Hybrid polymer composites with enhanced energy absorption

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Abstract: This paper presents the influence of the type and structure of reinforcement, on the epoxy resin matrix polymer composites mechanical and ballistic properties. Aramid, basalt, glass fabrics and their hybrid systems were used as reinforcement. Impact strength according to Izod and "falling arrowhead", flexural strength and structure of the obtained composites were tested. The specific gravity was also determined. The aramid-glass hybrid composites showed high flexural strength (397 MPa) and Young's modulus (21 GPa). However, aramid-basalt composites had high impact strength (116 kJ/m^2) and impact energy absorption (45 J).

Keywords: hybrid epoxy composites, reinforcement modification, aramid, glass and basalt fabrics, mechanical properties.

Hybrydowe kompozyty polimerowe o zwiększonej absorpcji energii

Streszczenie: W artykule przedstawiono wpływ rodzaju i struktury wzmocnienia na właściwości mechaniczne oraz balistyczne kompozytów polimerowych na osnowie żywicy epoksydowej. Jako wzmocnienie zastosowano tkaniny aramidowe, bazaltowe, szklane oraz ich układy hybrydowe. Zbadano udarność wg Izoda i „spadającego grota", wytrzymałość na zginanie i strukturę otrzymanych kompozytów. Oznaczono również ciężar właściwy. Hybrydowe kompozyty aramidowo-szklane wykazały dużą wytrzymałość na zginanie (397 MPa) i moduł Younga (21 GPa). Natomiast kompozyty aramidowo-bazaltowe cechowały się wysoką udarnością (116 kJ/m^2) i dużą absorpcją energii uderzenia (45 J).

Słowa kluczowe: hybrydowe kompozyty epoksydowe, modyfikacja wzmocnienia, włókniny aramidowe, szklane i bazaltowe, właściwości mechaniczne.

Obtained in 1965 by Stephanie Kwolek, poly(phenylene-1,4-diamide) started a revolution in armors, both in equipment and personal protection for soldiers. Polymer composites often replace the steel used in the production of helmets, vests, and tank armor. The reason for this phenomenon were the unique properties achieved by polymer materials reinforced with synthetic fibers [1–3]. These materials show similar strength parameters and much lower density, compared to steel components. Heavy steel helmets and vests, which negatively affected soldiers' mobility, were replaced by lightweight and thin ballistic composite materials providing comparable protection against combat agents [4–6]. In the defense industry, hybridization of reinforcement is used to reduce the cost of armor production. Production of details reinforced only with aramid fiber or UHMWPE is too expensive and becomes unprofitable. Therefore, fiberglass and basalt reinforcements are additionally used. The second and most important aspect, from the utilitarian point of view, of the reinforcement hybridization is the additivity of positive properties, while simultaneously compensating for the disadvantages possessed by each type of fiber. As a result, the composite acquires unique properties [4, 7–9].

The mechanical strength of composite materials depends on the reinforcement used. A key aspect in the design of composites for the designer is to have knowledge of reinforcement types and properties, as well as an awareness of the manufactured component final purpose. High-performance fibers are used as reinforcements in polymer composite armors. This group includes p-aramid, ultra-high molecular weight polyethylene (UHMWPE), glass, and basalt fibers [10, 11]. The most popular of the high-performance fibers is p-aramid fiber, which has been strongly associated with the defense industry for several decades. P-aramid fibers

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have high tensile strength in the longitudinal direction and abrasion resistance. They are characterized by low density, high modulus, and the ability to absorb impact energy in the form of plastic deformation [11–13]. They also exhibit excellent thermal stability, retaining their properties over a wide temperature range from –196 to 427°C. Disadvantages of aramid fibers include UV sensitivity, as well as poor compressive strength [12, 14, 15]. UHMWPE fibers, like aramid fibers, have excellent mechanical properties that are key to the defense industry. They exhibit high tensile strength and modulus [16]. They have high resistance to impact, wear, friction, and chemicals. In addition, they show low moisture absorption [17, 18]. Unfortunately, UHMWPE fibers have a low melting point, which hinders application possibilities. The high degree of crystallinity and lack of polar functional groups make these fibers have low surface energy. This translates into poor interfacial adhesion between the fibers and matrix. This requires UHMWPE fibers to have their surfaces modified before they can be used as reinforcements in composites [19, 20]. In the reinforcement industry, glass fibers are used because of the favorable ratio of suitable properties to low price. They exhibit good mechanical properties that are stable at both low and high temperatures. In addition, glass fibers have good impact resistance, fire resistance and low moisture absorption. However, they show sensitivity to alkalis, phosphoric acid, have low modulus and low fatigue resistance [7, 15, 21, 22]. Nowadays, basalt fibers are experiencing a renaissance. During World War II, work on basalt fibers for military applications was carried out by both the US and the Union of Soviet Socialist Republics. In the 1970s, interest in basalt declined in the US in favor of glass and aramid fibers [23, 24]. In recent years, there have been many papers describing the possibility of using basalt in lightweight armors or explosion-proof manholes. Scientists' interest in this material is due to its unique properties. Basalt fibers are non-toxic and chemically stable. They have good mechanical properties, tensile strength, which is in the range of 3000–4000 MPa, and Young's modulus of 80-110 GPa. The maximum service temperature of basalt fibers is 1255°C. Unfortunately, these fibers are characterized by brittleness. During compression or impact tests, they easily break [11, 25, 26]. Basalt fibers have better properties than glass fibers, and they are cheaper than carbon fibers, which puts them as a potential replacement [27, 28].

