

Thermal Degradation Studies of Perchlorate-Doped Conductive Polymers

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SYNOPSIS

The thermogravimetric behavior of polypyrrole (PPY), polybithiophene (PBT), and polyaniline (PAN) perchlorate complexes and their corresponding base polymers have been studied. The PBT-perchlorate complex and base polymer decompose at significantly lower temperatures than do the PPY and PAN counterparts. All three perchlorate complexes retain half or more of their original conductivities with no changes in the doping levels after one cycle of heating to 150°C in air and cooling to room temperature. However, after heating at 150°C for 24 h, only the PAN-perchlorate complex shows no significant change in the doping level, although its conductivity decreases by more than two orders of magnitude. X-ray photoelectron spectroscopy analyses of the perchlorate complexes reveal the presence of at least two distinct chlorine species. The thermal decomposition of the perchlorate anions results in the formation of volatile chlorine species as well in chlorine covalently bonded to the polymer. The thermal decomposition of the PPY-perchlorate complex also results in the conversion of positively charged nitrogens to iminelike structures.

INTRODUCTION

Among the electrically conducting polymers, polyaniline and the polyheterocycles, polypyrrole and polythiophene in particular, have attracted considerable attention because of their high conductivities and greater environmental stability as compared to polyacetylene. Highly conductive films of polypyrrole, polyaniline, and polythiophene have been prepared by electrochemical polymerization and oxidation of the corresponding monomer with various counterions incorporated into the polymer.¹⁻⁵ Alternatively, various chemical methods have also been developed for synthesizing these conducting polymers using appropriate oxidants, for example, FeCl₃ (Ref. 6) and halogens (Ref. 7) for pyrrole, (NH₄)₂S₂O₈ (Ref. 8) for aniline, and metal halogenide (Ref. 9) for thiophene. Recently, it has been reported that highly conducting polypyrrole, polyaniline, and polybithiophene can all be synthesized chemically using copper perchlorate as the oxidant.¹⁰⁻¹²

In this paper, we report on the thermal degradation of polypyrrole, polyaniline, and polybithiophene doped with perchlorate. X-ray photoelectron spectroscopy is used to study the structural changes of the polymers associated with the thermal degradation process. The use of a common dopant provides a good opportunity to see how the different polymers behave and interact with the dopant during thermal degradation. Furthermore, the thermal degradation behavior of these perchlorate-doped polymers can be compared with those of similar polymers doped with other oxidants to elucidate the role of the dopant.

EXPERIMENTAL

The polypyrrole, polybithiophene, and polyaniline perchlorate complexes were synthesized via the oxidative polymerization of pyrrole, 2,2'-bithiophene, and aniline, respectively, by copper perchlorate [Cu(ClO₄)₂·6H₂O] in acetonitrile according to the method of Inoue et al.¹⁰⁻¹² For the polybithiophene (PBT)-perchlorate complex, the Cu/S ratio in the reaction mixture was 1.5. The corresponding Cu/N

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ratios for the polypyrrole (PPY)-perchlorate and polyaniline (PAN)-perchlorate complexes were 1.5 and 3, respectively. These ratios have been shown by Inoue et al. to result in highly conductive complexes. The undoped base polymer was obtained by treating the complex with excess 0.5M NaOH for 24 h, followed by rinsing with deionized water and vacuum drying.

The weight changes of the complexes and the base polymers with temperature were monitored with a Netzsch STA409 simultaneous thermogravimetric (TG)-differential thermal analyzer (DTA) at a heating rate of 10°C/min in N₂. For each run, about 8 mg of sample was used with 100 mL/min of N₂ at approximately atmospheric pressure. The electrical conductivity (σ) of the complexes was also measured at various temperatures. For these measurements, the powdery polymer was compressed into pellets of 1.2 cm diameter and about 0.1 cm thickness and the standard collinear four-probe method was used. The four-probe assembly consists of a Teflon frame with platinum contacts. In determining the variation of σ with temperature, the ambient (air) temperature was slowly increased to 150°C and the σ was measured at regular intervals. The samples were then either left to cool in a desiccator or maintained at 150°C for 24 h before cooling to room temperature.

