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ORIGINAL RESEARCH

Wear response of glass fiber and ceramic tile-reinforced hybrid epoxy matrix composites

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Abstract
This study presents the tribological behavior of epoxy matrix composites containing two different fillers. The composites contain fillers with different particle sizes (< 90 µm and in the range of 150–300 µm) and different wall tile to glass fiber waste ratios (55:5, 50:10 and 40:20). The total amount of filler was fixed to 60 wt% in all of the composites to reveal the effect of variations in filler content on properties. The physico-mechanical properties, such as bulk density, porosity, impact resistance, shore hardness, flexural strength and wear characteristics of developed materials were determined. Tribological tests were carried out by ball-on-disk configuration and rotational sliding at room temperature against 6-Co/WC ball with a diameter of 3 mm. 3N of constant test load was applied and the wear distance was kept as 400 m. The results showed that the steady-state coefficient of frictions was changed in the range 0.38–0.47 and the wear rate was obtained between 1.74×10^{-5} and 1.09×10^{-4} mm^3/Nm. When the coarser particle size filler was used, it resulted in worsening of the wear resistance. It can generally be concluded that the properties were improved with the increasing amount of wall tile addition, and also better properties were obtained in both types of filled composites compared to the epoxy matrix.

Keywords Hybrid composites · Epoxy · Fabrication · Ceramics · Mechanical properties · Tribology

Introduction

Polymer matrix composites serve several advantages such as combination of light weight, high specific modulus and strength, impact resistance, etc., over metal equivalents [1]. Hybrid composites are one of the class of composites which are made by either combining two or more different types of reinforcements or at least two different matrix types. They offer a range of properties that cannot be achieved with a single reinforcement [2]. Moreover, a balance in cost and performance can be achieved with hybrid composites.

More importantly, in terms of waste usage, which has become one of the most important issues in recent times, waste particles can be used as reinforcement/filler in hybrid polymer matrix composites [3, 4]. By this way, environmental pollution can be eliminated and the value-added products can be produced. For example, a gigantic quantity of boron [5], marble [6], tile [7], glass fiber [8–10] waste, etc., is generated in Turkey, recently. Hence, nowadays, the studies on the reuse and regain of these kinds of waste materials for different applications have been started to become one of the most important topics of research. For example, ceramic wastes which obtained from tile production have been started to use as additive for porcelain stoneware body mixture [7]. The utilization of boron wastes in ceramic and construction industries is one of the possible ways for the recycle of boron wastes. Karasahin and Terzi investigated the usage of marble wastes as a filler in the asphalt mixtures [6]. Kavas studied the usability of boron wastes as a fluxing agent in brick production [11]. Christogerou et al. investigated use of boron wastes in the production of heavy clay ceramics [12]. Kavas and co-workers studied the formation of artificial light-weight aggregates from boron-containing wastes [13]. Özdemir and Öztürk examined utilization of clay wastes containing boron as cement additives [14]. Celik studied recycling of boron waste for the production of ceramic wall tile [15]. Ercenk and co-workers studied the influence of boron waste addition on the

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properties of glass and glass–ceramics [16]. Uslu and Arol studied the use of boron waste as an additive in red bricks [17]. Ribeiro and co-workers studied addition of glass fiber-reinforced polymer waste materials into polyester material-based mortars, as a replacement for sand aggregates or filler [18]. Yaman and Calis Acikbas developed 60 wt% boron waste-containing epoxy matrix composites, and investigated the effect of boron waste addition and the particle size of boron waste on the some properties [19]. Calis Acikbas and Acikbas studied the use of thermoset industrial toilet seat wastes as reinforcement for the production of polymer composite [20]. Thomas and co-workers used carbon powder wastes in epoxy matrix for enhancing mechanical properties [21]. Acikbas and Gocmez studied the effect of industrial sanitary ware waste content on the properties of polyester matrix composite [22, 23]. Uygunoglu et al. studied the properties of boron-added polymer matrix composites [24]. The studies show that these kinds of waste materials can be evaluated for the production of polymer matrix composites. By this way, cost-effective products can be produced and it may also help to decrease environmental pollution.