It can be concluded that each reinforcement, in addition to a number of advantages, also has some disadvantages, which eliminates it from the "ideal reinforcement" role. The solution to this problem is multilayer hybridization of reinforcement, that is, the use of several different types of reinforcing fibers simultaneously. This hybridization makes it possible to compensate for the disadvantages and synergize the positive features of the initial reinforcements, thus obtaining an "ideal reinforcement." There are few ways to include different types of reinforcement in a composite. The most important of them are shown in Figure 1. Hybridization can involve the use of a clique or all fibrous reinforcement types, e.g.: fabric, mat, roving, 3D fabric, additionally made of different fiber types. This type of hybridization is called interlayer, which also includes combining fibrous and core reinforcements, e.g.: synthetic or metal honeycomb structures, porous ceramic or polyurethane structures. When two materials (a fabric in which the weft and warp yarns are two different fibers) enter within a single layer of reinforcement, we are dealing with intralayer hybridization. Furthermore, the yarn may consist of different fiber types or fibers of the same type coated with nanofiller, or both possibilities at the same time. This is an example of intrayarn hybridization [29–33].

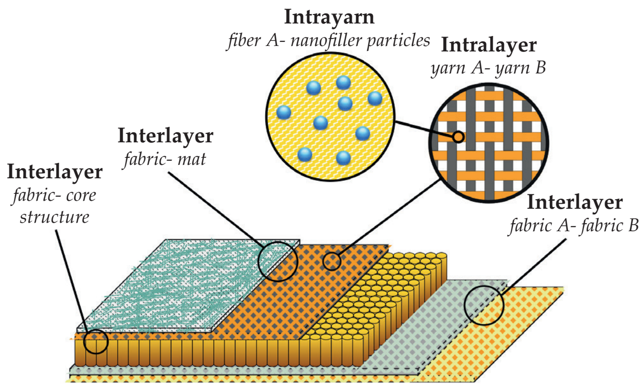

Fig. 1. Types of reinforcement hybridization used in polymer composites

Over the past four years, the hybridization topic of polymer composites reinforcement has been of particular and continuing interest to researchers. This is evidenced by numerous publications on intra- and interlayer hybridization. Chen *et al.* [34] investigated the intralayer hybridization effect on the axial crushing of tubes that mimic the structural component of automobile bodies. For this purpose, they made double-hat-shaped laminates reinforced separately with carbon, glass and carbon-glass fabric with yarn arrangement: carbon 90°/glass 0° and carbon 0°/glass 90°. As criteria for evaluating the strength of the details, the researchers took the values of mean crushing force (MCF) and absorbed energy (EA). Intralayer hybridization slightly improved MCF and EA relative to the carbon details. Samples reinforced with only carbon and glass fabric achieved similar values of absorbed energy. Among the hybrids, the tube reinforced with 0° carbon/90° glass fabric had higher MCF and EA values. Based on the results, the researchers concluded that the use of carbon-glass fabrics, can be an excellent compromise between the price and weight of the target part. Hashim [35] and his team tested the effect of fibers orientation on tensile and low-cycle fatigue of hybrid composites. They made 10-layer epoxy composites reinforced with an aramid-carbon hybrid fabric with a canvas weave, which had a carbon fiber as the matrix thread and an aramid fiber as the weft. They then cut the shapes at 0°, 45° and 90° to the direction of the carbon fiber. The 0° samples had the highest tensile strength (554 MPa) and Young's modulus (595 GPa). For 90° and 45° specimens, the tensile strength reached 468 and 111 MPa, respectively, and the modulus reached 24 and 6.4 GPa. For the 45° samples, high ductility related to fibers rotation was observed, which translated into an elongation-at-break result of 61.42 mm, which was about 6 and 10 times higher than the 0° and 90° samples, respectively. In addition, fatigue tests showed, a slower rate of fibers degradation in 90° shapes, compared to 0°. The mechanical and thermal properties of carbon reinforcement, basalt reinforcement and their intralayer hybrid were described in their work by Azimpour-Shishevan [36] and his team. They made epoxy matrix composites using an infusion method, which they subjected to tensile, 3-point bending and short beam shear tests. The epoxy-carbon composite exhibited the highest tensile, flexural and interlaminar shear strengths, as well as the highest Young's and flexural modulus. The composite containing the hybrid reinforcement exhibited slightly worse performance than the carbon laminate. Compared to the epoxy-basalt composite, hybridization significantly improves mechanical properties. A similar relationship occurred in the thermal conductivity test, where samples with carbon, basalt and hybrid reinforcement achieved conductivities of 0.6025, 0.47, 0.58 W/mK, respectively. A summary and comparison of the hybrid reinforcements mechanical properties described in [34–36] are reported in the work of Cho and Park [37]. The researchers evaluated the tensile strength, 3-point flexural strength and impact puncture of composites reinforced with carbon fabrics (CF/CF) and hybrid fabrics, in which the warp yarn was carbon fibers, and the weft was glass (CF/GF), basalt (CF/BF) and aramid (CF/AF), respectively. The matrix of the composites was thermoplastic Polyamide 6. In tension and bending, the specimens were evaluated in the warp and weft directions. The CF/CF composite achieved the highest values of tensile strength (419 MPa), flexural strength (232 MPa), flexural modulus (16 GPa) and Young's (25 GPa) in the weft direction. In the matrix direction, the CF/AF composite achieved the highest flexural modulus and flexural strength of 33.7 GPa and 188 MPa, respectively. The CF/GF composite had the best tensile strength and flexural modulus. In both tests, the CF/BF composite had the worse results, which the authors explain by the difficulty of supersaturation and thus poor impregnation of the basalt fibers by the polyamide matrix. However, in the "falling arrowhead" test, CF/CB achieved the highest peak energy of 18.3 J. In this test, the CF/GF sample performed the worst. When interlayer type hybridization is used, its effectiveness is determined by the type of appropriate selection of fibrous materials and their arrangement order. Pujar *et al.