The structural changes associated with the thermal degradation of the complexes were studied by X-ray photoelectron spectroscopy (XPS). The XPS measurements were carried out on a VG ESCALAB MkII spectrometer with a MgK α X-ray source (1253.6 eV photons). The polymer samples in powder form were mounted onto standard sample studs by means of double-sided adhesive tape. The X-ray source was run at 12 kV and 10 mA. The pressure in the analysis chamber was maintained at 10⁻⁸ mbar or lower during measurements. All binding energies were referenced to the C1s neutral carbon peak at 284.6 eV. In spectral deconvolution, the full-width at half-maximum (fwhm) of the Gaussian peak components was kept constant in a particular spectrum. Surface elemental stoichiometries were obtained from peak area ratios that had been corrected by experimentally determined instrumental sensitivity factors.

RESULTS AND DISCUSSION

Thermogravimetric Analysis

The thermogravimetric (TG) scans of PPY-perchlorate, PBT-perchlorate, and PAN-perchlorate complexes in N₂ are compared in Figure 1. The TG

scans of the PPY-perchlorate and PAN-perchlorate complexes are quite similar, with a small initial weight loss followed by a major rapid weight loss step commencing at about 250°C. Simultaneous TG-DTA scans indicate that the major weight loss step is exothermic in nature. This second weight loss step is attributed to the decomposition of the perchlorate anions (see later section). The PBT-perchlorate complex also shows a small initial weight loss and an exothermic second weight loss step. However, the second weight loss step commences at about 150°C, which is substantially lower than the temperature at the onset of the second weight loss step of PPY-perchlorate and PAN-perchlorate complexes. Earlier studies¹⁰⁻¹² have indicated that these three perchlorate complexes are hydrated, and hence, the initial weight loss is probably due to the loss of water. The weight loss from the PBT-perchlorate complex in the second weight loss step from 150 to 400°C is less than those of the other two

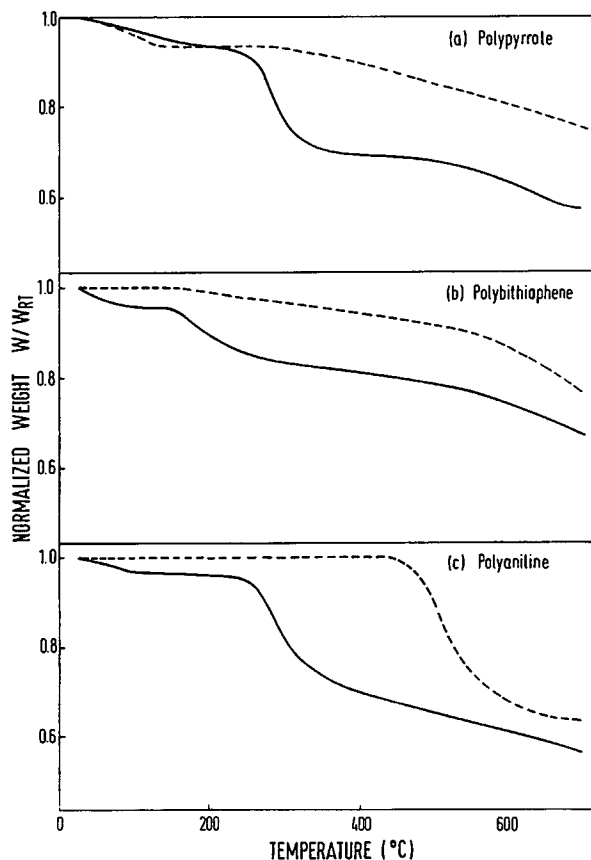


Figure 1 Thermogravimetric scans of (a) polypyrrole, (b) polybithiophene, and (c) polyaniline in N₂. Solid lines are for polymer-perchlorate complexes. Dashed lines are for the base polymers. W_{RT} is the weight at room temperature.

complexes. This is consistent with the fact that the PBT-perchlorate complex has a significantly lower weight fraction of ClO_4^- anions (see later section).