To the best of our knowledge, no study is available in the literature on the tribological behavior of the wall tile/glass fiber-reinforced hybrid epoxy matrix composites. Waste inventory of wall tile factories (Kale Ceramic Company, Turkey) involves 600 tons year^{-1} and for glass fiber (Şişecam Company, Turkey) 5000–25,000 tons year^{-1}. These waste quantities are quite high and they should be re-evaluated. Usage of these kinds of waste materials in composite production might provide enhanced tribological properties, allow recycling of wastes and obtain wide range of properties by reinforced hybrid composites. In our previous study, it was observed that incorporation of ceramic wastes (wall tile and glass fiber) enhances the mechanical properties of epoxy matrix [8]. However, in many applications, tribological behaviors of materials are very important for life time of product. Therefore, this study was conducted to investigate the wear and frictional behavior of hybrid polymer matrix composites used in solid stone, kitchen and laboratory counter-tops, machine parts, etc., where wear protection is required. The effect of filler particle size and the ratio of wall tile to glass fiber waste on the physico-mechanical properties were evaluated and the relationship between these properties with the tribological behavior was investigated.

Experimental

Materials

The starting materials for the production of hybrid composites was glass fiber waste (Şişecam Glass Fiber Company, Gebze, Turkey) and wall tile waste (Bien Ceramic Company, Bilecik, Turkey). Epox Acast 690, chosen as a matrix material, was procured from Smooth On Limited, Canada. The weight ratio of the epoxy and hardener was 73:27. The preparation and characterization of waste materials for filler are given elsewhere [8]. The true density of the glass fiber and wall tile powder was 2.61 and 2.54 g/cm^3, respectively. The values were determined by a Micromeritics Accupyc II 1340 model He-gas pycnometer.

Fabrication of hybrid composites

A total of six glass fiber/wall tile-reinforced hybrid epoxy composites were fabricated using casting technique. The prepared compositions are given in Table 1. A model (carton picture) representing hybrid composite of glass fiber and wall tile filler is given in Fig. 1, and fabrication steps are shown in Fig. 2. Epoxy:filler ratio was kept as constant for all compositions as a 40–60 by weight. In our previous study, the pourable mixture of epoxy to ceramic filler was found as 40:60 wt% [8]. Therefore, this ratio was used in this study. The coding was done as the first figure representing the particle sizes. F letter in the code means fine and the particle size is below 90 microns, and C letter in the code means coarse particle size between 150 and 300 microns. The two figures following F and C represent the

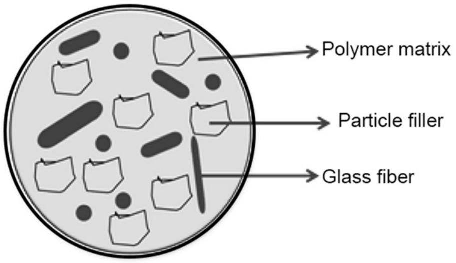


Fig. 1 A model (carton picture) representing hybrid composites

Table 1 Details of the fabricated hybrid epoxy matrix composites

	F5505	F5010	F4020	C5505	C5010	C4020
Epoxy	40	40	40	40	40	40
Wall tile waste	55	50	40	55	50	40
Glass fiber waste	5	10	20	5	10	20

