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Mechanical characterization and modeling of cyclic and time dependent behavior of a soft 3D printed photopolymer

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Abstract

TangoBlackPlus © (TB+) is a 3D printable photopolymer frequently employed in advanced materials, as additively manufactured biomimetic composites and metamaterials, or soft actuators based on shape memory effects. Thorough knowledge of its mechanical response is essential to identify constitutive laws for the modelling of innovative devices or fine tuning of simulations of the fracture process. In the present work we investigated the response of TB+ under different types of mechanical loading, including uniaxial tension at different strain rates, stepped stress relaxation and cyclic imposed loads. The experimental results were used to compare the predictive capabilities of different visco-plastic models. Among these, the Bergstrom-Boyce (BB) model showed the highest accuracy, as also verified through FE implementation, allowing prediction of large deformation, time-dependent response and residual strain resulting from cyclic loading.

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1. Introduction

TangoBlackPlus © (TB+) is a flexible elastomer available to PolyJet © technology, an additive manufacturing (AM) process based on photopolymer jetting. This technology involves the deposition of droplets of photopolymer resins onto a print bed, followed by curing with ultraviolet (UV) lamps to form a polymerized layer. Then, similarly

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to other AM methods, the part is built layer-by-layer, with the peculiar advantage that the production of multi-material components is also possible, as reported in the review by Patpatiya (2022). The elastomeric nature of TB+ makes it particularly attractive for many advanced applications, including soft robotic and actuators, soft modules for logic gates and computation, and biomedical devices, as for example described, among other works, by Dämmer (2019), Kamrava (2022) and Khalid (2020). Shape memory ‘4D materials’, as well as different configurations of metamaterials, were instead printed by Ding (2017), Dalaq (2023) and others. Biomimetic composites represent a further active field of research where TB+ can be used. AM processes such as Polyjet allow researchers to mimic, with 3D-printed materials and composites, the complex and specific hierarchical organization of natural materials. Composites consisting of stiff and soft constituents were designed by Wang (2023) to improve energy absorption capabilities, whereas Libonati (2017) and Aguilar Coello (2023) developed novel damage-tolerant architectures by replicating the nacre’s toughening mechanisms with a combination of stiff and soft polymers and a proper 3D printed spatial architecture. In this context, flexible materials like TB+ play a key role, serving as a matrix to provide support for large strains, dissipating big quantities of energy when mechanical deformation is applied, and acting as a viscoelastic glue that contributes to the peculiar fracture mechanisms of these composites. Due to the complex nature of the above-mentioned applications, accurate material characterization and modeling are especially relevant to anticipate the response of sophisticated devices or to tune specific properties of a 3D-printed material architecture, so to reduce time and cost of the prototyping phase. This is particularly true for elastomeric materials like TB+, in which the ability to undergo large deformations may be associated with time-dependent properties, as a consequence of their viscous nature, or may change under cyclic loading. Different approaches were reported in literature for characterization and modelling of the mechanical behavior of TB+, depending on the goal and type of problem. The most straightforward modelling strategy consists in considering only the quasi-static response, neglecting any time-dependency effect, assuming a linear elastic model, as done by Shen (2014), or hyper-elastic response to account for large deformations, as in the papers by Dämmer (2019) or Wickramasinghe (2023). The use of time-independent hyper-elastic laws has also been shown to be appropriate for modelling quasi-static response of composites based on TB+ matrix, including fracture processes, as assumed in Aguilar Coello (2023). However, Slesarenko (2018) noted that the comparison of the different proposed models may lead to some inconsistencies in shear modulus evaluation and that anisotropy in the material may be present, although this is usually neglected. On the other hand, MacCurdy (2016) showed that the mechanical behavior of TB+ is not only non-linear, but also viscoelastic and characterized by strain rate sensitivity and stress relaxation effects. Considering strain-rate, experimental data were reported by Khalid (2020), showing that by increasing the strain rate of testing an increasing stiffness could be noticed. In the same study a reduction in stiffness and tensile strength response was reported with increasing temperature in the range 0°- 50°C. Slesarenko (2018) reported tensile tests on TangoPlus (TP), an elastomer very similar to TB+, in the strain-rate range 1.2×10^{-3} to $1.2 \times 10^{-1} \text{ s}^{-1}$. In this case, the ultimate strain of TP was more than 1.5 times higher when comparing the fastest test with the slowest one. Abayazid (2020) adopted five strain rates between 3×10^{-3} to $3 \times 10^{-1} \text{ s}^{-1}$ in tension and between 5×10^{-3} to $5 \times 10^{-1} \text{ s}^{-1}$ in compression, again for TP. Also in this case, the ultimate strain increased with increasing strain rate, with the material showing a progressively stiffer response. This study included stress-relaxation data, with the stress profiles, normalized by peak stress, showing a remarkable overlap for tests in the range 5 %–20 % of imposed strain. Kundera (2014) investigated stress relaxation at component level, showing that photopolymer O-rings made of TB+ exhibited stiffness relaxation that can be reproduced with simple rheological models. When dealing with elastomeric materials, the influence of cyclic loading should also be considered, since they may exhibit stiffness variations after the first cycle or cyclic softening and changes of hysteresis cycles, as noted for TPU elastomers by Avanzini (2011). At present the only investigation reported in literature for Tango materials family is the study by Abayazid (2020), in which samples were compressed three times, eventually incorporating in the sequence a resting interval. The results of cyclic tests up to 0.6 strain did not show any permanent plastic deformation and highlighted a marked difference between loading and unloading path, the latter being less sensitive to cycling. Notably, long term behavior in relation to fatigue damage was instead studied in Moore (2015). Overall, for TB+ tests at different strain rates, stress-relaxation and cyclic response have not been investigated yet.

