penetration of water in polymeric films may be transported through micro cracks. Absorbed water may induce irreversible changes like chemical degradation, cracking and debonding. Cracking and blistering cause high water absorption, while leaching of small molecules results in decrease in weight\textsuperscript{23}. A slight gain in weight may also due to surface solvolysis of polymer chains. The chemical resistance of PS-3 and PS-4 is found in accordance to other polysulfonates of this type\textsuperscript{7-14}.

#### Thermal analysis

The knowledge of thermal properties is very essential since it provides wealth of informations on characteristics degradation pattern, molecular architecture and degradation mechanism. Thermal stability of polymers is also useful for designing high temperature applications. DSC and TG-DTA thermograms of PS-3 and PS-4 are presented in Figs 3 and 4, respectively. The DSC and DTA transitions along with the initial decomposition temperature,
decomposition range, % weight loss and temperature of maximum weight loss \(T_{\text{max}}\) data for PS-3 and PS-4 are reported in Table 5 along with PS-1\textsuperscript{10} and PS-2\textsuperscript{7}. DSC thermograms showed \(T_g\) at 143.5°C and 134.5°C, respectively for PS-3 and PS-4 and is comparable with PS-1 (138°C) and PS-2 (141°C). The DSC exothermic transitions of PS-3 at 351.9°C and 400°C are due to some physical change and confirmed by no weight loss in TG curve over corresponding transition temperature range, while endothermic transition of PS-4 at 362.5°C confirmed decomposition reaction and it is supported by the weight loss over that transition range. Similarly exothermic transition of PS-3 at 407.7°C confirmed some physical change and other exothermic transition at 567.1°C confirmed degradation reaction. Exothermic DTA transitions of PS-4 at 378.3°C and 553.7°C supported degradation reactions.

PS-3 and PS-4 are thermally stable up to about 360°C and 340°C, respectively and involved two step decompositions. PS-3 and PS-4 have same thermal stability as that of PS-1\textsuperscript{10} and PS-2\textsuperscript{7} (340°C). Thus, thermal stability is slightly decreased due to methyl group. The first step decomposition of PS-3 involved 58.9% weight loss, while that of PS-4 involved 45.2%. The \(T_{\text{max}}\) of PS-3 for both the steps is higher by about 20°C than PS-4. Very small amount of residual weights indicated that both the polymers ultimately decomposed into low molecular weight substances. Thus, thermal stability of PS-1 to PS-4 is in the range of other polysulfonates\textsuperscript{9,11-14} containing other disulfonate linkages. From thermal data of studied polysulfonates of different chemical constitution indicated very little variation in thermal stability.

Sulfonate linkage in the main chain and methyl and chlorine side substituents are weak points, which degrade selectively forming free radicals. These radicals may further recombine to form new compound(s), which degrade at elevated temperature. The formation of new compound(s) is supported by DSC/DTA transitions and is further confirmed by no weight loss in TG curves over those transition ranges. Polymer degradation is a complex reaction and

<table>
<thead>
<tr>
<th>Polym.</th>
<th>DSC</th>
<th>DTA</th>
<th>TGA</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>(T_g)</td>
<td>Transition Temp., °C</td>
<td>Transition Temp., °C</td>
</tr>
<tr>
<td>PS-1\textsuperscript{10}</td>
<td>125</td>
<td>382 (exo)</td>
<td>125</td>
</tr>
<tr>
<td></td>
<td></td>
<td>292.9 (exo)</td>
<td></td>
</tr>
<tr>
<td>PS-2\textsuperscript{7}</td>
<td>140</td>
<td>360 (exo)</td>
<td>141</td>
</tr>
<tr>
<td></td>
<td></td>
<td>351.9 (exo)</td>
<td>340</td>
</tr>
<tr>
<td>PS-3</td>
<td>143.5 (endo)</td>
<td>400 (exo)</td>
<td>360</td>
</tr>
<tr>
<td></td>
<td></td>
<td>378.3 (exo)</td>
<td></td>
</tr>
<tr>
<td>PS-4</td>
<td>134.5 (endo)</td>
<td>362.5 (endo)</td>
<td>340</td>
</tr>
<tr>
<td></td>
<td></td>
<td>553.7 (exo)</td>
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</tbody>
</table>

Fig. 4—TG-DTA thermograms of PS-3 and PS-4 at heating rate of 15°C/min in an N\textsubscript{2} atmosphere
involves a variety of reactions such as cross-linking, branching, rearrangement and decomposition of segments.

Conclusions

On the basis of the experimental observations, it is concluded that PS-3 and PS-4 possess excellent solubility in common solvents, hydrolytic stability towards water, acids, alkalis, and salt solutions, moderate tensile strength, excellent electrical properties and good thermal stability. No change in tensile strength is observed but volume resistivity has increased 1.5 to 54.2 times, while electric strength has increased 2.5 to 3.5 times upon introducing methyl and chlorine groups alone or in combination. The improvement in volume resistivity and dielectric constant is due to cancellation of partial charges present on the polymer molecules.

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