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# Description of the Yield State of Bioplastics on Examples of Starch-Based Plastics and PLA/PBAT Blends

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The present work concerns the description of the yield state of biodegradable materials. As examples, biodegradable polymers are chosen – cornpole CRP-M2, starch fatty acid ester, and PLA/PBAT, poly(lactic acid) (PLA) blended with poly(butylene adipate/terephthalate) (PBAT) [1, 2]. These biodegradable, plant-derived bioplastics are a promising alternative to petroleum-based plastics. To describe the onset of plasticity in the bioplastics under discussion, Burzyński 's hypothesis of material effort has been applied [3, 4]. The applied criteria account for the differential strength effect and for the shear correction resulting from the difference between experimental and theoretical values, obtained as a result of the Huber-Mises approach [5, 6]. In general, these properties of yield state are characteristic for polymers. The description of yield state for bioplastics is an issue that has hardly been investigated, which illustrates the novel nature of this paper in which this topic is discussed.

**Key words:** bioplastics, strength differential effect, shear correction, yield surface, Burzyński yield condition.

## 1. Introduction

The amount of plastics used in industrial applications is increasing from year to year. In contrast to metals which are easily recoverable and recyclable, plastic waste does not decompose easily and quickly causes considerable damage to the environment. Plastics are more than 12 percent of municipal solid waste, which is a huge increase since 1960, when plastics were less than one percent of this waste [1]. One way of solving the increasing problem of plastic waste is by introducing the concept of isofunctional recycling, e.g. [7]. Another solution can be biodegradable plastics. Biodegradable plastics can be broken down by microorganisms and will eventually decompose fully into water and carbon dioxide, which makes them friendly to people and to the environment [2]. Biodegradable plastics (plastics that can decompose in the natural environment) and biomass plastics (plant-derived or recyclable resource-based plastics) have been extensively investigated in recent times; new biodegradable and biomass plastics are being continuously developed. The input materials for the production of these polymers may be either renewable (based on agricultural plant or animal products) or synthetic. Bioplastics are biodegradable, less expensive and are more environmentally friendly in their production process and the distribution of waste materials. They can be used in many industries: the textile industry, the food and packaging industries e.g. the production of plastic cutlery, alimentary product containers, bowls and bags, as well as in medical applications and confectionary [1, 2, 8, 9]. Bioplastics can also be used as structural elements: interior components of cars, computers and cell-phones. Bioplastics are a promising alternative material to petroleum-based plastics, consequently knowledge about their properties and behavior under different loadings can be helpful in the further improvement of bioplastics.

In this study, the following are discussed as examples of biodegradable plastics: cornpole CPR-M2 – the starch fatty acid ester and PLA/PBAT polymer alloy – poly(lactic acid) (PLA) blended with poly(butylene adipate/terephthalate) (PBAT)

Cornpole (the brand name of Nihon Cornstarch Co., CPR-M2, [2]) – the starch fatty acid ester is produced by extracting and refining starch from corn. The processes of extracting corn-starch and the production of materials based on this substance were patented in the 1970s (e.g. [10]), but the increase in interest in bio-derived plastics started at the beginning of this century, when the development of environmentally friendly polymeric materials became more ecologically urgent [11–13]. Starch is a useful material for biodegradable plastics because of its natural abundance and low cost. It is the major carbohydrate in plant tubes and seed endosperm, where it is found as granules. Each granule contains several million amylopectin molecules accompanied by a much larger number of smaller amylose molecules. The largest source of starch is corn and other commonly used sources are wheat, potatoes and rice [14]. Starch is fully biodegradable, contains no harmful substances and can be composted. In general, cornpole polymers are water resistant and the heat of combustion is less than that of petroleum plastics. This kind of corn-based polymer is highly compatible with other biodegradable polymers such as polyester, PHBV, PLA, and PCL, which allows a wide variety of new composite materials to be produced [2]. The chemical composition and aspects of production of corn-based plastics are described in [15]. The effect of strain rate and temperature on compressive properties of starch-based biodegradable plastics was examined in the paper of [16].

Poly(lacticacid) (PLA) is a linear aliphatic biodegradable polyester derived from biomass through bioconversion and polymerization. Lactic acid is firstly produced from various starches (corn or sugarcane and other biomass materials through biological fermentation) and then chemically converted to poly(lactic acid) [17]. This bioplastic is applicable to various industrial needs in the areas of packaging, biomedical, disposable cutlery, pharmaceutical, etc. [18, 19]. The properties of PLA, including values of yield strength, high transparency, elastic modulus and thermal stability, allow this material to be thermoplastically processed like conventional plastics. Compared to conventional polymers (PS, iPET, iPP), PLA presents the highest mechanical properties but the lowest thermal resistance [20]. However, as a crystalline polymer, PLA is very brittle and its low toughness and other drawbacks (low impact strength, poor heat resistance, it is also readily hydrolysable) limit its applications [21]. Polymer blends, alloys or the addition of natural fibers are used to overcome the brittleness and low impact resistance of PLA. To improve the resulting bio-material, PLA is blended with other flexible polymers; in this study, the poly butylene adipate-co-terephthalate (PBAT) was added. PBAT, as a blending polymer, is also fully biodegradable and flexible, though its Young's modulus and tensile strength are lower than in PLA, whereas the breaking strain is much higher than in PLA [8, 9, 22]. It is a flexible material; it has a high elongation at break, as well as good hydrophilic and processing properties. It can be used in the production of blown film and its associated membrane products [21, 23]. Polymer alloys of PLA/PBAT blends are very promising for the engineering industry because of their better ductility and higher strength in comparison to pure PLA bioplastics. Consequently, numerous studies present discussions on the mechanical, thermal and biodegradable properties of PLA/PBAT blends. The compatibility, crystallization and tensile properties of PLA/PBAT blends in different proportions are given in [24]. The conclusion states that PBAT effectively toughens PLA. The rheological properties of PLA/PBAT blends with increasing PBAT content are studied in [25]. The mechanical properties of the PLA, PBAT and PC blends and increase in tensile stress and impact strength of the PLA and

PBAT blends due to reactive extrusion are shown in [26]. Whereas the biodegradation behaviour of the blends, and the respective degradation behaviour under soil conditions is discussed in [21]. The effect of dialkyl peroxide blending on the tensile properties of PLA/PBAT blends is discussed in [9], whereas the dynamic compressive properties of these blends is examined in [8].

However, up to now no study has shown how different proportions of PLA/PBAT blends influence the yield state of the resulting polymeric material. Under a phenomenon called yielding, a material transition from the elastic state to the plastic state can be understood [27]. For uniaxial stress state, the moment of obtaining plasticity by the material is given by the yield strength. In cases of complex stress states, identifying the moment when the plastic state occurs in the material is possible if the components of the stress tensor fulfill the function called yield condition. As the usage of polymer materials in structural applications is increasing, researchers and engineers are looking more comprehensively at their mechanical properties, leading to more detailed investigations into their behavior under different loading conditions. Knowledge about the yield state of a material is essential to predict the response of a structure under different loadings and its susceptibility to various failure modes.