From a modelling point of view, Slesarenko (2018) proposed for TP a Quasi-Linear Viscoelastic (QLV) model, which combined hyper- and visco-elasticity phenomena, reporting good accuracy when using instantaneous Yeoh type strain energy density function. Abayazid (2020) followed a similar approach, but combining an Ogden-Type strain energy density function with a convolution integral to describe linear viscoelastic stress response, introducing a

relaxation function fitted with a Prony series. The model captured accurately the stress relaxation phenomena, whereas reproduction of strain-rate effects seemed less satisfactory. Xiang (2019) implemented instead a physically based visco-hyperelastic constitutive model which captured well strain rate sensitivity. However, none of these studies reported information about the incorporation of these sophisticated approaches into FEM code, which may represent a serious limitation when the focus is shifted from material modelling to component simulation. To the best of authors' knowledge, no attempt to model the cyclic behavior of TB+ or TP has been reported in the literature yet. To overcome these limitations, in the present study we adopted a more extended test protocol, capable of providing data useful for evaluating predictive capabilities in different scenarios. The mechanical response of TB+ was first investigated with tensile tests at different strain rates. Time-dependency was further investigated with a step-relaxation test with increasing strain level. Then, tests with consecutive load cycles or stepped load cycles were carried out to investigate the influence of cyclic loading. The results of this experimental campaign were then fitted using the Bergstrom-Boyce (BB) model (Bergstrom, 2015), which has been shown to be potentially suitable to capture time-dependent or cyclic related effects in elastomer-like materials.

2. Materials and Methods

2.1. Material, Specimens and Test Protocol

TB+ is a rubber-like proprietary material developed by Stratasys. The samples tested in the present research were produced using an Objet500 Connex 3 multi-material 3D printer, as described in a previous work by some of the authors (see Aguilar Coello (2023)) where biomimetic composites were investigated. The printer is based on PolyJet © technology, which makes use of two different printing heads to print different materials, if necessary, supplied as liquids inside cartridges. The polymer is then immediately cured by ultraviolet light, with a typical layer thickness of about 15 μm . Miniature dogbone shaped specimens were cut from plates and used for uniaxial and cyclic tests. Relevant dimensions of the specimens are detailed in Fig. 1(a), for all specimens the thickness was 2.5 mm. All the tests were carried out using in-house built test rig, specifically designed to test soft materials under uniaxial or biaxial loading. The test bench, described in detail in Avanzini (2016), consists of four independent linear actuators that can be moved with a load or displacement/speed control, with load accuracy lower than 0.05 N and displacement resolution lower than 1 μm . The test bench is equipped with an optical strain measurement system based on digital tracking of markers placed on the surface of the specimen. The software for bench control and optical measurements was developed within a NI Labview real-time environment. After the tests, another piece of code developed via NI Labview by the authors (see Pola (2019)) was used to process all the images, extract the displacements of the markers and calculate the stress and strain outputs. For all the tests carried out in displacement control, the corresponding strain rate was estimated based on the optically measured displacements of the markers.