* [38] conducted a study on improving the tensile strength of epoxy-glass composites by introducing carbon fabric. For this purpose, they made four 10-layer composites using the vacuum bag method. Three of them were hybrids , in which carbon fabrics were placed sequentially: as the outer layers (H1), the third from the outside (H2) and the middle (H3). The last epoxy-glass laminate was a representative sample (G1). The introduction of carbon fabrics improved the mechanical properties in each case. The tensile strength and modulus of elasticity increased with the placement of carbon reinforcement layers towards the central part of the composite. For the H3 sample, they were 669 MPa and 14.4 GPa, respectively that is, there was an improvement in these parameters by 36% and 51%, respectively, compared to the representative sample. Karamooz [39] and Rezasefat [40] and their colleagues studied the effect of interlayer hybridization and stacking order to improve impact puncture resistance. The first team made 13-layer aramid-basalt hybrids and representative composites reinforced only with Kevlar and basalt fabric using a vacuum bag method. Epoxy resins were used as the matrix. Karamooz [39] used two types of sequencing: alternate, where the first and last layers were basalt fabric (BK-HI) or aramid fabric (KB-HI), and package, where 7 layers of basalt fabric (BK-HS) or aramid fabric (KB-HS) formed the core of the composite. The researchers performed a comprehensive puncture resistance test at two impact energies: 40 J and 60 J, using three types of arrowheads: flat, conical, and hemispherical. The highest absorption of impact energy had composites with an alternating sequence and then with a packet sequence. Laminate reinforced only with aramid fabric showed the lowest energy absorption capacity. The use of arrowheads with an increasingly sharper end, resulted in longer impact times, increased surface damage area and specimen penetration. As the sharpness of the arrowhead decreased, the value of the peak force decreased, and the absorption energy increased. On the other hand, a second research team under the direction of Rezasefat [40] used hybridization of aramid and glass fabrics in an epoxy matrix. The researchers made eight composites, including two representative specimens, using the infusion method. Hybrid laminates have been designed in such a way that aramid fabrics alternate with glass fabrics, constituting the outer layers, the core part, and the back layers of the laminate (3–5 layers). The obtained composites were subjected to puncture impact tests at three impact energies of 19, 37 and 72 J, using a hemispherical arrowhead. The post-test specimens were subjected to structure and response surface methodology (RSM) analysis. The composite with aramid fabrics as the last layers had the highest energy absorption coefficient for each impact energy. In the case of hybrid composites, the difference between the absorption coefficient and arrowhead displacement is insignificant for impact energies of 19 and 36 J. The lowest value of energy absorption and arrowhead displacement was observed for the epoxy-glass composite, which, in turn, had the highest peak force among all samples. In addition, the authors hypothesized that in the hybrid systems the damage to the matrix was bigger on the impact side and smaller on the back side compared to the represented composites. Bigger damage (delamination) was noticed between layers of glass and aramid fabric than between reinforcements of the same type. An attempt to use fabrics made of natural fibers (flax and kenaf) in structural components by hybridizing them with synthetic fabrics (glass and carbon) was carried out by Saroj and Nayak [41]. They made eight seven-layer composites by hand lamination. Four of them were laminates reinforced with only one type of fabric. The other four were hybrids, in which the outer layers were two synthetic fabrics (carbon or glass) and the inner layers were 3 layers of linen or kenaf reinforcement. The cut specimens were subjected to the following tests: 3-point bending and Izod impact tests. Hybridization of synthetic and natural reinforcement resulted in a huge improvement in mechanical properties compared to composites reinforced with natural fabric only. The hybridization of flax and carbon fibers showed the maximum effect, causing an increase of 600% in flexural strength from 54 to 364 MPa and modulus from 4 to 25 GPa. In comparison, the decrease in flexural strength of the hybrid relative to the epoxy-carbon composite was 24% and in modulus by 34%. For impact strength, a maximum increase in impact strength from 10.1 to 80.2 kJ/m^2 (800%) was observed for kenaf and glass fiber. Compared to glass laminate, the decrease was 66%.

In this paper, the effect of aramid, glass and basalt reinforcement hybridization on ballistic properties of epoxy resin composites was investigated. In addition, the possibility of using basalt fiber as a replacement for glass fiber, which is the main reinforcement used next to aramid fiber in ballistic composites, was investigated.