The thermal stability of the perchlorate-doped complexes should also be compared with that of the corresponding halogen-doped complexes. The TG scans of chemically synthesized PPY- Cl_2 , PPY- Br_2 , and PPY- I_2 in N_2 indicate the onset of major weight loss at 135, 140, and 184°C, respectively.¹³ These temperatures are substantially lower than that at the onset of major weight loss of the PPY-perchlorate complex. Thus, the anion has a strong effect on the thermal degradation behavior of the PPY complex. In contrast, the PAN-HCl complex commonly synthesized via the oxidative polymerization of aniline by $(\text{NH}_4)_2\text{S}_2\text{O}_8$ in aqueous HCl shows a TG behavior in N_2 that is quite similar to that of PAN-perchlorate. In the former, there is also an initial weight loss attributable to the loss of water and a second weight loss at 225°C attributable to the loss of the chlorine anions as volatile species.¹⁴ However, XPS analysis indicates that the chloride anions in PAN-HCl undergo reactions with the polymer in air, resulting in covalent chlorine species at 150°C before any significant weight loss is observed.¹⁴ In the case of PAN-perchlorate, XPS analysis shows no significant reaction of the ClO_4^- anions with the polymer in air at 150°C (see below).

In addition to the different temperatures at which the ClO_4^- anions in the complexes decompose, the polymer backbone may also exhibit varying degrees of thermal stability depending on the monomeric unit. The base polymers can be obtained by treating the perchlorate complexes with NaOH. The elemental analysis of the PBT base indicates that its structure can be represented by $\text{C}_{3.99}\text{H}_{1.91}\text{S}_{1.0}$. The corresponding representations of PPY base and PAN base are $\text{C}_{4.03}\text{H}_{2.68}\text{N}_{1.0}$ and $\text{C}_{6.08}\text{H}_{4.43}\text{N}_{1.0}$. The composition of the PBT base is close to that expected of a linear chain of disubstituted thiophene rings. In addition, the three elements C, H, and S account for over 97% of the mass, indicating minimal oxidation of the chain during synthesis and NaOH treatment. The composition of the PAN base corresponds well with that of emeraldine base with equal numbers of reduced (amine) and oxidized (imine) repeat units.⁸ The elemental analysis of the PPY base suggests that the NaOH treatment has resulted in deprotonation of some of the nitrogens. This is consistent with the XPS analysis that indicates the presence of iminelike structures in the PPY base polymer, but not in the pristine PPY-perchlorate complex. Thus, the PPY base does not truly represent a chain of pyrrole units.

The TG scans of the PPY, PBT, and PAN base polymers in N_2 are shown in Figure 1. In general, the base polymers show greater thermal stability than do their doped counterparts. However, the PBT base suffers from a slow but steady weight loss above 160°C. This temperature is only slightly higher than the temperature at which the PBT-perchlorate complex decomposes. Thus, it is possible that the decomposition of the ClO_4^- anions in this complex is initiated by structural changes in the polymer backbone since the ClO_4^- anions in PPY-perchlorate and PAN-perchlorate complexes do not decompose until about 250°C. The PPY base undergoes a small initial weight loss below 120°C, and the major weight loss commences above 300°C. The thermal stability of the PPY base can be improved if the base polymer is treated with phenylhydrazine to reduce the deprotonated nitrogens.¹⁵ Of the three base polymers, the PAN base appears to be the most stable for temperatures below 440°C. There is no significant weight loss from this polymer below this tempera-