In general, polymers are characterized by different properties in tension in comparison to compression [28]. The compressive behavior of polymers at different strain rates has been a topic of many investigations [e.g. 29–32]. The determination of a tensile behavior of polymers, especially in high strain rates, is not an easy task due to the complex nature of the experimental techniques. However, it is important to understand the mechanics governing deformation at high rates and to help improve the physics-based constitutive modeling of the high-rate behavior. The yield behavior of polymeric materials exhibits pronounced hydrostatic pressure dependency. That conclusion is confirmed by many scientific studies e.g. [33–36]. First basic yield criteria (e.g. von Mises-Huber [5, 6], Tresca [37]) are unable to describe that phenomenon occurring for polymers. Further investigations proved that to describe the yielding of polymers properly, the first invariant of the stress tensor must be incorporated to involve the influence of hydrostatic pressure on the yield behavior, e.g. [34, 38, 39]. It was concluded that all polymers which deform relatively homogeneously (e.g., polyvinylchloride (PVC), epoxy resin, and high-density polyethylene (HDPE)) should obey a criterion which accounts for hydrostatic influence (e.g. modified von Mises criterion [34]. Polymers which deform inhomogeneously by the deformation of shear bands (e.g., polyethylene terephthalate (PET)) should follow a modified Tresca criterion, also known as the maximum shear stress theory [38]. More modern approaches not only include the first invariant of the stress tensor (the hydrostatic pressure influence) but also the third invariant of the stress tensor – in the case when the beginning of the plasticity in the material mainly corresponds to the evolution of shear bands [35, 40, 41]. In [34], it is shown that the criteria including only hydrostatic pressure are not capable of predicting the viscoplastic responses of all thermoplastic polymers subjected to any general biaxial stress state. The introduction of the third invariant of the deviatoric stress tensor does not affect the yield stress in uniaxial tension, uniaxial compression, or in purely hydrostatic pressure. However, differences in the calculated yield state are observed under pure shearing. Consequently, to improve the prediction of the yield behavior of isotropic polymers and to establish a generalized yield criterion under any ranges of biaxial stress state (so, accounting for shear when the main deformation mechanism is related to shear banding along with hydrostatic pressure dependency), the first and the third invariants of the deviatoric stress should be accounted in the expression of the yield equation. The criterion, formulated in such a way, applied to the experimental data on PMMA, PC and PS showed some improvement over the yield loci drawn in comparison to criteria including only hydrostatic pressure [34]. Also, in the case of Nylon 101, the research of [41] confirmed that the criterion in the form of $f(I_1,\ I_2,\ I_3)$ allows the accurate modeling of yield state to be obtained.

Our objective is to propose a criterion with a simplified account for the aforementioned SD effect and shear influence on material deformation – these are phenomena which greatly affect the yielding of polymers, considered as isotropic and homogeneous materials. Our proposal, based on the Burzyński criterion, is a yield model that is a simple to use and easily applicable, in which only three material parameters are required: the yield limits in tension $\sigma_Y^T$, compression $\sigma_Y^C$ and shear $\tau_Y$ [3, 4, 36, 42]. The difference of the values of yield stress in tension and compression, leading to the asymmetry of the elastic range, and the deviation of the yield strength in shear from the theoretically anticipated value are included in the proposed formulation. The criterion proposed in the paper incorporates the first (hydrostatic pressure influence) and third (certain correction of the yield limit in shear) invariants of deviatoric stress. It may be used to model the yield state of isotropic materials (as regards elastic and plastic yield properties). The criterion discussed is described in the area of principal stresses by a paraboloid of revolution for the yielding of the isotropic materials and by an elliptic paraboloid for materials for which the yield limit in shear varies from the theoretical value (based on the Huber-Mises approach [5, 6], Eqs. (3), (4)).

In the study presented here, the results of tension, compression and shear tests for the cornpole CPR-M2 and the PLA/PBAT blends, mixed in the different ratios (60:40, 70:30, 80:20) with the addition of the cross-linking agent at a weight ratio of one are used to estimate the onset of plasticity using the Burzyński criterion. This criterion, accounting for the SD effect and the shear correction, describes the yield state of polymers in general and, in particular, the yielding state of the discussed bioplastics. The description of the onset of plasticity in the case of the bioplastics, which are relatively new materials in engineering applications, seems to be a topic which should be explored further scientifically. The aim of this paper is to add some information about the yielding description of the exemplary bioplastics.

In Sec. 2 the results of mechanical tests of CPR-M2 cornpole and the PLA/PBAT blends are discussed. The description of the proposed yield criterion is given in Sec. 3. Section 4 presents the discussion and visualizations of the onset of plasticity for the given bioplastics. The conclusions are presented in Sec. 5.

## 2. Mechanical properties of bioplastics CPR-M2 and PLA/PBAT blend

In this section, the results of the quasi-static strength tests: compression, tension and shear for cornpole CPR-M2 are presented. In a further part of the section, the results of quasi-static and dynamic compression and tension tests of PLA/PBAT polymer blends are discussed.

In the quasi-static compression tests performed for cornpole CPR-C2, specimens with a diameter and thickness of 6 mm and 9 mm respectively were used. In the tensile tests, the flat dog-bone specimens presented in Fig. 1 were produced from the 5-mm-thick plates using a milling machine. The gage mark area measured 5×10 mm, Fig. 1. Both the compression and tension tests were carried out at strain rate $\dot{\varepsilon} = 0.001$ s$^{-1}$.

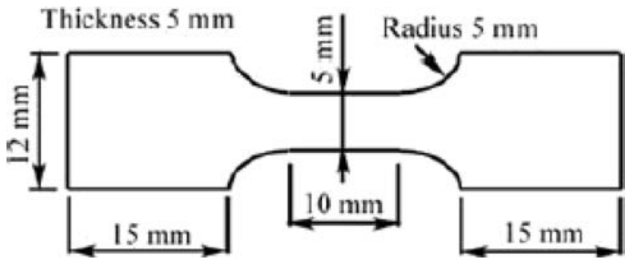



Fig. 1. Specimens for quasi-static tension test, used in the case of the cornpole CPR-M2.

The double shear tests were performed at quasi-static strain rates varying from $10^4$ s$^{-1}$ to 0.1 s$^{-1}$. A modified double shear (MDS) specimen is presented in Fig. 2. The double shear test is described in e.g. [43–45]. The geometry presented in Fig. 2 is chosen to minimize error resulting from any non-homogeneity of the shear stresses and strains at the shear zone of the specimen [45].

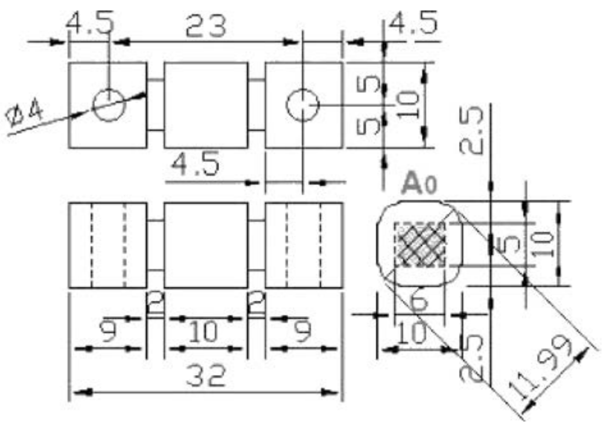



FIG. 2. Geometry of specimen for double shear test, the shear zone is indicated by gray shading.

In Fig. 3 the results of the compression and tension tests performed at strain rate $10^{-3}$ s$^{-1}$ are presented. The stable behavior of the cornpole without hardening was observed during the compressive test at quasi-static strain rate. During the tensile test, stress increased linearly with increasing strain – after obtaining the maximum stress (for $\varepsilon = 0.12$), the stress decreased quickly. Cracks were observed at maximum stress and finally the specimens broke at the fracture point. These observations confirm that the material analyzed is brittle. It is clearly noticeable that there is a large difference between levels of stresses for tension and compression.