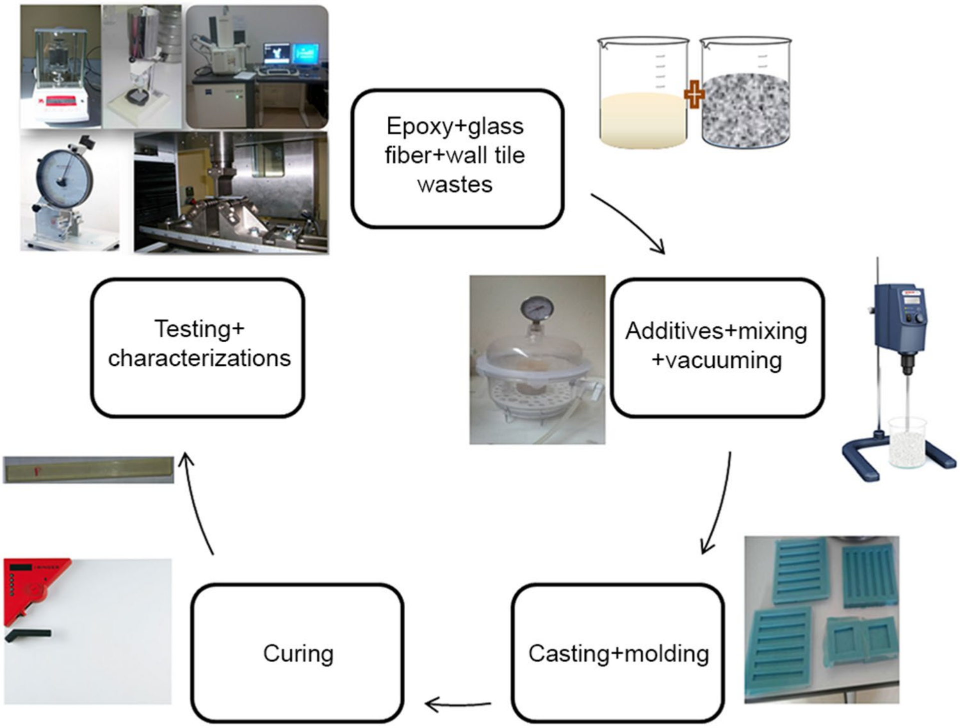


Fig. 2 Fabrication steps of hybrid composites

wall tile waste (WT) content and the last two figures represent the glass fiber (GF) amount.

The fabrication process of WT–GF waste particles–epoxy composites can be summarized as follows: The silicon mold was treated by a release agent polyvinyl alcohol (PVA) and after the drying process, the silicon mold was ready for casting. Epoxy resin, hardener and waste particles were blended in a propeller mixer at 500, 1000 and 1500 rpm for 5 min at each speed. Next, vacuum application was applied to avoid bubble formation so that porosity evolution was eliminated. The composite compositions were cast into silicone molds and left to rest for 1 day at 25 °C to complete the curing process. After that the hybrid composite samples were removed from the molds.

Density and porosity measurements

The Archimedes principle was used to determine bulk density (B.D.). The hybrid composite samples were weighed by an analytical balance to determine the bulk density values:

$$\text{Bulk density} = \frac{W_1}{W_3 - W_2}\rho_{\text{water}}. \quad (1)$$

In this equation, W_1 is the dry weight, W_2 is the wet weight suspended in water, and W_3 is the wet weight. Theoretical density (T.D.) of hybrid composites was calculated from the volume fractions and the theoretical densities of the constituting materials. The percentage theoretical density and percentage total porosity (T.P.) were calculated by

$$\text{T.D.}(\%) = \frac{\text{B.D.}}{\text{T.D.}} \times 100, \quad (2)$$

$$\text{Total Porosity } (\%) = 100 - \text{ T.D. } (\%). \quad (3)$$

Mechanical tests

The hardness of samples was measured by a Shoremeter with a dimension of 5 × 5 cm. TS-985 EN ISO 178 standard was used for the determination of three-point-bending strength of composites with 80 × 10 × 4 mm ($L \times W \times T$) dimension and bending modulus was also calculated. ISO EN 180 U standard was used for the determination of impact resistance of developed composites. The samples were in 80 × 10 × 4 mm ($L \times W \times T$) sizes. For each test, at least three measurements were performed to obtain accurate results.

Tribological tests

All composites were polished to achieve an average surface roughness (Ra) of 0.1 µm. For the investigation of the sliding wear behavior of hybrid composites, ball-on-disk wear tests (CSM tribometer, Switzerland) were carried out by applying a constant normal 3N load in the contact while rotating the ball at constant speed of 30 cm s^{-1}. The friction force was continuously measured and stored during the test. Wear contact occurred at normal room temperature and humidity in the laboratory. All the experiments were run for 400-m distance. A tungsten carbide ball with 3 mm diameter was attached to the testing machine sample holder. Speed, time and load were kept constant throughout for all the experiments. The following is a common equation used to compute the wear rate:

$$W = V/(D \times L). \tag{4}$$

Here, W represents the wear rate, V the wear volume, D the sliding distance, and L is the load.

Microstructural investigations

After wear tests, SEM (Jeol JSM 5600 model) equipped with an energy-dispersive spectroscope (EDS) was used to examine the morphology and analysis of the wear characteristics occurred in the wear debris. The analysis was carried out at 20 kV.

Results and discussion

Density and porosity content

The bulk density, theoretical density, total porosity (%) and theoretical density (%) results of hybrid composites are given in Figs. 3 and 4. The bulk density values are almost similar and changing between 1.54 and 1.60 g/cm^3. The composites containing finer particles show a little bit higher bulk density values (Fig. 3). Increase in GF waste content leads to lower bulk density and higher porosity content, and hence, shows lower theoretical densities. Therefore, total porosity (%) values decrease and theoretical density (%) increases with increase in bulk density (Fig. 4). Since theoretical densities of the glass fiber and wall tile powder were very similar (GF: 2.61 g cm^{-3} and WT: 2.54 g cm^{-3}), change in WT/GF ratio did not cause much difference in theoretical density of hybrid composites.