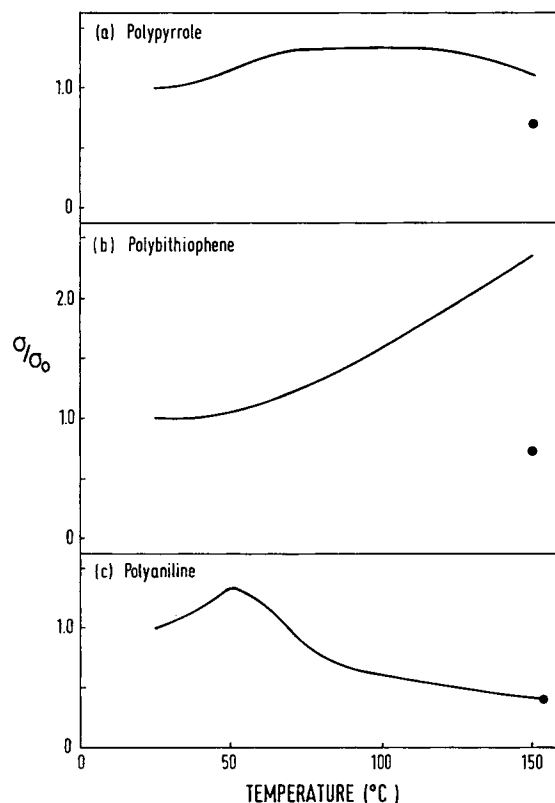


Figure 2 Effect of temperature on the electrical conductivity (σ) of (a) PPY-perchlorate, (b) PBT-perchlorate, and (c) PAN-perchlorate complexes. σ_0 is the conductivity prior to heat treatment. The symbol (\cdot) denotes the normalized conductivity at room temperature after cooling from 150°C.

ture. Thus, the equal numbers of reduced and oxidized repeat units in the PAN base appear to give rise to a polymer backbone that is less susceptible to thermal degradation than are those of the polyheterocycles, PPY and PBT.

Conductivity Changes

The σ 's of the PPY-perchlorate, PBT-perchlorate, and PAN-perchlorate complexes are 3, 0.5, and 1.5 S/cm, respectively. Of these three complexes, the PBT-perchlorate complex exhibits the highest degree of thermal activation when heated from room temperature to 150°C (Fig. 2). Although this complex begins to lose weight above 150°C, its σ increases continuously upon heating for temperatures below 150°C. Upon cooling the sample to room temperature in a desiccator, the σ is 0.72 of the original value (i.e., $\sigma/\sigma_0 = 0.72$). For electrochemically synthesized polythiophene-perchlorate, it has been reported that after one cycle of heating to 150°C and cooling to room temperature, the σ/σ_0 ratio has decreased to 10^{-2} .¹⁶ Electrochemically synthesized polythiophene-FeCl₄⁻ films exhibit excellent thermal stability as compared to polythiophene-perchlorate. The former films withstand 10 such cycles of heating and cooling with no significant loss of conductivity, and, also, no significant weight loss is detected during TG experiments at 150°C for 24 h.¹⁶ Thus, the method of synthesis and the choice of anions appear to have an important effect on the thermal stability of polythiophene. The σ of the PPY-perchlorate complex shows less variation with temperature from room temperature to 150°C than does that of the PBT-perchlorate complex. The former increases to 1.3 times the room temperature value upon heating to 70°C and is then constant to about 120°C before slowly decreasing. The room temperature σ/σ_0 after this heat treatment is similar to the corresponding value for PBT-perchlorate complex. Comparisons of the data in Figure 2(a) with those of chemically synthesized PPY-halogen complexes indicate that the conductivities of the latter complexes degrade rapidly above 70°C or 130°C, depending on the halogen dopant.¹³

Although the PAN-perchlorate complex possesses the highest thermogravimetric stability among the three complexes, its σ decreases at the lowest temperature (52°C) and the room temperature σ/σ_0 ratio attains the lowest value after the heat treatment. The temperature dependence of σ of the PAN-perchlorate complex [as shown in Fig. 2(c)] is very similar to that of PAN-HCl.¹⁷ The decrease in σ of PAN-HCl upon heating to 150°C

has been attributed to the initial loss of absorbed water vapor followed by the conversion of chloride anions to covalent chlorine.¹⁷ The loss of absorbed water vapor as indicated by the initial weight loss in the TG scan [Fig. 1(c)] probably also contributes to the decrease in σ of the PAN-perchlorate complexes. However, XPS analysis of the PAN-perchlorate complex after heating to 150°C shows no increase in the covalent chlorine content (see next section). Even after the PAN-perchlorate complex has been exposed to 150°C for 24 h, its perchlorate anion and covalent chlorine contents show only minor changes, although the room temperature σ/σ_0 ratio has decreased to 6×10^{-3} . In contrast, after a similar heat treatment, the composition of the PPY-perchlorate complex shows significant changes although the σ/σ_0 ratio is the same as that of PAN-perchlorate. The PBT-perchlorate complex shows the most drastic change in composition after being maintained at 150°C for 24 h, and this is also reflected in the decrease of the σ/σ_0 ratio to 10^{-4} .

