Abrasive wear performance of various poly amides

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Abstract

Polyamides (PAs) form a major class of tribo-polymers used in almost all types of wear situations. They excel most of the engineering polymers especially in the case of abrasive wear performance. Since methylene (-CH₂-) to amide (-CONH-) group ratio in PAs control the physical and mechanical properties, it was thought interesting to examine its influence on tribo-performance also. Hence, two types of PAs (addition and condensation) were selected for the present study. Abrasive wear studies on 10 PAs were done in a single pass condition by abrading a polymer pin against a waterproof silicon carbide (SiC) abrasive paper under various loads. It was observed that CH₂/CONH ratio had a significant influence on various mechanical properties such as tensile strength, elongation to break, fracture toughness, fracture energy and therefore, on the abrasive wear performance. It was observed that CH₂/CONH ratio and various mechanical properties when plotted individually did not show linear relation in most of the cases, while the specific wear rate as a function of some mechanical properties showed good correlation.

Keywords: Polyamide; Methylene to amide ratio; Abrasive wear; Structure-property correlation

1. Introduction

Polymers unlike other materials have two structural units such as molecular chains and molecular chain segments. Physico-mechanical properties of polymers such as glass transition temperature (T_g), intermolecular forces, conformation and the efficiency of molecular packing, etc. depend mainly on their chemical structure, length of the backbone, entanglement density of polymer chains, etc. [1,2]. In the case of polyamides (PAs), for example, the linear hydrogen bonding density increases with the increasing amount of amide groups. This restricts the segmental mobility of long chain aliphatic methylene groups in the polymer backbone and increases tensile strength [3] and the melting point [4,5].

A lot is reported on the performance evaluation of various polymers and composites in various wear modes. In fact, literature is flooded with the investigations on the influence of solid lubricants and fibres in various forms and/or their combinations on the tribo-performance of composites. Though the molecular weight, crystallinity, microstructure, etc. definitely affect the mechanical and tribo-properties of the polymer, very little is reported on these aspects (Table 1) [6-18]. All the same less efforts have been focussed on investigating the influence of basic chemical structure in a

class of polymers on the tribo-performance. This research area, still in its infancy needs to be explored for fundamental understanding of tribology of polymers.

Mori and Iwata [19] studied the effect of substituent chemical groups such as flexible alkyl chain derivatives and rigid cyanobiphenyl group on the tribological properties of the liquid crystal polymers (LCPs). It was observed that the flat structure of the biphenyl group played important role on the tribological properties of the LCPs. It was also observed that the introduction of alkoxy group into the cyanobiphenyl moiety increased the friction coefficient, while the substitution in the biphenyl group resulted in the destruction of the planar nature of the moiety, which proved to play very important role in deciding the tribological properties of the LCPs. Eiss and co-workers [20,21] studied the effect of different chemical groups in the backbones of the two types of polyimides (PIs) on the fatigue wear and friction. Type I PIs were developed from pyromellitic dianhydride (PMDA) while that of type II PIs were based on benzophenone tetracarboxylic dianhydride (BTDA). Backbone of type I was more flexible than that of type II and wear rates of type I were 10 times lower than the type II PIs. Type I PIs contained functional groups, viz. methylene, oxygen (highly flexible) and carbonyl (highly polar) while type II contained methylene and oxygen groups. It was found that the highly flexible polyimide with oxygen linkage showed less wear rate as compared to the other two rigid ones. The rigid polyimide