EXPERIMENTAL PART

Materials

For this study, the epoxy resin used was Epidian 624, with a density of 1.11 g/m^2 and an epoxy number of 0.485–0.51 mol/100 g, together with Z1 hardener from Ciech Sarzyna S.A. (Nowa Sarzyna, Poland). As reinforcement, aramid, glass, and basalt fabrics were used, all with a canvas weave and weights of, respectively: 220 g/m^2, 350 g/m^2 and 620 g/m^2 supplied by Rymatex Sp. z o.o. (Rymanów, Poland).

Composites preparation

Epoxy composites containing glass (K.S), aramid (K.A), basalt (K.B), aramid-basalt (K.AB) and aramid-glass (K.AS) reinforcements were obtained. Each composite consisted of four layers of reinforcement. In the case of K. AB and K.AS, it contained two layers each of a particular type of reinforcement, arranged alternately (Figure 2). The composites were obtained by infusion, percolating with Z1-cured resin in a weight ratio of 100:13. From the resulting laminates, samples were cut in the form of 60 × 60 mm plates and 10 × 80 mm and 15 × 60 mm beams.

Methods

To determine the specific gravity of the obtained composites, the density was calculated by weighing and sizing the cut plates. The impact strength of notched specimens was determined by the Izod method according to EN ISO 180 using an Instron Ceast 905 impact machine (Pianezza, Italy), with a hammer impact energy of 5.5 J. In addition, the machine recorded the absorption of impact energy. The impact test (impact puncture) by the falling arrowhead method was carried out using a drop tower designed by Proximo Areo sp.z.o.o. (Rzeszow, Poland), in accordance with PN-EN ISO 6603-2. A weight of 10 kg was dropped onto 60 × 60 mm specimens from a height of 1 m, at a speed of 4.4 m/s. A hemispherical arrowhead with a diameter of 20 mm was used for the test. 3-point bending was tested using an Instron 5967 testing machine (Grove City, USA), and performed in accordance with PN-EN ISO 14125. The speed of the bending head displacement was 5 mm/min. An Olimpus model DSX510i (Tokyo, Japan) optical-digital microscope was used to analyze the structure of the samples after the impact puncture test. The samples were cut in half in the place of the sample perforation.

RESULTS AND DISCUSSION

Specific gravity analysis

Figure 3 shows the changes in specific gravity of the obtained composites. The composite containing only aramid reinforcement had the lowest specific gravity, while the glass reinforcement had the highest. Hybridization increases the specific gravity of the com-

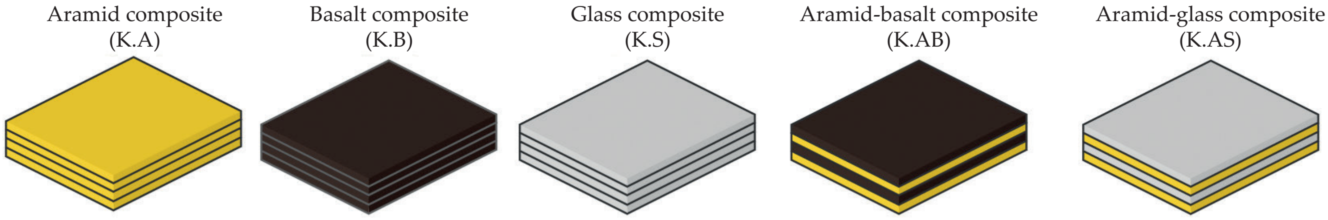

Fig. 2. Construction of the reinforcement structure

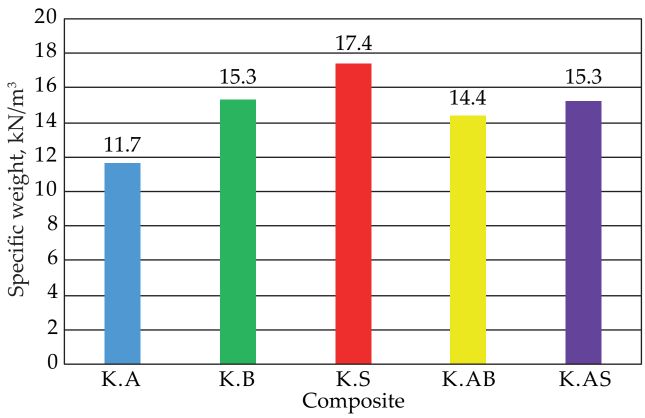

Fig. 3. Distribution of the tested composites specific gravity

posite in relation to K.A. This disadvantage is compensated by lower production costs. Due to the final weight of the detail, the use of basalt fabrics for reinforcement hybridization turns out to be a better choice.

Impact strength analysis

Among the tested composites, K.B had the highest impact strength and energy absorption values, which were as high as 205.1 kJ/m^2 and 79.4%, respectively. The worst result showed K.A composites, whose impact strength was 78.8 kJ/m^2, and the degree of absorbed impact energy reached a value of 17.8%. Analysis of the results presented in Figures 4 and 5 showed that for composites based solely on aramid reinforcement, hybridization of the reinforcement contributes to the impact strength and energy absorption improvement. Comparing K.AB and K.AS hybrid composites, much better results were achieved by the former. The aramid-basalt hybrid achieved impact strength the same as the glass laminate and by about 20 kJ/m^2 from the K.AS composite. In addition, the energy absorption level was 33.5%, for KA composites. K.S and K.AS did not exceed 20%.