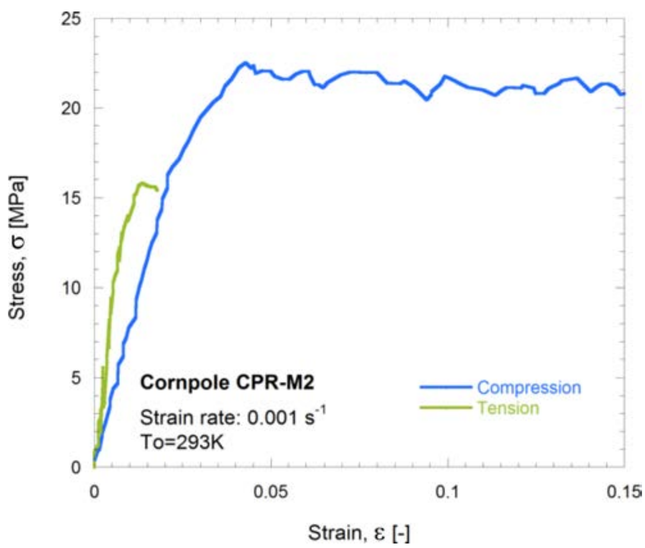
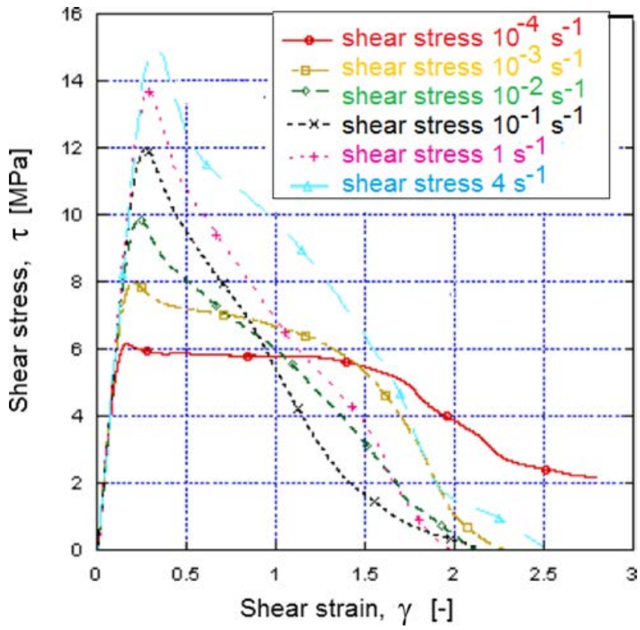



FIG. 3. CPR-M2. Results of tension and compression tests.

In Fig. 4 the results of double shear test – performed for a wide range of strain rates – are presented. Curves account for the correction resulting from the inho-

a)




b)

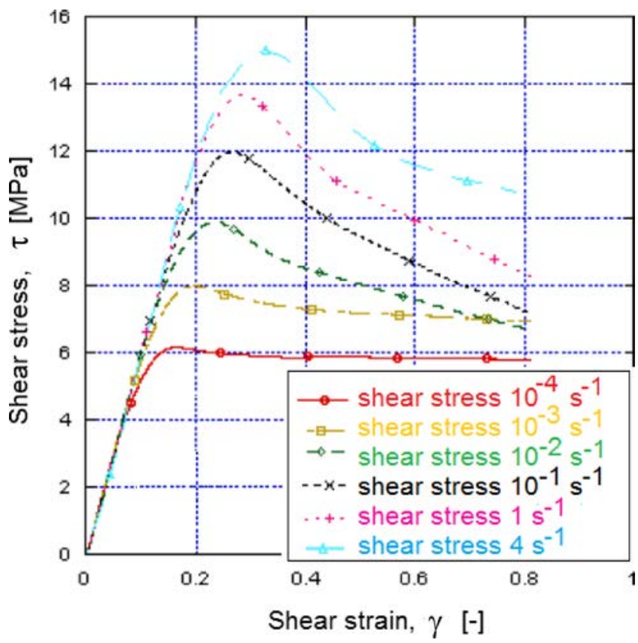



Fig. 4. CPR-M2. Shear stress-strain curves at several quasi-static strain rates, [59].

mogeneous shear zone. The origins of correction are clarified in [45]. The results are also corrected in that the same shear modulus is imposed, $G_{\text{theor}} = 60$ GPa. The shear test results show a significant softening by a drop of shear stress-strain curves. The effect can be explained from a thermodynamic point of view by the transformation of a part of mechanical work into heat. According to this hypothesis, the heat spreads through the material, causing an increase in temperature which depends on the heat flux exchanged between the material and the surroundings. Assuming that the strain rate of the test is high enough that heat exchange does not occur between the MDS specimen and the room environment, the increase in temperature causes a significant stress relaxation – characteristic for polymers.

In the next part of this section, the results of PLA/PBAT blends of the mixing ratios (weight fraction) of PLA and PBAT 80:20, 70:30 and 60:40, investigated under compressive and tensile loadings, are presented. The effect of the cross-linking agent – dialkyl peroxide, is also examined. The subsequent blends 80:20:1, 70:30:1 and 60:40:1 were tested. For the purposes of this study, the PLA/PBAT alloys were prepared using PLA from Toyota Motor Corporation (Eco-plastic S-17) and PBAT from BASF SE (Ecoflex). The polymer alloys were produced using a twin-screw extruder (TECHNOVEL CORPORATION) at 180°C. As a cross-linking agent – dialkyl peroxide (NOF Corporation, PERHEXA 25B) was added. The screw speed was 400 rpm and the feed rate was 100 g/min. After melt mixing, the strands prepared by the twin-screw extruder were cooled rapidly, pelletized, and then dried. The 5-mm-thick plates were prepared using a conventional hot press at 190°C and 5 MPa for 30 min [8, 9]. In the final products, the effects of the addition of dialkyl peroxide (cross-linking agent) and the different mixing ratios of PLA and PBAT on mechanical properties were examined. In [8, 9], the behavior of the bioplastic obtained was studied under compressive loading. The stress-strain curves of PLA/PBAT polymer alloys were obtained using a universal testing machine [A&D Co., Tensiron RTM-500] and a split Hopkinson pressure bar system. In [8, 9] the discussion about the same polymers is presented, but bioplastics were examined under tensile loading in quasi-static strain and dynamic strain rates.

The stress-strain curves of PLA/PBAT blends were obtained at high strain rates (600–900 $\text{s}^{-1}$) by using a split-Hopkinson pressure bar for the compression tests and a tensile split-Hopkinson bar for the tensile tests. The tests were carried out for tensile loading at strain rates from $\dot{\varepsilon} = 0.0001\ \text{s}^{-1}$ to $\dot{\varepsilon} = 10^3\ \text{s}^{-1}$ and compression test were performed at strain rates from $\dot{\varepsilon} = 10^{-4}\ \text{s}^{-1}$ to $\dot{\varepsilon} = 10^4\ \text{s}^{-1}$. For the dynamic compressive tests cylindrical specimens of diameter 15 mm and thickness 5 mm were used [8]. In the quasi-static tests, the specimens with a diameter and thickness of 6 mm and 9 mm were used [8]. The results of dynamic compression tests are given in [8]. For the blend PLA:PBAT = 80:20, the stress-strain curve reaches its maximum just after the elastic limit and then the stress decreases gradually with the increasing strains. As the PBAT ratio increases, the peak of the stress-strain curve becomes smaller, and the yield stress and Young's modulus decreases. When PLA:PBAT = 70:30, the flow stress remains almost constant. When PLA:PBAT = 60:40, the flow stress increases slightly in line with the hardening of the material. The yield stress and Young's modulus decrease with increasing PBAT ratios. The addition of dialkyl peroxide reduces the yield stress and Young's modulus of the specimens and increases the work hardening [8]. Results presented in [8] show the effects of PBAT content on the nominal stress-nominal strain curves at dynamic and quasi-static strain rates. It can be observed that during the experiment, stress increases linearly with the increasing strain. After obtaining the maximum stress, the stress decreases gradually. The maximum stress and Young's modulus decreases and the elongation increases with increasing PBAT content. It can be concluded that PBAT improves the brittleness and fracture toughness of PLA. When PLA:PBAT: dialkyl peroxide = 60:40:1, the stress remains steady at approximately 25 MPa in a strain range of 0.04 to 0.11. After that, it decreases gradually. Necking starts at a strain of 0.125, and a clear crack is observed at a strain of 0.185. After that, the specimen breaks. The mixing ratios of components affect the shape of the stress-strain curves. The yield stress decreases and the elongation at break and strain energy increases with increasing PBAT content when dialkyl peroxide is used. The effects of the addition of the cross-linking agent on the final blends, their Young's modulus, the yield stress, the elongation at break and fracture morphology are examined and concluded in [8, 9].