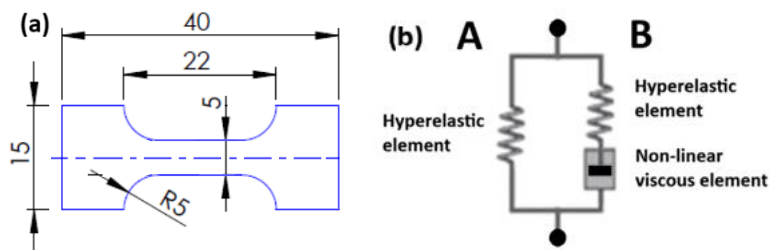


Fig. 1. (a) Specimen for uniaxial tests, (b) Rheological model for Bergstrom-Boyce approach

2.2. Bergstrom-Boyce model and calibration

Polymeric materials are mostly characterized by elastic and viscous (time dependent) responses when loaded. The simplest way to model this behavior is through linear visco-elasticity, as previously done by Slesarenko (2018) and Abayazid (2020). On the other hand, visco-plasticity is the most accurate material model framework available to

represent the mechanical response of all polymers to capture the viscous time-dependent response (Bergstrom 2015). In particular, the BB model has been reported to provide great accuracy for predicting the non-linear time-dependent, large-strain behavior of elastomer-like materials (Bergstrom, 2001), including traditional engineering rubbers and soft biomaterials. As schematically shown in Fig. 1(b), the basic foundation of the BB model can be represented by two interacting parallel networks, A and B, in which the key difference with linear viscoelasticity is that viscoelastic flow response of network B has to be non-linear. Details on theoretical background and model framework can be found in (Bergstrom 2015). The calibration of the model requires determining nine material parameters and testing the material at different strain rates, preferably with loading-hold-unloading cycles (Bergstrom, 2015). The test protocol thus included the following experiments:

- Uniaxial tension at different strain-rate ($5 \times 10^{-3} \text{ s}^{-1}$, $1 \times 10^{-1} \text{ s}^{-1}$, $2.25 \times 10^{-1} \text{ s}^{-1}$, 1 s^{-1})
- Stepped stress relaxation (uniaxial, displacement control)
- Repeated cyclic at a fixed stress level (uniaxial, load control)
- Cyclic step loading at increasing stress level (uniaxial, load control)

Given the complexity of the constitutive model and the number of material parameters to be determined, the calibration is not trivial. Fitting of material parameters was carried out using the software MCalibration (Veryst Engineering, Needham, MA).

Finally, it should be noted that by means of the associated Polyumod library, the results of material calibration can be transferred to FEM commercial codes as subroutines to be used as if they were built into the FE program.

3. Results and Discussion

3.1. Experimental tests

The results of uniaxial tensile tests at different strain rates and biaxial tests are summarized in Fig. 3, showing that the material has a highly non-linear behavior, with a globally stiffer response as strain rate increases and an ultimate tensile strength of 0.9 MPa, in line with the results reported by Khalid (2020), Slesarenko (2018) and Abayazid (2020).

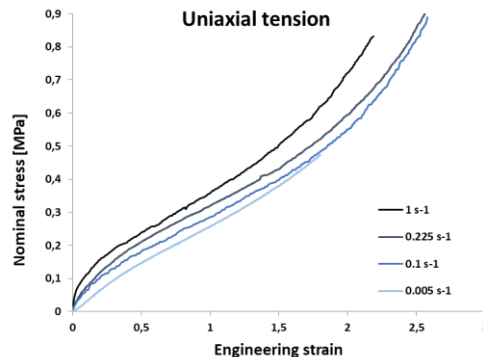


Fig. 3. Uniaxial tensile tests at different strain rates

In comparison, we found slightly higher values of the ultimate strain, in the range 1.8 – 2.55 against 1.3 – 2.25 of Slesarenko (2018) and 1.1-1.8 of Abayazid (2020) works. In the same studies, the ultimate strain of TP was also reported to increase with strain rate. This effect can be noticed also in the present study for the range of strain rates between $5 \times 10^{-3} \text{ s}^{-1}$ and $2.25 \times 10^{-1} \text{ s}^{-1}$, whereas for the test with higher strain rate (i.e. 1 s^{-1}), the ultimate strain at failure is instead lower (2.20).