In the impact puncture test, perforations occurred in all the tested composites except K.B (Fig. 6). Figure 7 shows the amount of impact energy absorbed by each laminate. In the case of the K.B composite, the impact energy was absorbed in 96%, which is reflected in the appearance of the specimen. The aramid-basalt hybrid allowed an increase in absorbed energy of 37.77 J compared to the K.A composite. In the case of the aramid-glass hybrid, no real improvement was observed. The increase in absorbed energy was only 0.85 J. The highest values of maximum energy and breakthrough (Figures 8 and 9) were characterized by K.B samples and the

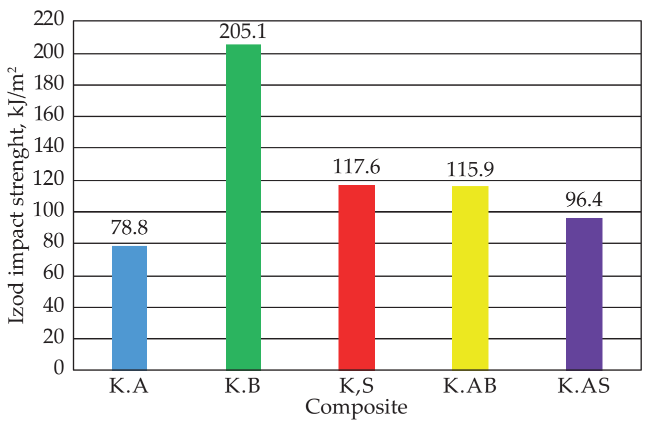

Fig. 4. Izod impact strength of the hybrid composites

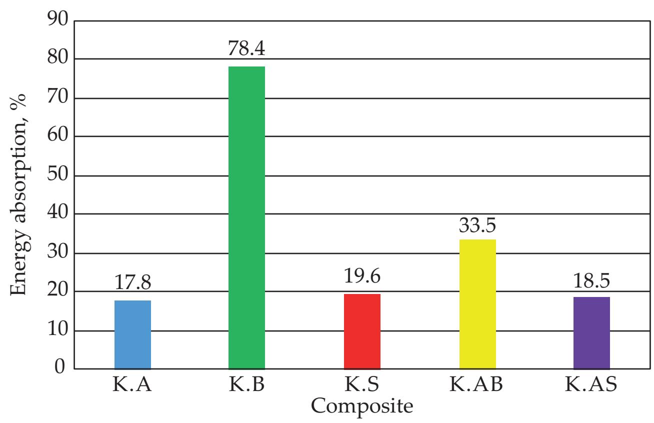

Fig. 5. Energy absorption of the hybrid composites determined in the Izod impact test

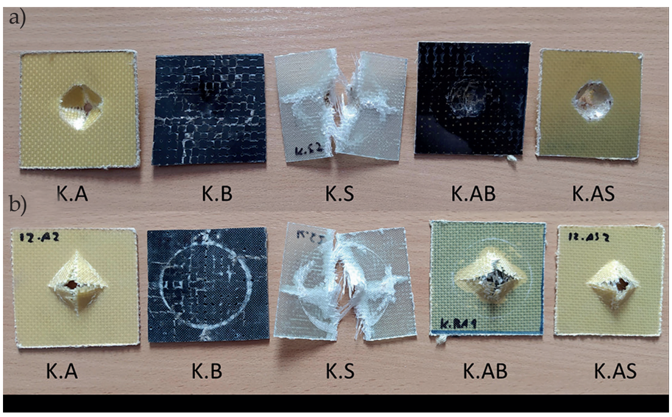

Fig. 6. The appearance of the samples after the falling arrowhead impact puncture test, a-front, b-back

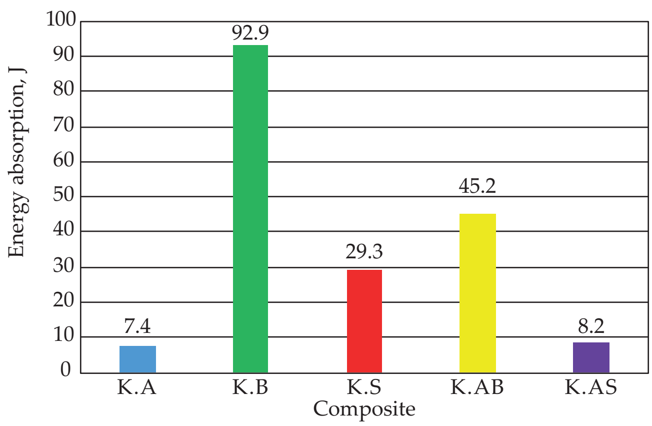

Fig. 7. Energy absorption in the falling arrowhead impact puncture test

T a b l e. 1. Mechanical properties of the tested hybrid epoxy resin matrix composites