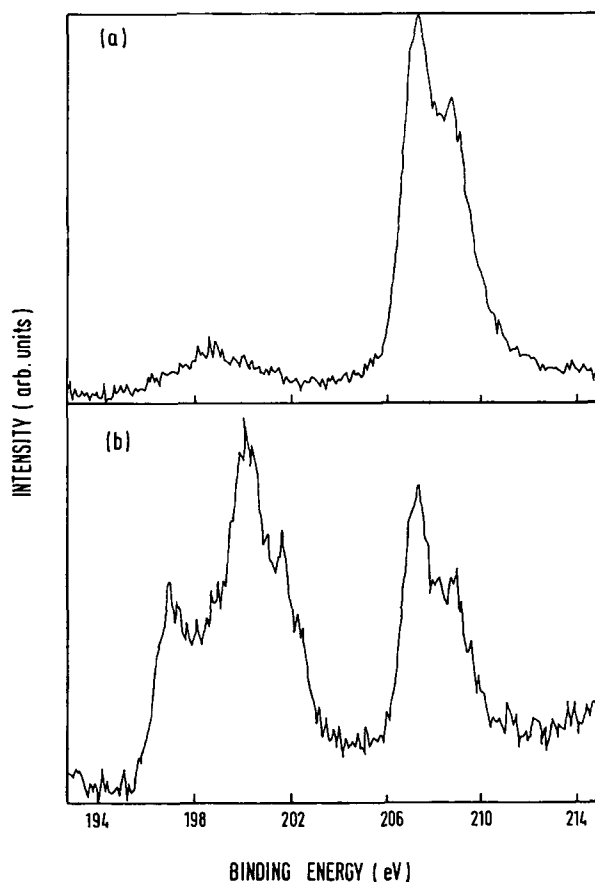


Figure 3 XPS Cl2p core-level spectrum of (a) pristine PPY-perchlorate complex and (b) PPY-perchlorate complex after heating to 260°C.

This drastic change in the composition of the PBT-perchlorate complex is consistent with the TG scan [Fig. 1 (b)], which indicates the onset of weight loss occurs at about 150°C.

XPS Analysis

The XPS Cl2p core-level spectra of the PPY-, PBT-, and PAN-perchlorate complexes indicate the presence of at least two distinct chlorine species. This is illustrated in Figure 3(a) by the two Cl2p envelopes of the PPY-perchlorate complex. The Cl2p component at the binding energy (BE) of 207.3 eV corresponds to that of the ClO₄⁻ anion.¹⁸ An earlier XPS study on PPY-Cl₂ (Ref. 19) indicated that chlorine anions would result in a Cl2p_{3/2} component at a BE of about 197 eV, whereas Cl covalently bonded to the pyrrole ring results in a Cl2p_{3/2} component at about 200 eV. Thus, the lower BE envelope shown in Figure 3(a) probably comprises the contribution from both partially ionic and covalent Cl species. Since the ClO₄⁻ anion shows a high reduction potential, it may undergo reactions to produce Cl⁻ ions,²⁰ which may then, in turn, react with the polymer, resulting in partially ionic and covalent chlorine. However, the X-ray satellite lines from the 207.3 eV component can also result in some contribution to the 200 eV envelope. The ratio of total Cl to N or S would not accurately reflect the degree of doping since some of the chlorine can exist as non-perchlorate species. Therefore, in the present work, the doping level is calculated from the ratio of the corrected XPS spectral areas of ClO₄⁻ and either N or S.

The ratios of the perchlorate anions to S and the nonperchlorate chlorine to S of the pristine PBT-perchlorate complex and the samples after heating to 150°C and after 24 h at 150°C are compared in Table I. In the pristine sample, the ClO₄⁻/S ratio of 0.18 is comparable to the anion to ring ratio of

Table I XPS Surface Stoichiometries of PBT-Perchlorate Complexes

Sample	Ratio of ^a	
	ClO ₄ ⁻ /S	Cl ^b /S
Pristine	0.18	0.02
After heating to 150°C	0.18	0.02
After 24 h at 150°C	0.03	0.05

^a Based on the corrected chlorine to sulfur core-level spectral area ratios.