Table 1
Details of the literature survey on structure-property correlation

Serial no.	Resin	Property correlated	Conclusion from that study	Reference
1	PA 46, PA 66, PA 6	Microstructure	Crystallinity, water absorption and thermal properties affected the morphology of polyamides, which in turn affected the tribological properties of PAs also	[6,7]
2	PP	Spherulite size	Spherulite size has very little effect on the friction coefficient of PP	[8]
3	PTFE/graphite	Crystallinity	For neat PTFE the wear rate was directly proportional to the crystallinity	[9]
4	HDPE	Blending with PA 6	Blending of PA 6 reduced the reciprocating wear resistance of the HDPE	[10]
5	PA 6/GF	Morphology	Tribological properties of the composite were altered by manipulating the microstructure of the composite under different thermal histories	[11]
6	PA 12, PE	Interpenetrating network technique (IPN)	IPN improved the sliding wear characteristic of the base resins because of its inherent lubricating ability	[12]
7	Surgical grade UHMWPE	Microstructure	Alignment of crystalline lamellae of UHMWPE resulted the higher wear rate against CoCrWNI pins in bovine serum medium	[13]
8	PE, PP	Drawing ratio	Specific wear rate decreased with increasing drawing ratio	[14]
9	PEEK	Drawing ratio and molecular weight	Specific wear rate decreased with the increase in drawing ratio for all the three directions and with increasing molecular weight at higher speed and loads	[15]
9	PTFE	Spherulite size	Reduction in the specific wear rate at the lower spherulite size	[16]
10	PA 6	Molecular weight and shear viscosity	For low molecular weight annealed samples the specific wear rate decreased with increasing annealing temperature	[17]
11	PTFE/graphite	Microstructure	Concentration of graphite altered the microstructure of the composite which in turn affected the wear behaviour	[18]

with highly polar group led to increased interaction with the counterface which in turn, resulted in the higher traction and as a result higher wear rates. It was also observed that the incubation time was characteristic of the chemical structure of the PI and the wear rate was directly proportional to the rigidity of the molecular backbone. Thus, it was confirmed that lower the T_g , the rigidity of the polymer was lower too and hence, the wear and friction. Thus, rigidity, modulus of elasticity, elongation to break and rupture energy showed good correlations with tribo-properties. Thomas [22] studied the frictional properties of the films of a homologous series of poly(n-alkyl methacrylates) (PnAMA) by investigating their interface shear strength (τ). It was found that the molecular backbone became more rigid up to four carbon atoms. Further increase in the chain length reduced the activation energy for rotation which, in turn decreased the interfacial shear strength and consequently reduction in frictional force.

Polyamides form a major class of engineering tribo-polymers with a good combination of properties such as thermal, mechanical and tribological, along with the proces-

sibility (ease of moulding) and cost. They are of two types, viz. addition and condensation. By varying CH₂/CONH ratio, several PAs with varying properties have been synthesised. Influence of such ratio on physical properties such as density, melting point, glass transition temperature, etc. has been well documented [3,4]. Few of them have been investigated for their tribo-potential. The literature is available on the tribo-performance of various PAs, such as PA 6 [23], PA 66 [24], PA 46 [25], PA 11 [26], PA 12 [27], PA 1010 [28], aromatic PA [29], etc. They are widely used in various wear situations where moderate friction coefficient and high abrasion resistance as is required in the case of tyres. In fact, PAs are known to show very good abrasion resistance. In the recent work in the authors' laboratory, very interesting correlation emerged in the case of erosive wear of series of PAs and mechanical properties [30].

Hence, it was thought worthwhile to select the same series of both addition and condensation PAs and to examine their abrasive wear performance and correlation with mechanical properties, if any.