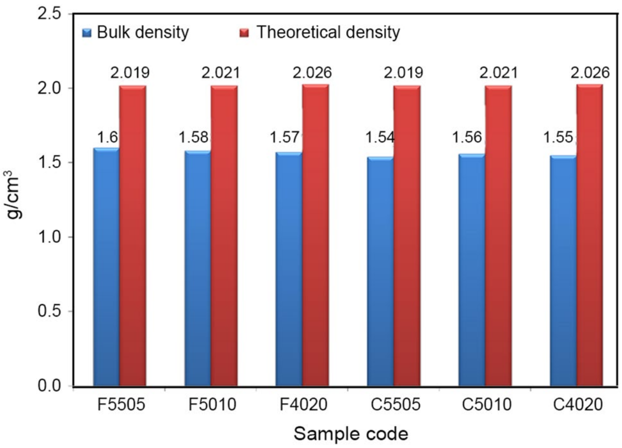


Fig. 3 Bulk density and theoretical density of fabricated hybrid composites

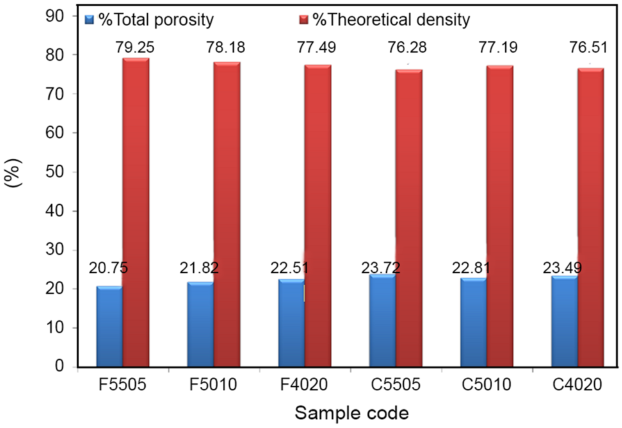


Fig. 4 Total porosity and theoretical density (%) of fabricated hybrid composites

Mechanical properties

Table 2 shows mechanical and physical properties of samples and their correlation with tribological behavior. The neat epoxy sample has the highest T.D.(%) value. Addition of fillers into epoxy provides more porous structure due to formation of hard mixable mixture with increased viscosity. The flexural strength of neat epoxy is the highest one due to plastic behavior. Addition of GF and WT hard ceramic fillers into epoxy resin results in decreasing flexural strength values due to more brittleness structure. Neat epoxy has the lowest flexural modulus and hardness as expected. Addition of hard particles enhances the flexural modulus as well as hardness. Addition of GF and WT particles decreases the impact resistance. Hard GF and WT particles have positive effect on wear rate since they carry load and assist in strengthening the surface which will result in enhancing the tribological behavior of the polymer. The results indicated that the composites reinforced with fine particle size showed better wear resistance related to enhanced mechanical properties. Increase in glass fiber content in composites leads to poor mechanical properties and results in lower wear resistance. The highest wear rate has been obtained for C4020, even higher than that for the neat epoxy, which contains the highest glass fiber content and particle size.

Table 2 Mechanical, physical and tribological properties of developed composites

	μ	T.D. (%)	Flexural strength (MPa)	Flexural modulus (GPa)	Shore D hardness	Impact resistance (kJ mm^{-2})	Wear rate (mm^3 Nm^{-1})
Epoxy	0.298	98.90	106.00	3.20	81.00	12.90	6.70080E−05
F5505	0.424	79.25	85.80	4.94	94.50	11.00	1.73515E−05
F5010	0.425	78.18	84.05	4.88	93.00	10.55	2.44583E−05
F4020	0.447	77.49	84.35	4.50	92.13	10.35	2.50084E−05
C5505	0.458	76.28	70.40	3.90	90.33	7.25	3.85607E−05
C5010	0.466	77.19	64.80	3.73	89.50	7.00	5.12650E−05
C4020	0.476	76.51	61.90	3.67	88.33	6.90	1.09000E−04