^b Chlorine that does not exist as ClO₄⁻.

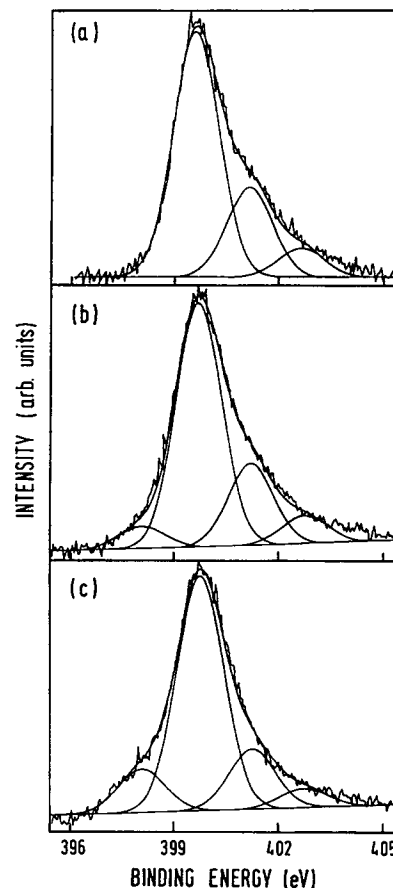


Figure 4 XPS N1s spectrum of (a) pristine PPY-perchlorate complex, (b) PPY-perchlorate complex after 24 h at 150°C, and (c) PPY-perchlorate complex after heating to 260°C.

1 : 7 reported for oxidized polybithiophene films grown electrochemically from solutions containing bithiophene and a perchlorate electrolyte.¹⁸ The XPS data indicate that when the PBT-perchlorate complex is heated to 150°C and then cooled to room temperature there is no change in the doping level. This is consistent with the σ of the heat-treated sample being about 0.7 of the pristine value. However, if the sample is maintained at 150°C for 24 h, the doping level as given by the ClO₄⁻/S ratio decreases to 0.03 and the amount of nonperchlorate chlorine shows a small increase. Thus, the XPS data confirms that the decomposition of the ClO₄⁻ anions occurs during the second weight loss step observed in the TG scan of the PBT-perchlorate complex. As expected, the decomposition of the ClO₄⁻ anions resulted in the loss of conductivity of the complex.

The N1s XPS core-level spectrum of the PPY-perchlorate complex is shown in Figure 4(a). It reveals the presence of a single major component at

Table II XPS Surface Stoichiometries of PPY-Perchlorate Complexes

Sample	Ratio of ^a		Proportion of		
	ClO ₄ ⁻ /N	Cl ^b /N	=N— ^c	—NH—	N ⁺
Pristine	0.38	0.05	—	0.68	0.32
After heating to 150°C	0.39	0.06	0.02	0.66	0.32
After 24 h at 150°C	0.27	0.09	0.06	0.65	0.29

^a Based on the corrected chlorine to nitrogen core-level spectral area ratios.

^b Chlorine that does not exist as ClO₄⁻.

^c Neutral imine structure.

about 399.7 eV, characteristic of the pyrrolylium nitrogen (—NH— structure) and a high BE tail attributable to the positively charged nitrogen.²¹ The proportions of nitrogen that exist as —NH— or amine structure and positively charged nitrogen (N⁺) are given in Table II together with the ratios of the two types (i.e., ClO₄⁻ and non-ClO₄⁻) of chlorine species to N. The ClO₄⁻/N ratio of the pristine PPY-perchlorate complex is similar to the value reported for electrochemically synthesized PPY-perchlorate complex.¹⁸ The N⁺/N ratio agrees quite well with the ClO₄⁻/N ratio, which is to be expected for charge neutrality to be maintained. As in the case of the PBT-perchlorate complex, heating the PPY-perchlorate complex to 150°C and then cooling it causes little change in the doping level of the complex and the σ after heat treatment is also still 70% of the pristine value. A very small amount of the amine structure is oxidized to the =N— or iminelike structure upon heat treatment. Increasing the period of heat treatment at 150°C for 24 h results in an increase in the imine structure. This is illustrated in Figure 4(b), which shows an imine component at about 398 eV that is absent in the spectrum of the pristine complex [Fig. 4(a)]. An increase in the temperature will result in a further increase in the imine-type structures and a decrease in the positive nitrogen [Fig. 4(c)]. When the PPY-perchlorate