Table 2
Details of the selected materials

Property	Materials									
	Polyamide addition type				Polyamide condensation type					
	PA 6	PA 11	PA 12 (VE)	PA 12 (N)	PA 46	PA 66	PA 612	PA 66/610 (BM 20SBG)	Polyetheramide	Aromatic PA
Designation	A	B	C	D	E	F	G	H	I	J
Manufacturer	Du Pont (India) Ltd.	Elf Ato Chem, France	EMS Chemie, Switzerland	EMS Chemie, Switzerland	ERTA NV, Belgium	Du Pont (India) Ltd.	Du Pont (India) Ltd.	EMS Chemie, Switzerland	Elf Ato Chem, France	Elf Ato Chem, France
Trade name	Zytel	Rilsan	Grilamid L20	Grilamid L16 GM	Ertalon 46	Zytel	Zytel	Grilon BM 20SBG	PEBAX	Cristamid
CH2/CONH ratio	5	10	11	11	4	5	8	12	-	-
Specific gravity	1.15	1.05	1.01	1.01	1.18	1.14	1.06	1.09	1.15	1.06
Average crystallite size (Å)	233	204	126	103	-	156	204	182	-	-
Cohesive energy (kJ/mol)	62.5	85	89.5	89.5	116	125	152	268	-	-
Tensile strength (MPa) ASTM D 638	55.4	40.7	47	38.2	89	67	43	31	14	68
Elongation to break (%)	378	420	446	321	30	433	9	100	480	11
Flexural modulus (MPa) ASTM D 790	59	12	37	29	-	64	48	31	4	51
Notched impact strength (kgm/m) ASTM D 256	4.92	5.90	10.82	5.90	8	3.93	3.93	5.90	-	1.96
Shore D hardness	74	66	76	79	84	80	81	76	53	85
Glass transition temperature, T_g (°C)	50	46	46	46	78	57	-48	47	48	-
Melting temperature, T_m (°C)	222	179	177	178	287	258	215	200	161	230
Degradation temperature, T_d (°C)	440	440	455	450	-	430	-	460	-	410
Stress field intensity factor, K_{Ic} (MPam ^{1/2})	4.60	3.15	4.58	4.64	9.31	6.10	5.24	3.56	2.07	7.78
Fracture energy, $(\gamma_c \times 10^4 \text{ J/m}^2)$	5.37	5.89	3.90	4.43	9.29	6.52	3.78	3.90	7.97	6.60
Ductility factor (mm)	17.90	17.60	14.21	14.76	10.81	15.14	14.52	16.19	21.20	13.06
Fracture surface energy, γ ($\times 10^4 \text{ J/m}^2$)	2.69	2.95	1.95	2.22	4.65	3.26	1.89	1.95	3.99	3.30
Fracture stress, σ^* (MPa)	97.4	66.7	97	98.24	197.13	129.16	110.95	75.38	43.83	164.73
Critical crack length, C_c (µbm)	5140	5135	5144	5152	51.46	5141	5149	5146	5133	5144
Crack growth velocity, VQ (flm/s)	527	344	913	962	801	508.3	2077.2	557.7	24.8	1706.3
Time to failure under tensile stress, $t-p$ (s)	16.32	25.02	8.94	9.42	10.73	16.92	4.14	15.42	346.8	5.04

2. Experimental

2.1. Materials selected

Details of the selected poly amides are shown in Table 2. Among the two types of the PA 12 (Table 2), one was of medium viscosity grade and the other was of nucleated grade (as per supplier's information). The materials (except PA 46 and PA 612) were procured in the granular form. PA 612 and PA 46 were supplied in fibrous form and a moulded block form, respectively. The fibres were then chopped in small pieces prior to injection moulding. These granular and chopped pieces were dried in vacuum oven at 85 °C for 24 h before injection moulding into required shapes. These moulded materials were characterised for their thermal and mechanical properties (Table 2). Among these, cristamid was the only amorphous and transparent PA with aromatic groups in the molecular backbone while PEBAX was the only PA containing flexible ether group and was in rubbery form at room temperature. Crystallite size of the polyamides was measured by the X-ray diffraction (XRD) technique.

3. Abrasive wear studies

Abrasive wear studies were carried out on a single pin-on-disc machine, the schematic as shown in Fig. 1 and discussed elsewhere [31]. Prior to the experiment, polymer pin of size 10 mm x 10 mm x 4 mm was abraded against the waterproof 1200 grade (grit size $\cong 6$ (xm) silicon carbide (SiC) paper for uniform contact followed by cleaning with acetone, drying and weighing. The pin was then abraded in a single pass condition against 80 grade (grit size $\cong 175$ (xm) SiC paper fixed on a disc rotating with a constant linear speed of 0.05 m/s. After each traversal, new paper was fitted till the total abrading distance was 3.26 m. The applied

loads were in the range 4-10 N. The specific wear rate (K_0 , m^3/Nm) was calculated from the following equation:

$$K = \frac{AV}{Ld} \quad (1)$$

where AV is the wear volume (m^3), d the sliding distance (m) and L is the load (N). Each experiment was repeated for three times. The average was taken.

4. Results and discussion

Specific wear rate is plotted as a function of load in Fig. 2, while Fig. 3 indicates the relationship between the specific wear rate and the CH_2/CONH ratio. Specific wear rate as a function of various mechanical properties is shown in Fig. 4. Salient features of the present study are as follows.

- With the increase in load specific wear rates decreased almost linearly.
- The order of wear performance of the polyamides was as follows:

$$\begin{aligned} \text{PEA} &\gg \text{PA66} > \text{PA6} \cong \text{PA 11} > \text{PA 12 (MVE)} \\ &> \text{PA 12 (GPN)} > \text{PA66/610} > \text{PA612} \gg \text{PA46} \\ &\wedge > \text{aromatic PA (amorphous PA)} \end{aligned}$$

Polyetheramide (PEA), a polyamide with highly flexible ether linkage and showing rubbery properties even at room temperature, exhibited the best abrasive wear performance among all the polyamides. Whereas, an amorphous polyamide with aromatic groups showed the highest wear rate.