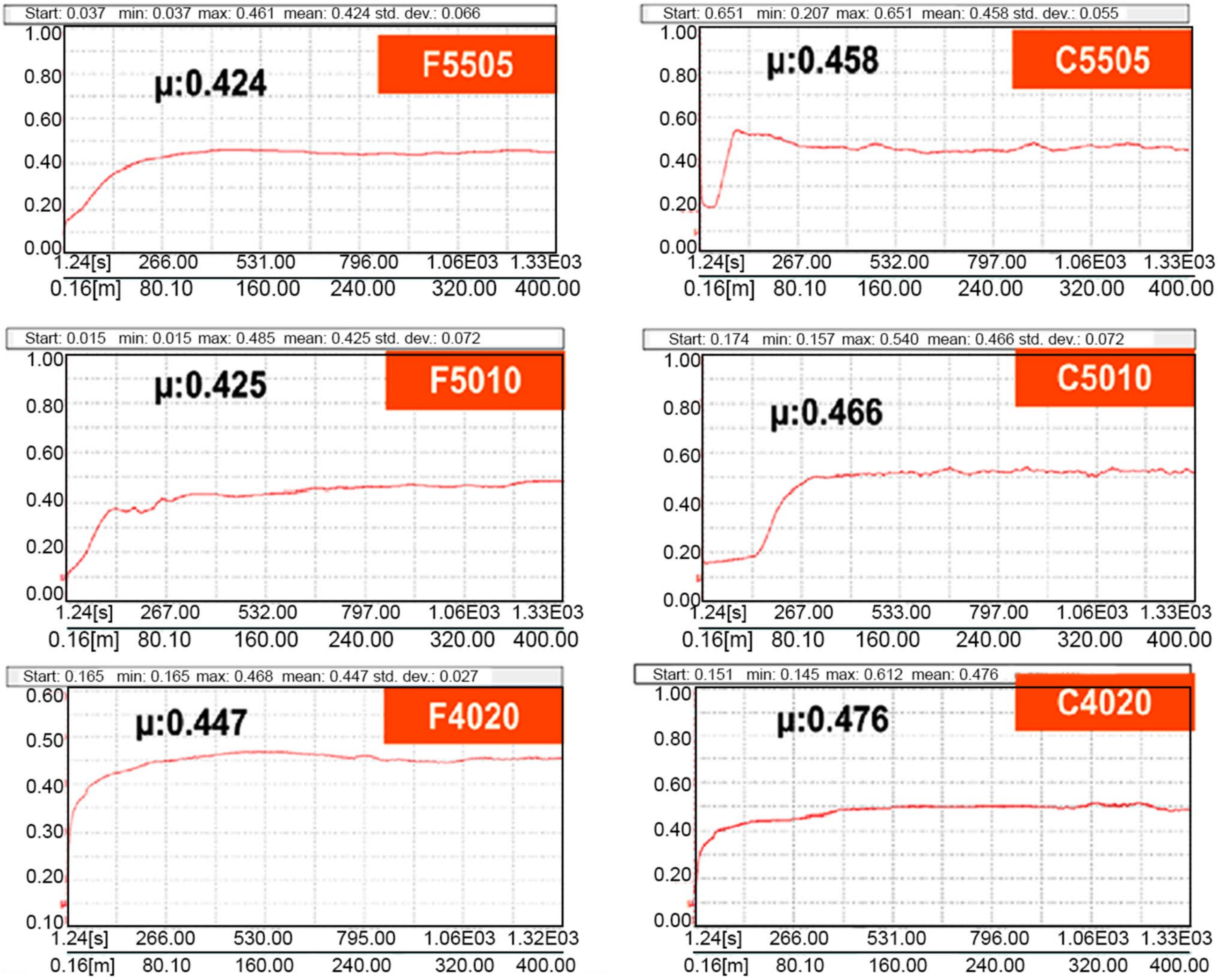


Fig. 5 Coefficient of friction graphics of produced hybrid polymer matrix composites

Tribological behavior

The coefficient of friction (CoF) graphics of produced hybrid polymer matrix composites are given in Fig. 5. The composites containing coarse particle size show higher friction coefficient and increasing glass fiber content which causes an increase in friction coefficient. This frictional behavior of these samples is in correspondence with wear rate results. Higher coefficient of friction leads to higher wear rates (Table 2). In comparison with the porosity and theoritical density (%) values (Figs. 3, 4), and wear properties, the rate of wear is increased in the samples with increasing porosity and decreasing density. In F4020 and C4020 composites, higher wear rates were achieved in comparison with the other samples of the same group (Table 2). It can be concluded that reducing the amount of porosity and increasing the density may lead to an increase in wear resistance in these composites. As it can be seen from Table 2, for group *F* samples as the hardness decreases, wear rates increase. Same tendency has been observed in the C group samples. It can be concluded that wear values have increased as the hardness values are decreased in both group of samples. This trend is often found in the literature. For example, Gore and Gates have investigated the effect of hardness on three different forms of wear in detail and the reported results show that by increasing hardness, wear is decreased [25]. When this is compared with the impact tests results, it is demonstrated that as the impact resistance is increased, wear resistance is also increased in both group of materials.