complex is heated to 260°C, the fraction of nitrogen existing as imine-type structures increases to 0.12 while the N⁺/N ratio decreases to 0.23. The fraction of amine structures has not changed substantially. At this temperature, a significant fraction of the ClO₄⁻ anion has decomposed to volatile species and chlorine covalently bonded to the polymer. The XPS Cl2p spectrum of the PPY-perchlorate complex after being heated to 260°C is shown in Figure 3(b). The covalent chlorine component (at about 200 eV) is now even larger than is the ClO₄⁻ component (at 207.3 eV). It is possible that the decomposition of the ClO₄⁻ anions resulted in some of the nitrogen being doped by the oxygen from the decomposition process since the N⁺/N ratio is now substantially higher than the ClO₄⁻/N ratio, which is 0.07.

In contrast to the obvious decrease in the doping levels of the PPY-perchlorate and PBT-perchlorate complexes after 24 h at 150°C, the PAN-perchlorate complex shows no significant changes in the ClO₄⁻/N and N⁺/N ratios under similar conditions (Table III). The proportions of the different N species in Table III have been obtained from the deconvolution of the N1s spectra based on the following peak assignments: imine peak at 398.1 eV, amine peak at 399.3 eV, and positively charged nitrogen at BE > 401 eV.²² The Cl2p spectra of the PAN-perchlorate complexes are similar to that of the pristine

Table III XPS Surface Stoichiometries of PAN-Perchlorate Complexes

Sample	Ratio of ^a		Proportion of		
	ClO ₄ ⁻ /N	Cl ^b /N	=N— ^c	—NH—	N ⁺
Pristine	0.55	0.08	—	0.55	0.45
After heating to 150°C	0.53	0.07	—	0.56	0.44
After 24 h at 150°C	0.52	0.09	0.02	0.54	0.44

^{a,b,c} Refer to footnotes to Table II.

PPY-perchlorate complex [Fig. 3(a)], indicating the presence of at least two distinct species of chlorine. The thermal stability of the ClO_4^- anions in the PAN-perchlorate complex is also in marked contrast to the Cl^- anions in PAN-HCl that undergo reactions with the polymer in air at 150°C to form covalent chlorine species.¹⁴ However, the σ of the PAN-perchlorate complex apparently does not exhibit improved thermal stability over those of PPY-perchlorate and PAN-HCl complexes. Since the doping level of PAN-perchlorate is not altered by the heat treatment, it is likely that the number of charge carriers does not change either. However, the mobility of these charge carriers in the PAN-perchlorate complex may have been affected by structural changes in the complex that are not manifested in the surface stoichiometries as determined by XPS.

CONCLUSION

The thermal decomposition of PBT-perchlorate complex commences at a temperature that is substantially lower than those of PPY-perchlorate and PAN-perchlorate complexes and is comparable to the decomposition temperature of the PBT base polymer. The three perchlorate complexes do not undergo significant changes in the doping level after one cycle of heating to 150°C and then cooling to room temperature. The conductivity after this heat treatment is about half or more of the original value. However, after 24 h exposure to 150°C in air, only the PAN-perchlorate complex shows no significant signs of decomposition, but its conductivity has decreased by over two orders of magnitude. XPS analysis reveals that in the pristine complexes a small fraction of the chlorine may exist as partially ionic and covalent species. During the thermal decomposition process, the perchlorate anions undergo reactions resulting in both volatile chlorine species and chlorine that is covalently bonded to the polymer. Comparison of the stability studies of the PPY, PBT, and PAN perchlorate complexes with those of the corresponding polymers doped with other anions indicate that the anion plays a more prominent role in affecting the thermal stability of the PPY and PBT complexes than in the PAN complexes.

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