Figure 5 shows the effect of strain rate on the yield stress, according to different proportions of the components – PLA/PBAT. For each blend, the yield stress increases with the strain rate, which is commonly noticed for most polymers – e.g. [31]. In the case of PLA/PBAT, the same tendency can be noticed, both for tension and compression results. However – the strain rate sensitivity for tension is smaller – Fig. 5b. Such a tendency may be explained by the strain-induced alignment of polymer chains. In tension, the molecular chains align uniaxially along the axis of elongation, whereas, in compression, the chains align in a plane normal to axis of compression, giving the very different strain hardening behavior [32].

The histograms with comparison among results in quasi-static tensile test and dynamic tensile test are presented in Fig. 6. The bars shows that there is not much difference – around 5–7% – in the values of yield points in tension compared between quasi-static and dynamic strain rate.

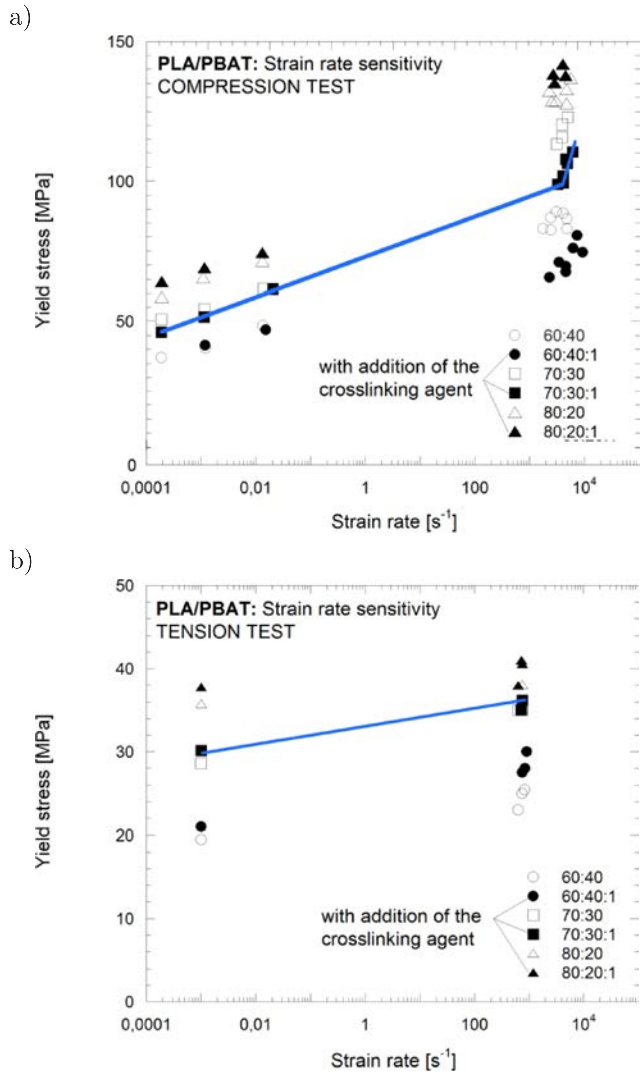



Fig. 5. PLA/PBAT. Strain rate sensitivity in a) compression and b) tension.

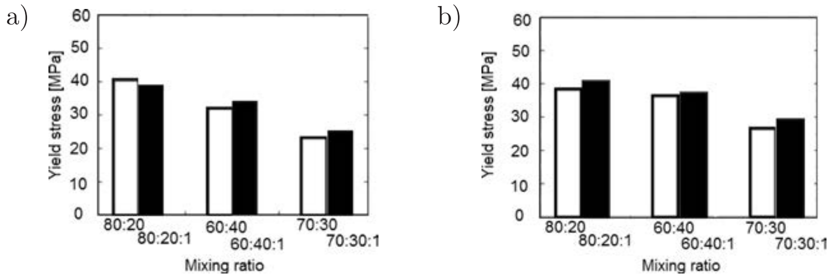



Fig. 6. Effect of PBAT content on the tensile yield stress at a) low strain rate and b) high strain rate.

In Fig. 7, the histograms of the yield stress in compression in quasi-static and dynamic tests are presented. At high strain rates, contrary to the results of tension tests, for compression tests the values of the yield stress are much higher than at quasi-static strain rates. In addition, the influence of the cross-linking agent is more ambiguous, the blends containing the agent have a yield stress 5–10% different (lower or higher) than the blends without it.

a) b)

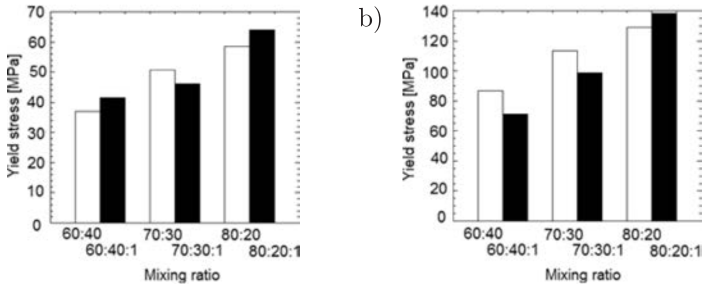



FIG. 7. Effect of PBAT content on the compression yield stress at a) low strain rate and b) high strain rate.

Knowing values of yield points for tension, compression and shear (for CPR-M2), it is possible to estimate the yield state of the discussed bioplastics. The yield hypothesis which is applied to describe the onset of plasticity is presented in the next section.

## 3. THE PROPOSED YIELD CRITERION

The energy-based hypothesis of material effort and resulting paraboloid yield criterion was originally proposed by Burzyński in the 1930s [3, 4], cf. also the comprehensive discussion of the Burzyński concepts in [42]. Generally, a superior yield criterion should have a physically based interpretation which can make the predicted result more reasonable. Energy as a multilevel scalar quantity can be assumed as an appropriate universal measure of the change of the strength of chemical bonds – material effort. The proposed concept is based on the hypothesis that certain portions of elastic energy density accumulated in the deformed body can be applied to define the measure of material effort. The concept of Burzyński was applied for the interpretation of the first investigations of plastic yield strength [46, 47] as well as for elaboration of data of modern isotropic materials in [36]. The recent implementation of the paraboloid yield condition in finite element code with use of Abaqus user subroutine UMAT is provided in [48].

The energy-based Burzyński hypothesis of material effort was formulated in particular to account for the strength differential effect described by the parameter $\kappa$ – the ratio of yield strengths in compression $\sigma_Y^C$ and in tension $\sigma_Y^T$, Eq. (3.1). The phenomenon of inequality of the tensile and compressive yield strengths is known as the strength-differential (SD) effect, it can be also denoted as an asymmetric effect. The SD parameter is defined by the following relation:

$$\kappa = \frac{\sigma_Y^C}{\sigma_Y^T}, \tag{3.1}$$

where $\sigma_Y^C$ is the yield strength in compression and $\sigma_Y^T$ is the yield strength in tension.