It is well known that the wear mechanism can be defined as the combination of physical, chemical and mechanical interactions that occur during wear contact. In general, friction and wear behavior of materials depend on wear mechanisms generated by many factors, such as load, sliding speed, and mechanical properties. Both SEM and EDX analyses have been carried out to understand the wear mechanisms of hybrid composites generated under the same test conditions as given in the experimental procedure section (Figs. 6, 7). In our previous study [19], tribological behavior of neat epoxy was investigated. It was found that tribochemical layer, which contained carbon and oxygen, was responsible for the low friction and reduced wear by protecting the composite surface and by providing a very smooth friction surface. Unfortunately, besides the tribochemical layer formation, deep wear scars were seen, which showed that the abrasive wear occurred on the surface of neat epoxy. According to microstructural characterization of worn hybrid composites, abrasive and adhesive wear, and the formation of tribochemical layer were also seen as those seen in neat epoxy. In all of the composites, it was detected that carbon (C), oxygen (O), silicon (Si), aluminum (Al), tungsten (W) and cobalt (Co) were as the main elements of the tribolayer. As can be seen in Fig. 6, the uniformity and thickness of this layer in fine particle-sized composites is increased with increasing filler contents, and the addition of more rigid particles into epoxy resin enhances the mechanical properties, such as hardness and modulus which might lead to higher abrasion resistance of composites than the epoxy matrix and coarser particle-sized samples. The high-strength surface layer (Fig. 6c) had the lowest wear rate among all composites.

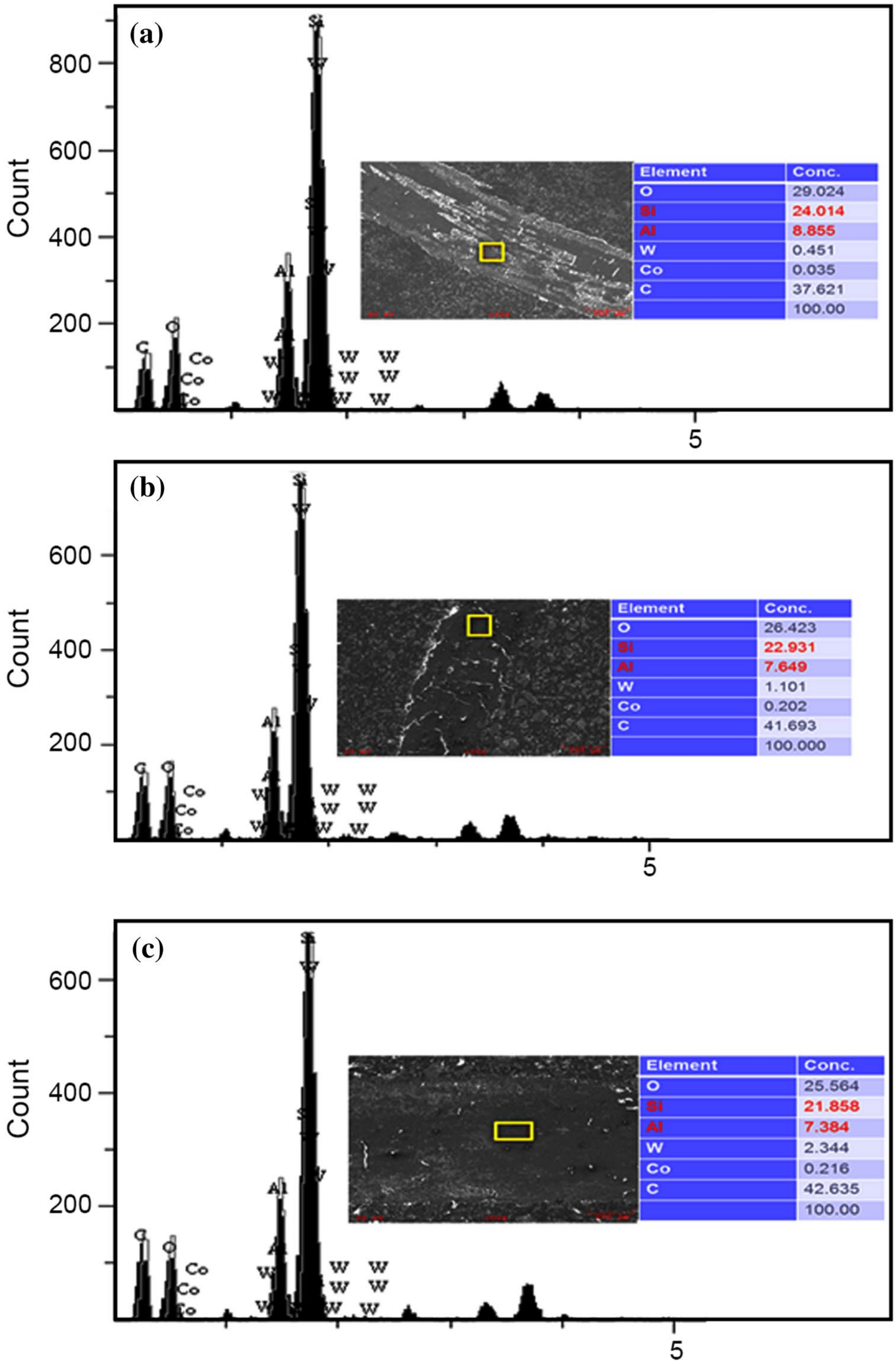
On the other hand, according to Fig. 7, in the coarse particle-sized composites, micro-cracks can be seen on the tribolayer and also the width of wear debris is larger than that in the fine particle-sized ones. The contact interference suffered from fragmentation of particles. Scars in composites containing 50 and 60 wt% fillers contained large amounts of wear debris, which formed a compressed layer over the track surface. Increase in particle size and increase in content of fillers might act as precursor points to initiate crack formation and propagation. Debonding between the fiber and matrix related to thermo-mechanical loading may enlarge the wear in these composites. Consequently, dominating wear mechanisms can be summarized as adhesive and abrasive wear, and formation of tribochemical layer under selected operating conditions.