- CH_2/CONH ratio proved to be the influencing parameter for determining mechanical properties and abrasive wear as well.

Fig. 1. Schematic of single pin-on-disc machine.

Fig. 2. Specific wear rate as a function of load (counterface 80 grade SiC abrasive paper: 175 μm grit size; speed: 5 cm/s; distance abraded: 3.26 m in a single pass condition; A: PA 6; B: PA 11; C, D: PA 12; E: PA 46; F: PA 66; G: PA 612; H: PA 66/610; I: polyetheramide; J: aromatic PA).

As seen in Fig. 2, specific wear rates decreased almost linearly with load which is as per expected trends and mathematical equation reported in [32]. It was observed from Table 2 that all the physical or mechanical properties are not simple linear functions of CH_2/CONH ratio. In fact, in the case of polymers especially in the case of PAs, various other properties such as molecular weight, molecular packing, combination of repetition of odd and even number of methylene groups in between the two amide groups, etc. also contribute simultaneously towards these properties [33]. As seen from Table 2, physical and mechanical properties of PAs do not vary linearly with increase in CH_2/CONH ratio. This could be because of other factors such as molecular weight, processing conditions, etc. which were different for different PAs. As seen in Fig. 3, fairly good correlation was observed between specific wear rate and CH_2/CONH

Fig. 3. Specific wear rate of PAs as a function of CH_2/CONH ratio (counterface 80 grade SiC abrasive paper: 175 μm grit size; speed: 5 cm/s; distance abraded: 3.26 m in a single pass condition; A: PA 6; B: PA 11; C, D: PA 12; E: PA 46; F: PA 66; G: PA 612; H: PA 66/610).

ratio of PAs. Complete linearity in this relation cannot be expected since other properties such as crystallinity, molecular weight, etc. also influence the wear rate of PAs.

Abrasive wear of polymers and composites is widely studied and various types of correlations with physical [6–18] and mechanical [23,24,34–39] properties have been reported (Tables 1 and 3, respectively). Abrasive wear rates of the selected PAs as a function of the inverse of ductility factor (DF), stress necessary to initiate crack (a^*), inverse of fracture surface energy (γ), critical crack length (C^*), tensile modulus (E), crack growth velocity (VC), square of the fracture toughness (KC) and time to failure under tensile stress (TF), are shown in Fig. 4. K_0 versus S^{-1} or $(Se)^{-1}$ (Ratner-Lancaster plot) generally shows good linear correlation with abrasive wear behaviour [34], where S is the tensile strength and e is the elongation to break. However, in the present work, since these relations did not show any uniform trend, they were not included in Fig. 4. In the present case, amorphous polyamide (designated as J in Table 2) showed excessively high wear and its behaviour could not be well correlated with various properties as shown in Fig. 4. This might be attributed to the distinct difference in the chemical structure of the backbone. This was thought to be due to the presence of cycloaliphatic and aromatic groups in the backbone of this polymer. K_0 has shown fairly good linear behaviour with various properties as is clear in Fig. 4 for all other polyamides. From Fig. 4 it is also evident that wear performance, though showed fairly good linearity with various properties such as ductility factor (a), stress for crack initiation (b), fracture surface energy (c), critical crack length (d), tensile modulus (e), crack growth velocity (f), square of the fracture toughness (g) and the time to failure under tensile stress (h), while it cannot be treated as a function of single property or a simple combination of few properties. Abrasive wear is related to the combination of various mechanical properties in a complex manner.