The SD effect is connected to the fact that the deformation and the failure stress of polymers are affected by the hydrostatic pressure, e.g. [35, 49]. It has been established that the viscoplastic flow of glassy polymers starts when the stress exceeds the resistance to molecular segments rotation. This effect also leads to a pressure dependence of the yield and failure stresses. The effect of hydrostatic pressure on the yield behavior of polymers was demonstrated experimentally in e.g.: [33–35, 50]. The investigation of polymer properties under hydrostatic loading is an importation issue, since pressure alters the character and mechanisms of the deformation processes, gives rise to transformations and affects the melting point and other material characteristics. Phase and polymorphic transformations, variations in porosity and the deformation mechanisms are also associated with the effect of hydrostatic pressure [35].

The details of the Burzyński hypothesis are precisely described in [3, 4, 36, 42], Burzyński paraboloid yield criterion for isotropic materials is presented in Eq. (3.2).

$$\frac{1}{2}\left[(\sigma_2-\sigma_3)^2+(\sigma_3-\sigma_1)^2+(\sigma_1-\sigma_2)^2\right]+(\sigma_Y^C-\sigma_Y^T)(\sigma_1+\sigma_2+\sigma_3) = \sigma_Y^C\sigma_Y^T. \tag{3.2}$$

In the area of principal stresses, the criterion has a form of paraboloid of revolution for materials with asymmetry of elastic range ($k \neq 1$). In the meriditonal plane, the graphical representation of the criterion is an arm of a parabola.

The criteria which incorporates the hydrostatic pressure can ascribed to the 1950s–1970s, e.g. [34], whereas the Burzyński proposal was published in 1928 [3, 4, 36, 42]. The Drucker-Prager yield criterion, one of the first pressure-dependent yield models, was published in 1952 [51]. In the area of principal stresses, it is represented by a conical failure surface. However, conical surfaces can only roughly approximate the real behavior of a material in the limited range of hydrostatic stress, additionally criterion formulated in such a form fails to properly describe the stress states near to the apex of the failure cone.

In the case of equality of yield stresses of compression and tension ($\kappa = 1$), the shape of yield surface is a cylinder of revolution. This is a representation of the Huber-Mises criterion [5, 6], which is applied to the bodies assumed as isotropic and insensitive to hydrostatic pressure. In the Huber-Mises criterion it is assumed that the yielding of materials begins when the second deviatoric stress invariant reaches a critical value. According to this theory, at the onset of yielding, the magnitude of the shear yield stress in pure shear is $\sqrt{3}$ times lower than the tensile yield stress in the case of simple tension [5, 6], Eq. (3.3).

$$\tau_Y^H = \frac{\sigma_Y}{\sqrt{3}} \tag{3.3}$$

and $\sigma_Y = \sigma_Y^C = \sigma_Y^T$.

If the strength differential effect occurs and $\sigma_Y^C \neq \sigma_Y^T$, what is accounted in the Burzyński criterion, Eq. (3.2), the yield strength in shear is calculated in the following way, Eq. (3.4):

$$\tau_Y^B = \sqrt{\frac{\sigma_Y^C \sigma_Y^T}{3}}. \tag{3.4}$$

Loading direction or stress state may result in a different response on the orientation of dominant slip-plane and the critical value of shearing stress. Yielding of element in a material often occurs when the shearing stress on a dominant slip-plane reaches its critical value [52]. The experimental observations show that the effect of the third invariant of stress tensor deviator appears due to shear processes [53, 54]. It means that the material starts to yield when it reaches the yield strength in shear. For polymeric materials, the deviation of shear strength is observed with respect to the value predicted in the Huber-Mises yield condition, given in Eq. (3.3). Such an effect can be described within the framework of the model for isotropic material by using the certain shear strength correction factor $\lambda$, Eq. (3.5).

$$\lambda = \frac{\sigma_Y^C \sigma_Y^T}{2\tau_Y^2} - 1, \tag{3.5}$$

where $\sigma_Y^C$ is the yield strength in compression, $\sigma_Y^T$ is the yield strength in tension and $\tau_Y$ is the yield strength in shear. In this particular case if $\lambda = 0.5$ and $\sigma_Y^C = \sigma_Y^T = \sigma_Y$ the classical Huber-Mises condition – Eq. (3.3) – is obtained.

An equation of paraboloidal Burzyński yield condition accounting for the shear strength correction factor $\lambda$ in the system of principal stresses takes the following form presented in Eq. (3.6).

$$\begin{aligned}(1-\lambda)(\sigma_2-\sigma_3)^2 + \lambda(\sigma_3-\sigma_1)^2 + (1-\lambda)(\sigma_1-\sigma_2)^2 \\ + (\sigma_Y^C - \sigma_Y^T)(\sigma_1+\sigma_2+\sigma_3) = \sigma_Y^C \sigma_Y^T.\end{aligned} \tag{3.6}$$

In Eq. (3.6) the sequence of principal stresses $\sigma_i \geq \sigma_j \geq \sigma_k$ with $i, j, k = 1...3$ is important and therefore Eq. (3.6) must consider the six different cases displayed in Table 1. Consideration of all the possible combinations of the principal stresses leads to obtaining multi-surface condition.

**Table 1**. The principal stresses – order cases.

| | Ordering of principal stresses | | |
|---|---|---|---|
| 1 | $\sigma_1$ | $\sigma_2$ | $\sigma_3$ |
| 2 | $\sigma_1$ | $\sigma_3$ | $\sigma_2$ |
| 3 | $\sigma_2$ | $\sigma_1$ | $\sigma_3$ |
| 4 | $\sigma_2$ | $\sigma_3$ | $\sigma_1$ |
| 5 | $\sigma_3$ | $\sigma_1$ | $\sigma_2$ |
| 6 | $\sigma_3$ | $\sigma_2$ | $\sigma_1$ |

Substituting all the above cases to Eq. (3.6), ordering its components, allows Burzyński multi-surface criterion in six detailed equations to be obtained. Let us observe that the cases 1 and 6, 2 and 4, as well as 3 and 5 are equivalents. Consequently, the resulting yield criterion is an intersection of three yield surfaces. To better present the idea of intersection of the yield surfaces, the intersection of yield curves in the plane of stresses $\sigma_2 = 0$ is described, Eqs. (3.7).

$$\begin{aligned}
\Phi_1(\sigma_1, \sigma_3) &= \sigma_1^2 - 2(1-\lambda)\sigma_1\sigma_3 + 2(1-\lambda)\sigma_3^2 \\
&\quad + 3\frac{\lambda}{1+\lambda}(\sigma_Y^C - \sigma_Y^T)\sigma_1 + 3\frac{1-\lambda}{1+\lambda}(\sigma_Y^C - \sigma_Y^T)\sigma_3 - \sigma_Y^C\sigma_Y^T = 0, \\
\Phi_2(\sigma_1, \sigma_3) &= \sigma_1^2 - 2\lambda\sigma_1\sigma_3 + \sigma_3^2 \\
&\quad + 3\frac{\lambda}{1+\lambda}(\sigma_Y^C - \sigma_Y^T)\sigma_1 + 3\frac{\lambda}{1+\lambda}(\sigma_Y^C - \sigma_Y^T)\sigma_3 - \sigma_Y^C\sigma_Y^T = 0, \\
\Phi_3(\sigma_1, \sigma_3) &= 2(1-\lambda)\sigma_1^2 - 2(1-\lambda)\sigma_1\sigma_3\sigma_3^2 \\
&\quad + 3\frac{1-\lambda}{1+\lambda}(\sigma_Y^C - \sigma_Y^T)\sigma_1 + 3\frac{\lambda}{1+\lambda}(\sigma_Y^C - \sigma_Y^T)\sigma_3 - \sigma_Y^C\sigma_Y^T = 0.
\end{aligned} \tag{3.7}$$

Each of the above equations is valid for a specific area; their inter-section gives the final formulation of the criterion. The inner contour of the three curves determines the yield condition in the plane. The Eqs. (3.7) are illustrated in Fig. 8.