Although no study is found in the field of wear properties of wall tile/glass fiber-reinforced hybrid epoxy matrix composites, when these results are compared to the wear behavior of glass fiber-added epoxy composites and neat epoxy in the literature, they seem consistent with our results. Agrawal et al. studied wear and friction characteristics of glass fiber-reinforced epoxy resin under three different sliding environments (dry sliding, oil-lubricated sliding and sliding in argon) under 40–120 N loads by pin-on-disk configuration. They reported that for epoxy matrix sample becomes softer with rising temperature during the test, and so, greater wear rate is obtained. In the case of glass fiber-added epoxy composites, glass fibers embedded to the matrix were separated more easily according to this thermal-softening behavior of matrix, and hence, wear amount was increased [26]. Kishore et al. compared the wear properties of the two different levels of graphite-bearing glass–epoxy (G–E) systems by pin-on-disk machine under 20-kg load cell and concluded that the tribological response of the glass–epoxy composite system is dependent on the filler type and its amount which affect the bonding strength between the fiber and matrix [27].

Conclusion

In this study, wear response of hybrid epoxy matrix composites containing two different fillers (wall tile and glass fiber waste) with different particle sizes (< 90 µm and in the range of 150–300 µm) and different wall tile to glass fiber waste ratios were evaluated. The composites reinforced with fine particle size showed better wear resistance which was related to improved mechanical properties. Increase in glass fiber content in composites led to poor mechanical properties and resulted in lower wear resistance. Coarse particle size-containing composites showed higher friction coefficient and increasing content of glass fiber caused increase in the friction coefficient. Dominating wear mechanisms can be summarized as

Fig. 6 SEM micrographs and EDX analysis results of the worn surfaces of **a** F5505, **b** F5010, **c** F4020



(a)