The basic Archard equation relating hardness of metals to the wear resistance does not hold good in the case of polymers and composites, though a linear trend has been reported [2] in the case of some polymers/composites. Various theories have been developed and combinations of several properties are claimed to control the wear resistance of polymers and composites. Lhymn et al. [24] developed the following equation for the abrasive wear of 30wt.% carbon fibre reinforced composites of polyphenylene sulphide (PPS), polyetheretherketone (PEEK), polycarbonate (PC), polyethersulphone (PES), polyetherimide (PEI), PA 66 and GF reinforced PA 66 and PPS based on fracture properties.

$$W_s = \frac{k\mu}{EsL_s} \quad (2)$$

where W_s is specific wear rate, μ the friction coefficient, E the elastic modulus, ϵ the critical wear strain and L_s is the inter-laminar shear strength. Lhymn [32], for example, developed a model based on the crack propagation theory and classical mechanics confirming the following relationship in

Fig. 4. Correlation between the specific wear rate and various mechanical properties of the selected PAs (counterface 80 grade SiC abrasive paper: 175 μm grit size; speed: 5cm/s; distance abraded: 3.26 m in a single pass condition; A: PA 6; B: PA 11; C, D: PA 12; E: PA 46; F: PA 66; G: PA 612; H: PA 66/610; I: polyetheramide; J: aromatic PA).

the case of glass fibre (GF) reinforced PA 66, polyoxymethylene (POM) and polybutylene terephthalate (PBT).

$$W_s = \frac{V_s}{E H \epsilon \mu} \propto \frac{1}{F_N^\delta} \quad (3)$$

where W_s is the specific wear rate, V_s the sliding speed, E the elastic modulus, ϵ the critical wear strain, μ the friction coefficient, V_c the crack growth velocity and F_N is the normal load. This model held good at lower sliding speed and loads. In the further work, Friedrich and Cyffka [36] also developed a model based on the fracture behaviour of the glass fibre and glass sphere filled polyethylene terephthalate (PET)

composites. Specific wear rate showed inverse relationship with the fracture energy of the composites as follows:

$$W_s = \frac{k \mu^2 H_{abr}^{2/2}}{G_c} \quad (4)$$

where W_s is the specific wear rate, H_{abr} the hardness of the abrasive particles, μ the friction coefficient and G_c is the fracture energy of the composites. The present work also indicated that wear is related to the combination of various properties in a complex manner. Thus, the work indicated the trends in mechanical, physical and abrasive wear properties of several PAs as a function of CH2/CONH ratio

Table 3
Literature on mechanical property-wear correlation

Serial no.	Resin/derivatives of alkyl compounds	Property correlated	Conclusion from that study	Reference
1	PA, PTFE, PMMA, PP, HDPE, POM, PVC	Ploughing component of friction (i_p), tensile stress (S), elongation to break (e), hardness (H)	$Koaii/(Hse)$	[23]
2	CF composites of PA 66, PPS, PEEK, PC, PES, PEI; PA 66/GF, PPS	ILSS (I_L), modulus (E), friction (ji), critical wear strain (ϵ)	$Koafj_/(EeI)$	[24]
3	Lancaster	Product of Se	$K_p\alpha/(Se)$	[34]
4	PTFE and -y-irradiated PTFE	Product of S^e	$K_p\alpha/(S^e)$	[35]
5	PET and its composites with reinforced GF and glass sphere	Hardness (H), microfracture energy (GC), probability factor for microcracking (Q)	$K_p\alpha HS^2/G_c$	[36]
6	PA 6, POM, PE, PMMA, PC, PP, PTFE, PPO, PVC	Cohesive energy (E_{coh}) of the materials	$K_0\alpha/E_{coh}^{1/2}$	[37]
7	POM, PA 6	Surface free energy	$K0\alpha/(\text{velocity of oil spreading in the bushes})$	[38]
8	PMMA, PES	Fracture toughness (KIC)	$K_0\alpha/K_{1C}$	[39]

and correlation with various properties. A mathematical model is to be developed for indicating perfect linearity in wear-property correlation.

5. Conclusion

It was concluded from the on studies various PAs from two classes, viz. addition and condensation that CH₂/CONH ratio significantly influenced the mechanical and physical properties of the PAs. However, a complete linear correlation was not observed because other physical properties such as crystallinity, molecular weight, microstructure, etc. also contribute towards controlling the mechanical properties. Specific wear rates (abrasive wear mode) showed fairly good correlation with various mechanical properties such as ductility factor, stress for crack initiation, fracture surface energy, critical crack length, tensile modulus, crack growth velocity and the time to failure under tensile stress, etc. Ratner-Lancaster plots, however, did not show good linearity. It was also concluded that abrasive wear of PAs is not controlled by a single material property. Combination of properties in a complex manner may show fairly good correlation.

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