For $\lambda = 0.5$ the criterion has a paraboloidal shape – the same as for the isotropic Burzyński condition, Eq. (3.2). For $\lambda < 0.5$ the obtained surface is not convex. If $\lambda = 1$ the criterion changes into the Tresca criterion because most parts of the relations analyzed are reduced to zero. Thus, the equations have

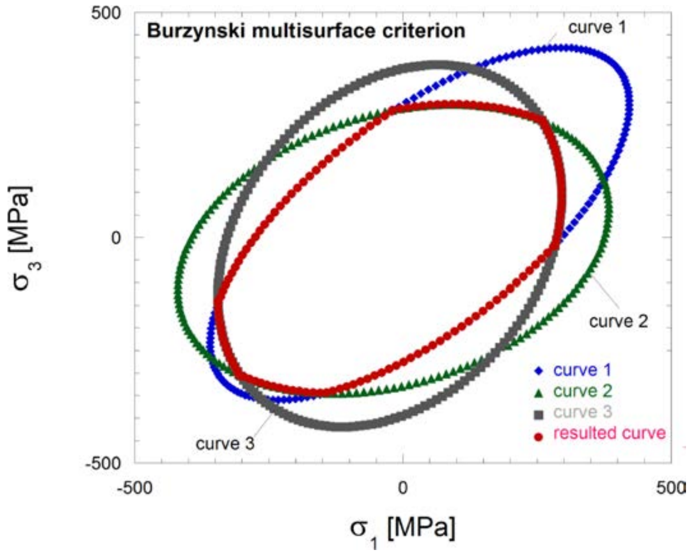



Fig. 8. Representation of elastic domain based on Burzyński multi-surface criterion in the plane $\sigma_2 = 2$, the resultant limit curve is expressed by the red line.

the forms $(\sigma_i - \sigma_j)^2 = \sigma_Y^C \sigma_Y^T$ and $i, j = 1...3$, and they are valid in suitable parts of the system of reference.

The analytical description based on the multi-surface approach, Eqs. (3.7), induces singularities at the intersection points. In such a case, a plasticity theory with so-called corners may be formulated, where the yield function is no longer differentiable, as in the model of the Tresca yield condition [37]. There are algorithms which provide a generalization of plasticity theory to accommodate such singularities, e.g. [55], but such an approach is not convenient for computational applications, mainly because of difficulties in defining the flow rule. Consequently, there is a tendency to round off the corners in the points where the stress is in the vicinity of singularity [56]. This is a reason for a 'smoothing out' curve resulting from the criterion given in Eqs. (3.7). Therefore, derivation of smooth resultant yield surface is proposed. The final formula of the yield criterion can be derived assuming continuity of the first derivatives at the points of intersections [36, 57]. The final formulation of the Burzyński criterion written in principal stress coordinates is given in Eq. (3.8).

$$\sigma_1^2 - R_B \sigma_3 \sigma_1 + \sigma_2^2 + (\sigma_Y^C - \sigma_Y^T)(\sigma_1 + \sigma_3) - \sigma_Y^C \sigma_Y^T = 0, \tag{3.8}$$

where

$$R_B = 2 - \frac{\sigma_Y^C \sigma_Y^T + 2(\sigma_Y^C - \sigma_Y^T)\sigma_Y^{CC}}{(\sigma_Y^{CC})^2} \tag{3.9}$$

and $\sigma_Y^{CC}$ is the yield strength in biaxial compression. If the experimental value of the yield strength in biaxial compression is unknown, then a purely mathematical relation given in Eq. (3.10) is obtained under the assumption that the point belongs to curve (3.8) and the values of abscissa and ordinate of $\sigma_Y^{CC}$ in the plane $\sigma_2 = 0$ are equal to each other. Equation (3.10) allows the value of the yield strength in biaxial compression to be calculated as a relation of the yield stresses in tension $\sigma_Y^T$, compression $\sigma_Y^C$ and a value of the experimentally measured yield strength in shear $\tau_Y$.

$$\sigma_Y^{CC} = \frac{2(\sigma_Y^C - \sigma_Y^T)\tau_Y^2 + \sqrt{4\tau_Y^4(\sigma_Y^C - \sigma_Y^T)^2 + 4\sigma_Y^C\sigma_Y^T\tau_Y^2(-\sigma_Y^C\sigma_Y^T + 4\tau_Y^2)}}{2(-\sigma_Y^C\sigma_Y^T + 4\tau_Y^2)}. \tag{3.10}$$

The final yield surface obtained with use of smoothed out criterion, Eq. (3.8), is an elliptic paraboloid with an axis of symmetry given by the axis of hydrostatic pressure. The cross-section of the yield surface is an ellipse. The $R_B$ coefficient is a relation between results of strength tests – yield strengths in tension, compression, shear and biaxial compression. However, as it is shown in [58], the biaxial compression test is an experimental technique demanding knowledge of many external conditions (friction, pressure, geometrical inaccuracies). It is possible to obtain the yield strength in shear experimentally in an easier way (though, careful analysis is also necessary [45]). Otherwise, with assumption that no shear correction is necessary, $\tau_Y$ may be calculated using Eq. (3.2) or (3.4). Consequently, assuming that yield stress in shear and biaxial compression are the points which belong to the limit curve, the yield stress in biaxial compression $\sigma_Y^{CC}$ can be given as a relation of yield stresses in tension, compression and shear, as it is shown in Eq. (3.10). This purely mathematical relation allows the yield strength in biaxial compression to be calculated, assuming that this point belongs to the limit curve given in Eq. (3.8).

In the next section the discussed criterion is applied for the exemplary bioplastics, presented in Sec. 2. These particular applications allow us to expect that Burzyński criterion may be used to describe the beginning of plasticity for polymers in general. A wide range of examples can be found in [36]. We conclude that macroscopic yielding in the processes driven by shear under an influence of hydrostatic pressure may be described by a single yield function, as the one given in Eq. (3.8).

## 4. Yield state of the cornpole CPR-M2 and PLA/PBAT polymer alloys

In this section the yield states of the bioplastics, i.e. the cornpole CPR-M2 and the blends of PLA/PBAT in different mixing ratio with or without the crosslinking agent, are described through application of the Burzyński criterion, Eq. (3.8).

The yield surface is presented in the stress space ($\sigma_1$, $\sigma_2$, $\sigma_3$) (the Haigh-Westergaard space), a cross-section of the surface in the plane $\sigma_2 = 0$ is also plotted. The limit curve, depicted in the meridional plane ($\sigma_e$, $\sigma_m$), where $\sigma_e$ is the equivalent Mises stress and $\sigma_m$ is the mean stress, shows a trace of the yield surface as a so-called meridional profile, containing hydrostatic axis. Such a representation allows a visual representation of loading states to be given depending on hydrostatic pressure $\sigma_m$. Limit curves are compared with these resulting from the Huber-Mises criterion. The Huber-Mises criterion is presented here as a reference yield criterion, most commonly used in science and industry.

Yield points in tension, compression and shear of the cornpole CPR-M2, identified for the offset strain $\varepsilon_{\text{offset}} = 0.002$, are collected in Table 2. Tests were performed in quasi-static strain rate $\dot{\varepsilon} = 0.001\ \text{s}^{-1}$.