Composites symbol	Izod impact strength kJ/m²	Energy absorption %	Maximum energy J	Puncture energy J	Energy absorption J	Young's modulus GPa	Bending deformation %	Flexural strength MPa	Specific gravity kN/m³
K.A	78.76 ± 6.44	17.84 ± 1.89	4.64 ± 0.06	6.40 ± 0.12	7.39 ± 0.30	11.82 ± 0.64	5.30 ± 2.73	223.10 ± 4.31	11.69 ± 0.32
K.B	205.07 ± 14.48	78.36 ± 12.93	73.72 ± 5.87	62.25 ± 2.63	92.92 ± 2.24	14.66 ± 1.33	1.87 ± 0.33	243.78 ± 26.16	15.32 ± 0.20
K.S	117.64 ± 7.28	19.57 ± 1.80	18.45 ± 0.38	22.75 ± 1.51	29.33 ± 1.55	22.37 ± 2.37	4.02 ± 0.46	658.69 ± 11.28	17.39 ± 0.82
K.AB	115.86 ± 5.60	33.50 ± 4.76	25.09 ± 3.50	31.31 ± 2.59	45.16 ± 2.41	15.56 ± 0.78	2.39 ± 0.36	296.31 ± 44.67	14.39 ± 0.23
K.AS	96.41 ± 7.50	18.49 ± 1.95	7.67 ± 0.15	9.81 ± 0.87	8.24 ± 2.86	20.60 ± 2.03	2.03 ± 0.11	396.75 ± 27.46	15.28 ± 0.04

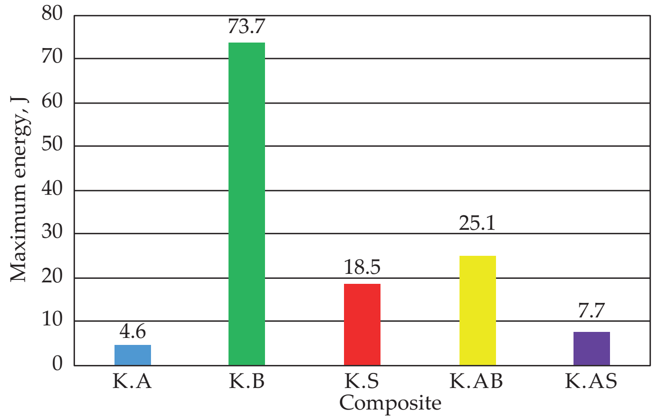

Fig. 8. Maximum impact energy of the composites

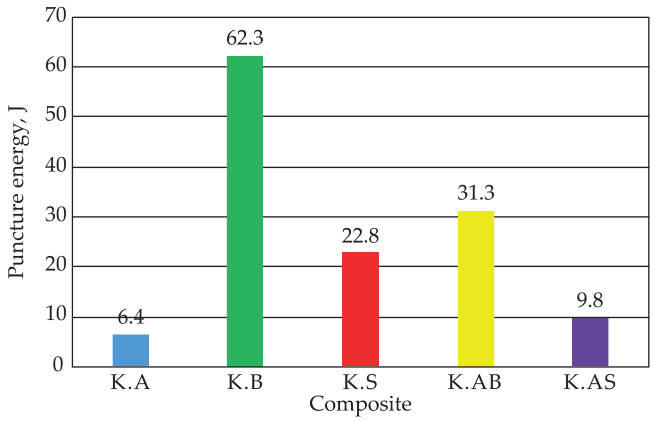

Fig. 9. Puncture energy of the composites

lowest, K.A. Hybrid laminates K.AB and K.AS, both in the case of maximum energy and breakthrough reached a higher value compared to K.A. Moreover, in the case of hybrids, the individual energies values achieved by K.AB are much higher than K.AS. This shows that the impact properties of the basalt reinforcement are fundamentally affected, while those of the glass reinforcement are insignificant. Although higher energy values were recorded for the K.S composite than for K.A, significant damage to the samples occurred, causing them to be sawn through. K.A perforation in terms of shape and size of damage is comparable to K.AB and K.AS perforations.

Flexural properties analysis

Figures 10-12 and Table 1 show the results of the fabricated composites 3-point bending. The highest flexural strength and modulus were characterized by composites reinforced entirely or in half with fiberglass fabric. In addition, K.S had a good strain of more than 4%, compared to composites containing basalt reinforcement, whose strain was about 47% lower. Of all the composites, the highest strain was achieved by the K.A samples, which at the same time had the worst flexural strength of 223 MPa, and a flexural modulus of 11.8 GPa. This represented almost three times lower strength and almost two times lower modulus, compared to composites con-