The results of cyclic tests for the different control modes and time histories investigated are reported in Fig. 4.

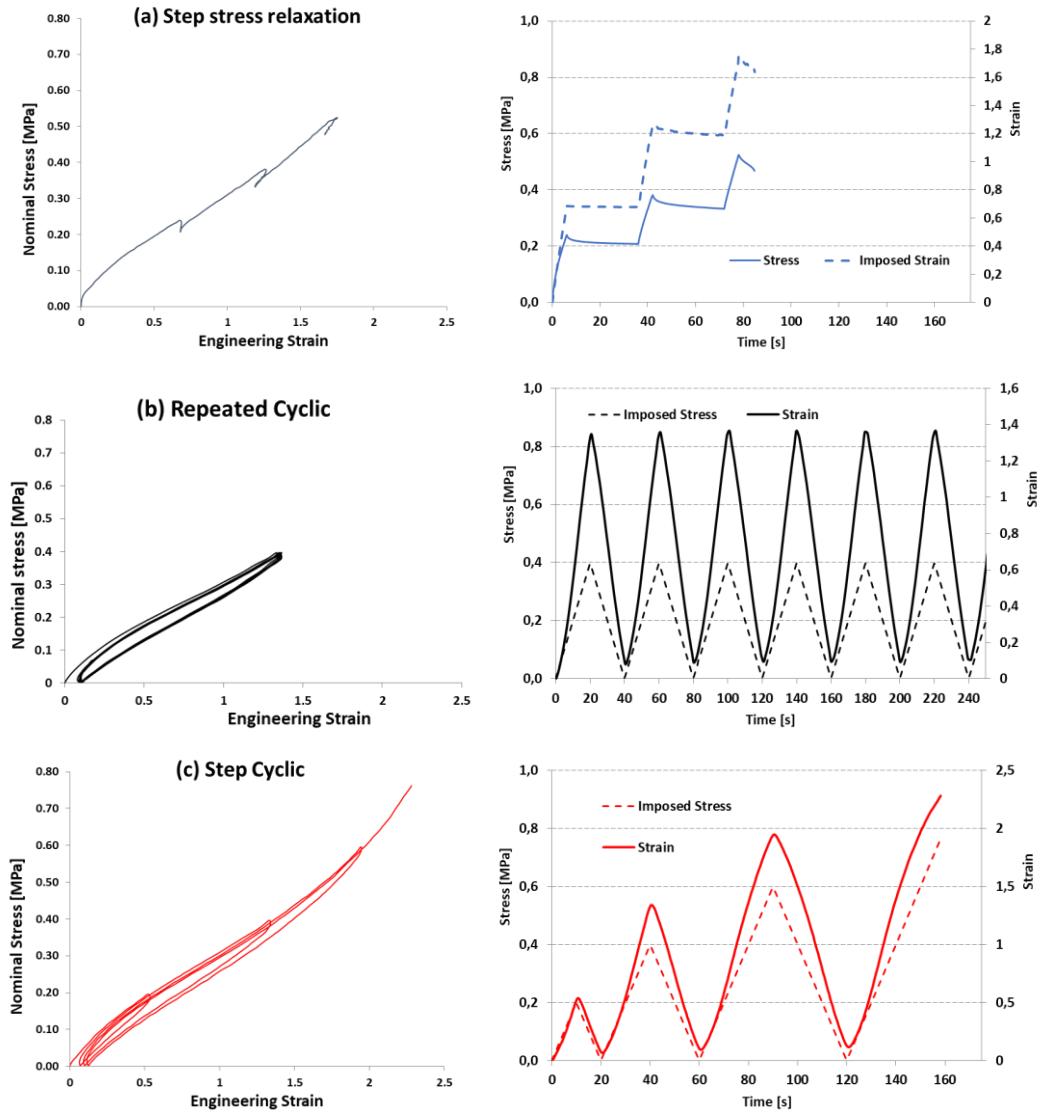


Fig. 4. (a) Step stress relaxation, (b) Repeated cyclic tests, (c) Step cyclic

All the tests terminated with specimen failure. Considering step stress relaxation (Fig 4(a)), under a fixed imposed displacement, the material clearly exhibits stress relaxation when the strain reaches the prescribed level. By comparing the relaxation behavior at different strain level, the stress decrease from the peak value reached at end of the displacement ramp, is more marked for increasing strain. Unfortunately, for the highest strain level the specimen broke before reaching an asymptotic stress, but these results suggest that a non-linear viscous response is present and should be accounted for when modelling. This behavior is different than what reported in Abayazid (2020), probably because in that study the stress relaxation was considered only up to a maximum strain of 0.2, much lower than the present study. The repeated cyclic test reported in Fig. 4(b) showed that the material exhibited hysteresis, undergoing limited softening in the first cycle, after which a stable response is soon reached with an almost fixed residual strain value upon unloading.

Considering step cyclic loading Fig 4(c), under incremental imposed load levels, again the presence of hysteresis can be noticed, as well as a residual strain increasing with the peak value of the stress previously reached. Notably, despite the stepped application of the load, the overall stress-strain response is very similar to that of the virgin material, but

the ultimate strength was lower, with a reduction of about 10% which may indicate that some form of progressive damage occurred. As could be better appreciated in Fig. 5, for the stepped cyclic test with load control, when the applied load is reversed after the first load ramp, hysteresis can be noticed, and the material follows an unloading path different than the loading phase. By applying further cycles, when the load level of the previous cycle is reached the material follows the path of the virgin stress-strain curve. At the end of the loading cycles, when zero load is reached, a residual strain is present, and its values get progressively larger with successive cycles.

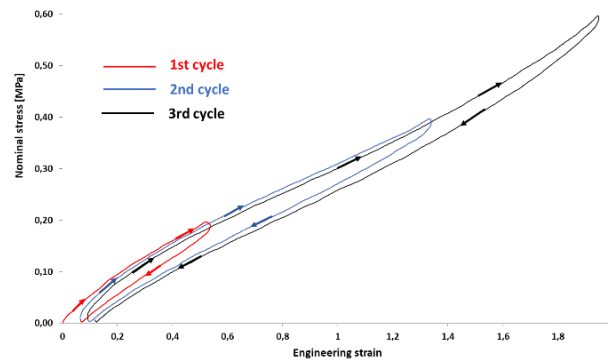


Fig. 5. Loading and unloading paths in cyclic test with increasing load