**Table 2**. Yield data of CPR-M2 identified for offset strain $\varepsilon_{\text{offset}} = 0.002$.

| | $\dot{\varepsilon}_{\text{offset}} = 10^{-3}$ | $\text{s}^{-1}$ |
|---|---|---|
| $\sigma_Y^C$ | 22 | [MPa] |
| $\sigma_Y^T$ | 15 | |
| $\tau_Y$ | 9.6 | |
| $\tau_Y^H$ | 10.48 | |
| $\kappa$ | 1.46 | |

In Fig. 3, it can be noted that there is a large difference between levels of stresses for tension and compression. The SD parameter, calculated by using Eq. (3.1), is equal to 1.46. The measured, experimental value of yield strength in shear, $\tau_Y = 9.6$ MPa, differs from the theoretical one, $\tau_Y^H = 10.5$ MPa, calculated according to the Huber-Mises approach – Eq. (3.3). The origin of such difference may be contributed to the brittle behavior of the cornpole CPR-M2. The amorphous structure is more sensitive to fracture due to the shear processes. Based on the above-mentioned observations, it may be concluded that the criterion properly describing an onset of plasticity for the cornpole should include the influence of hydrostatic pressure and the experimental value of the yield strength in shear. Values of yield points (collected in Table 2) allow us to describe the yield state of the cornpole. The criterion discussed in Sec. 2 is applied to the experimental data, Eq. (3.8). The yield surface and the yield curves, built based on the experimental data of the cornpole CPR-M2, are shown in Fig. 9a,b. The comparison between limit curves plotted using the Burzyński criterion and the Huber-Mises criterion is depicted in Fig. 9c,d.

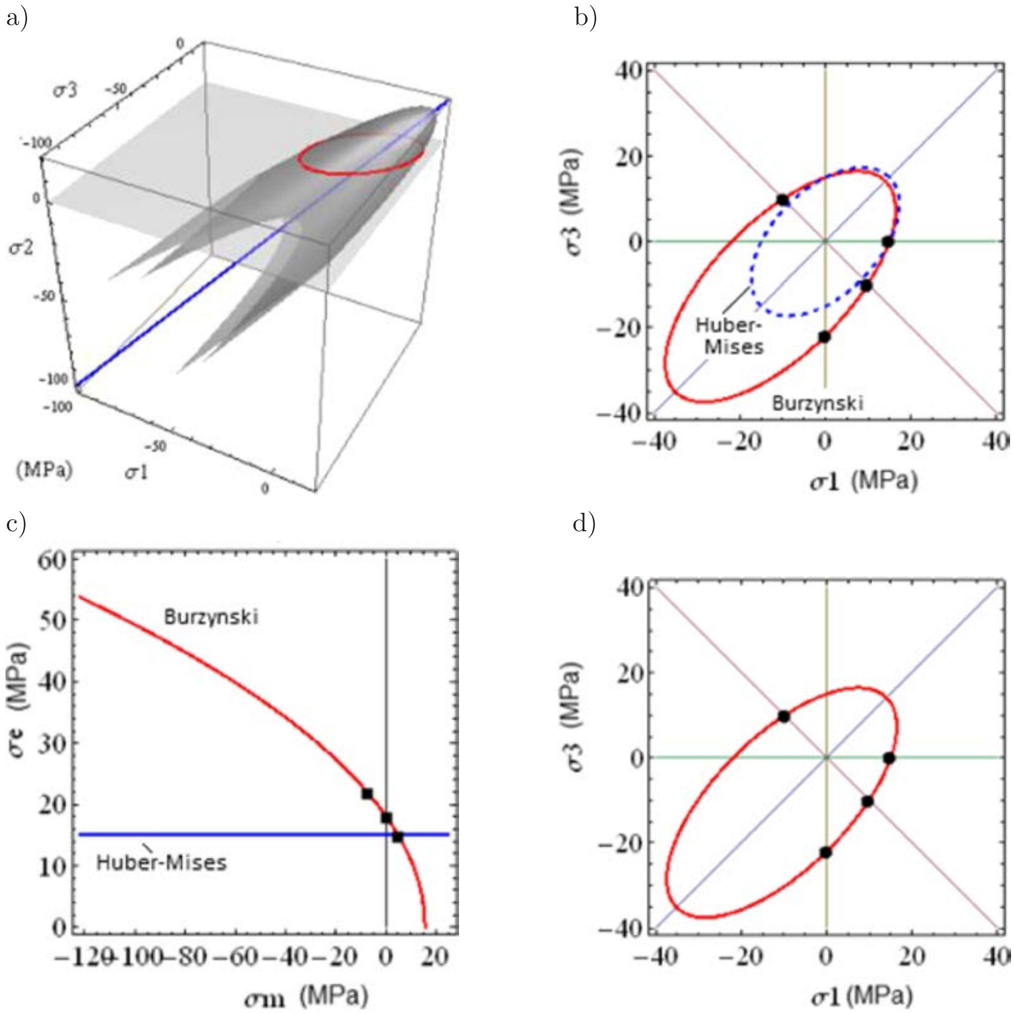



FIG. 9. Visualization of the yield state of the cornpole CPR-M2 according to the Burzyński condition Eq. (3.8) presented in: a) the stress space, b) the plane $\sigma_2 = 0$. Comparison with the Huber-Mises limit curves in: c) the meridional plane, d) the plane $\sigma_2 = 0$.

With reference to Fig. 9, it can be concluded that good agreement is found between experimental results and the analytical description provided by the Burzyński criterion. The difference can be observed for the prediction resulting from the Huber-Mises approach, which is especially visible in a plot given in the meridional plane. The Burzyński criterion described by Eq. (3.8) more properly reflects the differences in the increase of yield strength under the increasing hydrostatic pressure The largest difference between the Burzyński criterion and the Huber-Mises criterion is observed in the third quadrant of the plane $\sigma_2 = 0$.

This observation emphasizes an assumption that the biaxial compression state may be of essential importance in the description of the onset of plasticity.

Data which characterize PLA/PBAT polymers in different proportions with or without the addition of crosslinking agent in dynamic and quasi-static strain rates are collected in Table 3. The yielding points are identified for the beginning of inelastic strain equals to $\varepsilon_{\text{offset}} = 0.002$.

**Table 3**. Comparison between SD parameters $\kappa$ calculated for PLA/PBAT polymers in different proportions of components.

| Components | Quasi-static | Dynamic |
|---|---|---|
| | $\kappa$ | $\kappa$ |
| 60:40 | 2.04 | 3.4 |
| 60:40:1 | 1.9 | 2.6 |
| 70:30 | 1.78 | 3 |
| 70:30:1 | 1.79 | 3.27 |
| 80:20 | 1.68 | 3.52 |
| 80:20:1 | 1.86 | 3.34 |

Comparing values presented in Table 3, it can be concluded that at low strain rates the SD parameter decreases with the increasing content of PLA compound. Regarding the SD parameters calculated for blends with dialkyl peroxide addition and without it, it is observed that the SD parameter is almost unchanged for both types of blends. In the case of the results obtained under dynamic loadings, it can be noted that the value of the SD parameter increases with increasing amount of PLA component for polymers with and without the addition of the cross-linking agent.

Figure 10 plots a comparison between yield curves in the plane $\sigma_2 = 0$ and in the meridional plane for the quasi-static and dynamic ranges of strain rates. Based on the results for the cornpole, it seems that PLA/PBAT alloys could be sensitive to failure caused by shear loading. There is no data about the shear behavior of PLA/PBAT blends, it is assumed that the yield point in shear has a theoretical value and fulfils the relation given in Eq. (3.4).