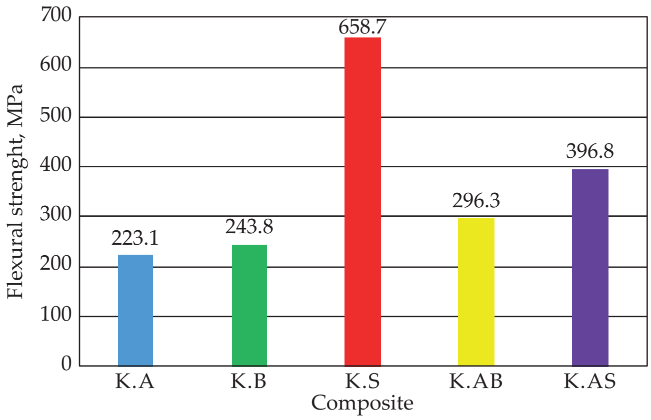

Fig. 10. Flexural strength of the composites

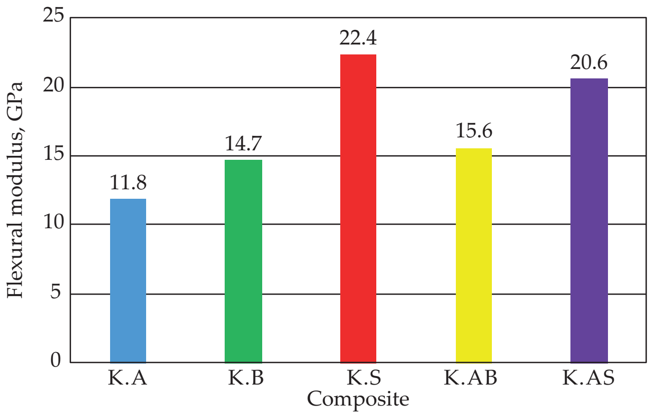

Fig. 11. Flexural modulus of the composites

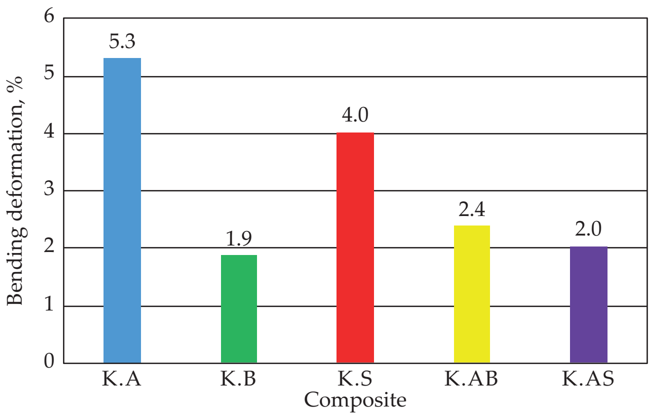

Fig. 12. Bending deformation of the composites

taining glass reinforcement. Hybridization of basalt and glass fiber reinforcements increased the flexural strength and modulus by 27.2% and 31.6% for K.AB, respectively, and by 77.8% and 74.3% for K.AS, respectively, compared to the composite reinforced with aramid fabric alone. The hybridization of aramid and basalt reinforcement made it possible to nullify the brittleness of basalt fibers, which in turn increased the modulus and flexural strength. The same but significantly better effect on flexural strength and modulus was obtained after hybridization of aramid reinforcement with glass reinforcement.