Element	Conc.
O	29.024
Si	24.014
Al	8.855
W	0.451
Co	0.035
C	37.621
	100.00

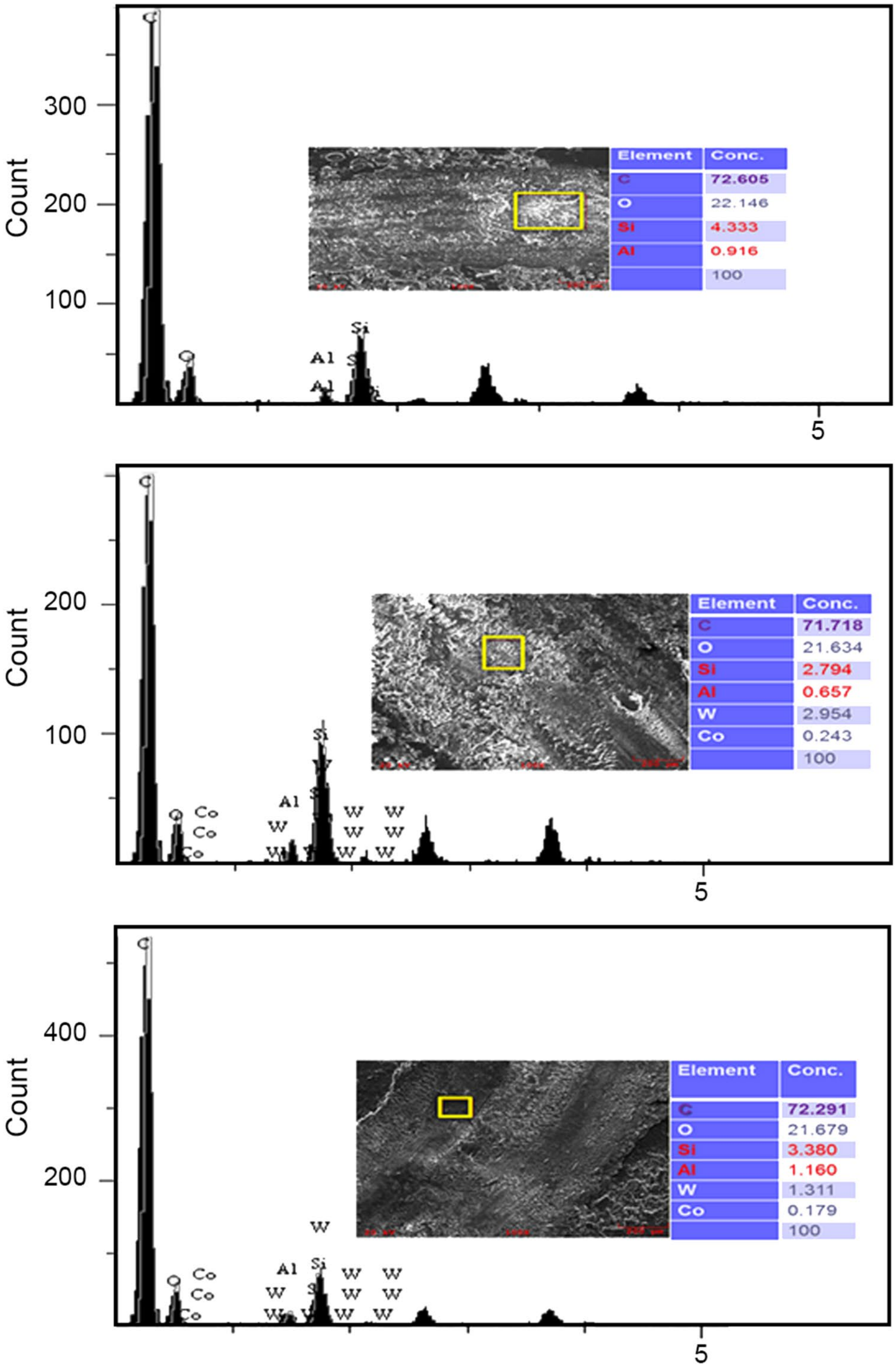
(b)

Element	Conc.
O	26.423
Si	22.931
Al	7.649
W	1.101
Co	0.202
C	41.693
	100.000

(c)

Element	Conc.
O	25.564
Si	21.858
Al	7.384
W	2.344
Co	0.216
C	42.635
	100.00

adhesive and abrasive wear, and formation of tribochemical layer under selected operating conditions. The results revealed that the addition of porcelain waste and glass fiber is an economic and effective method to improve mechanical and tribological properties utilizing ceramics industry wastes.



Element	Conc.
C	72.605
O	22.146
Si	4.333
Al	0.916
	100

Element	Conc.
C	71.718
O	21.634
Si	2.794
Al	0.657
W	2.954
Co	0.243
	100

Element	Conc.
C	72.291
O	21.679
Si	3.380
Al	1.160
W	1.311
Co	0.179
	100

Fig. 7 SEM micrographs and EDX analysis results of the worn surfaces of **a** C5505, **b** C5010, **c** C4020

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