To assess the yield state, Burzyński criterion with assumption that $R_B = 0.5$ is used. In Fig. 10, the yield curves for PLA/PBAT polymers of different proportions and for dynamic and quasi-static strain rates are plotted. The curves for the bioplastics with addition of dialkyl peroxide are indicated by a dotted line.

With regard to Fig. 10, it is noted that for the quasi-static strain rates, the addition of dialkyl peroxide has a negligible influence on the resulting yield curves. For dynamic strain rates, however, the addition of the cross-linking agent

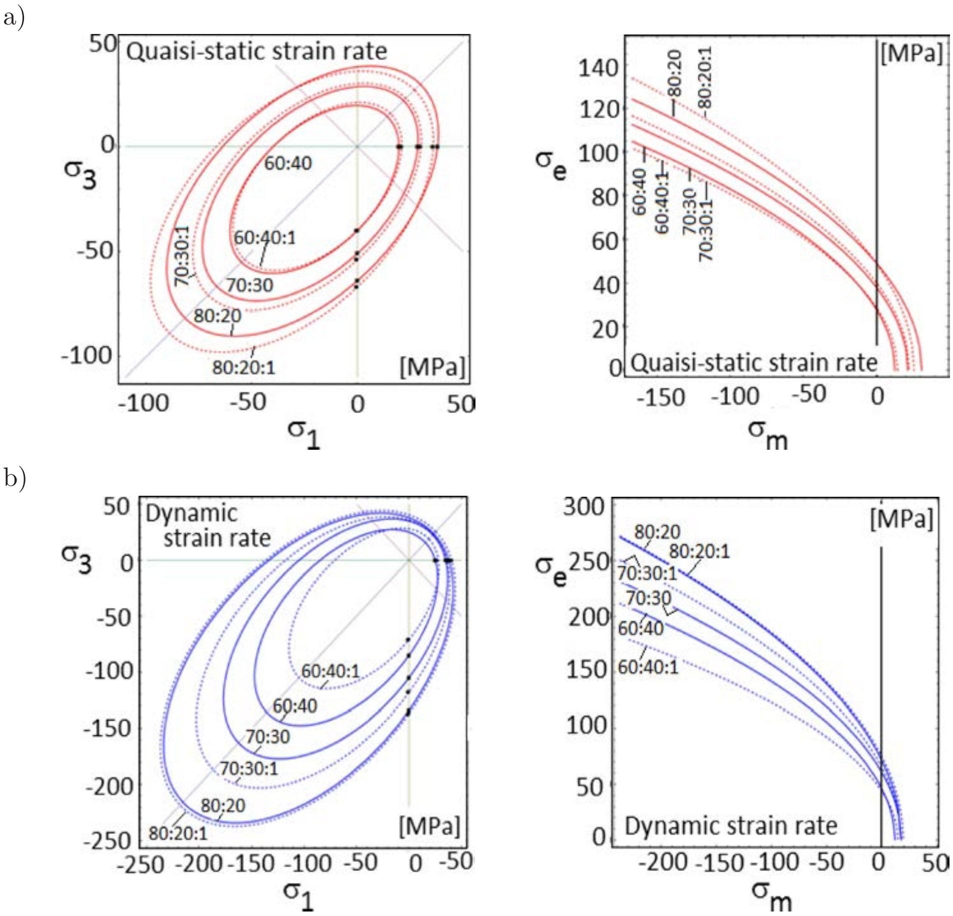



FIG. 10. Yield curves in the plane $\sigma_2 = 0$ and the meridional plane for polymers with different proportions of PLA/PBAT at: a) quasi-static strain rate, b) dynamic strain rate. Polymers with addition of dialkyl peroxide are indicated by dotted curves.

has a noticeable influence, especially in the case of the compressive stress states. A decrease in PBAT component results in more resistant material, especially for the compressive loadings. In Fig. 11, the comparison of the yield curves in the plane $\sigma_2 = 0$ and in the meridional plane is shown for the exemplary blend of PLA/PBAT 70:30 without addition of the cross-linking agent.

The largest discrepancy is seen regarding the strain rate in the compressive range of the stress state. A huge difference between values of yield limits in compression can be observed in comparison to a relatively small difference in values of yield limits in tension (in dynamic ranges the strength of the material is almost 3 times higher than in quasi-static ranges). The plots confirm the observation that the resistance of bioplastics to tensile loading is not high and

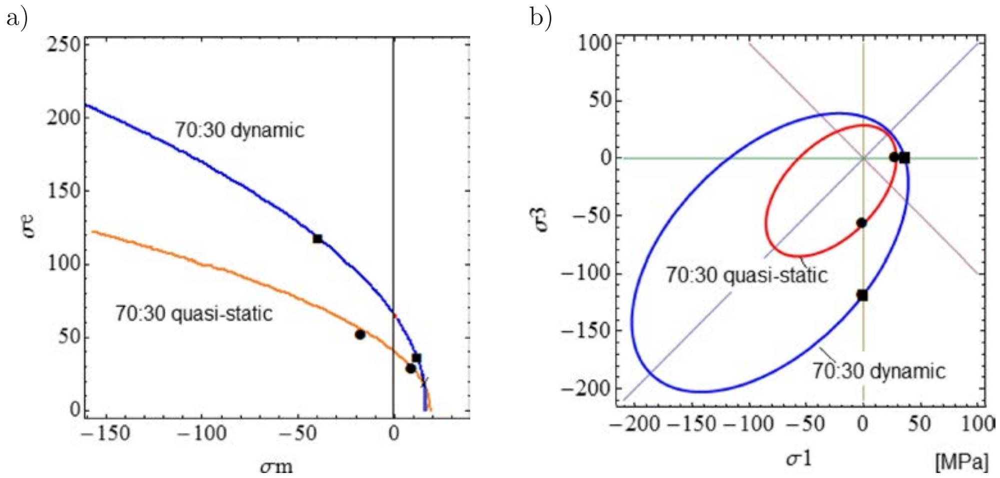



FIG. 11. The comparison of the yield curves in the plane $\sigma_2 = 0$ (a) and in the meridional plane (b) for the exemplary blend of PLA/PBAT 70:30 without addition of the cross-linking agent.

much lower than the resistance to compressive loading. The strength in tension is almost insensitive to the strain ratio of the loading process.

## 5. Conclusions

In the case of bioplastics, i.e. the cornpole CPR- M2 and PLA/PBAT blends, the pressure condition becomes more pronounced under the influence of the more compressive stress state. The asymmetry of elastic limits is characteristic for both bioplastics; the SD parameter in the case of the cornpole is equal to 1.46, whereas for the PLA/PBAT blends it varies between 1.7 and 3.5 depending on the compounds and the strain rate. For the cornpole, the measured value of the yield point in shear varies from the theoretical one, Eq. (3.3).

In the discussions in the paper, the Burzyński criterion [3, 4], the SD effect and the influence of shear on yielding are incorporated. In addition, its formulation is based on the additive character of elastic energy, which makes the Burzyński criterion grounded on the fundamental principles of material mechanics. In consequence, it is possible to predict the yield state of polymers which are assumed to be liable to the above-mentioned effects. It can be observed that the Burzyński criterion allows us to obtain good correlation between the predicted yield state and the experimental results, as confirmed by the visualizations of the yield states of the cornpole and the PLA/PBAT blends given in Figs. 9, 10. The criterion is relatively easy to apply (only 3 material data are needed – $\sigma_Y^T$, $\sigma_Y^C$, $\tau_Y$), which speaks in favor of its application for a wide variety of materials and for polymers in particular.

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