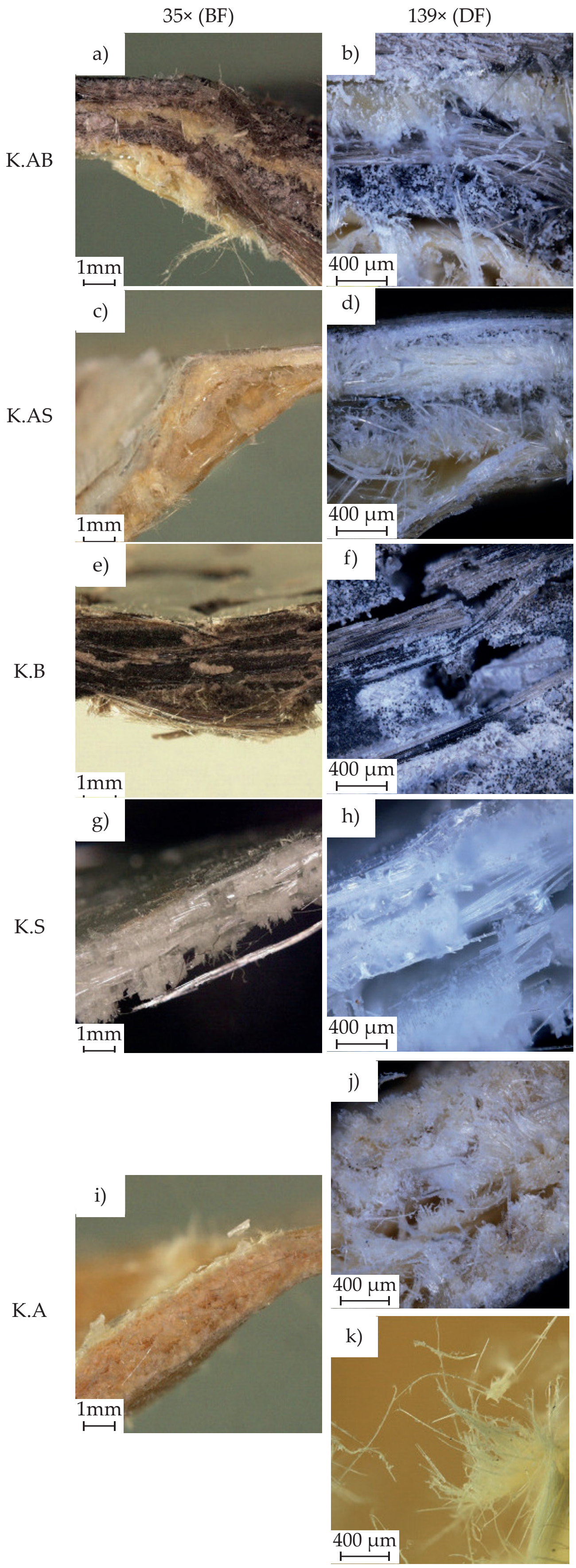

Fig. 13. Microscopic images of composites after impact testing

Structure analysis

Microscopic images were obtained at 35× magnification using the bright field (BF) technique (Fig. 13 a, c, e, g, i) and 139× using the dark field (DF) technique (Fig. 13 b, d, f, g, j, k). Interlaminar delamination was observed on each specimen, with the biggest degree of delamination occurring in the K.AS and K.AB hybrid composites (Fig. 13a and c). The least delamination occurred in K.B. Fig. 13f shows basalt yarn cracking and stretching, caused by the impact of the arrowhead. In addition, it can be seen that K.B and K.AB have good saturation of the fibers with epoxy resin. In the case of the K.AB composite (Fig. 13b), in addition to delamination, intra-layer delamination of the basalt fabric located between the aramid layers can be seen. In the case of K.S, the impact caused the sample to be cut in half (Figure 6). This resulted in breaking and tearing of the glass fibers, as well as the delamination seen in Figure 13g and h. For the K.A sample, fibrillation characteristic of aramid fibers can be seen in Figure 13k.

CONCLUSIONS

Hybridization of reinforcement in polymer composites can significantly improve their mechanical properties, as well as expand their range of applications. Replacing part of the aramid reinforcement layers with glass or basalt reinforcement can reduce the production cost. Unfortunately, this results in an increase in the specific weight of the resulting structural element. In conclusion, when designing ballistic shields made of such hybrid composites, due to the high flexural strength, modulus and appropriate deformation, aramid-glass hybrid should be used as an outer layer of the cladding. On the other hand, aramid-basalt hybrid, which is characterized by high impact strength and impact energy absorption, should be used as core layers, for example: a core layer located between two honeycomb structures.

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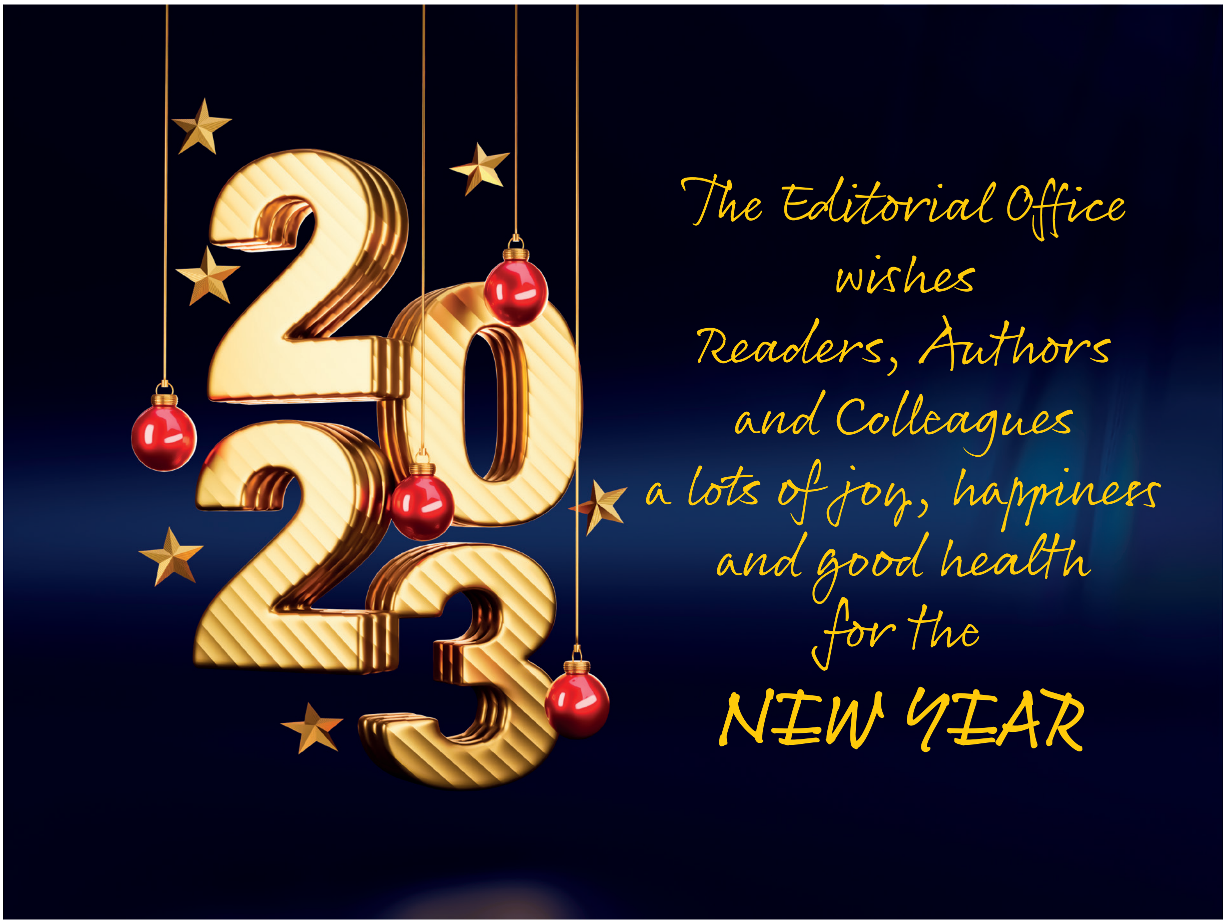

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