The presence of a residual strain was also reported in Abayazid (2020), in which case, however, it was completely recovered, when an idle period was included between compressive cycles, suggesting a visco-hyperelastic behaviour rather than visco-plastic. In our case this could not be verified, though the lower values of the ultimate strength of the cycled specimens in comparison with monotonic suggest instead that some damage may have occurred in the cycled material and that visco-plastic behaviour cannot be excluded. The step cycle test also provided the opportunity to gain some insight on the presence of the Mullins effect, often reported in filled and non-filled rubber-like materials. Briefly, as per the review by Diani (2009) the Mullins effect is associated with some general features, in particular a marked softening in the first unloading cycle. For subsequent loading, the stress-strain curve can follow the first unloading curve or a different path. If the material is subsequently loaded to a strain beyond the previous maximum strain, the loading curve follows the path of virgin loading. Despite the limited number of cycles, the behavior reported in Fig. 5, where arrows denote the loading/unloading path, supports the conclusion that a mild Mullins effect is present, but probably negligible when modelling. Overall, the tests provided evidence that in presence of cyclic loading time-dependency and hysteresis should be considered and that a visco-plastic model may be useful to account for the presence of some irreversible deformation.

3.2. Model calibration

The results of the calibration of BB model on all datasets are summarized in Fig. 6, where material parameters are also listed (see Bergstrom (2015) for definitions and physical meaning). Overall, considering the complexity of the input data, the BB model captures the time-dependent and cyclic response of TB+ remarkably well. In particular, the prediction is indeed accurate when considering stepped stress relaxation, in which case the non-linear viscoelastic response at different strain levels is captured, as well as cyclic response. Considering strain rate effects, the model captures the main features of the mechanical behavior of TP+, but with some limitations. In particular, the accuracy of the fit decreases as the strain increases and the model fits very accurately only the tensile test at higher strain rates. It should be noted that by feeding the fitting algorithms with more experimental datasets, the accuracy of the prediction under different loading conditions potentially improves, but reaching convergence with a unique set of material parameters for all the different experimental tests can prove very difficult.

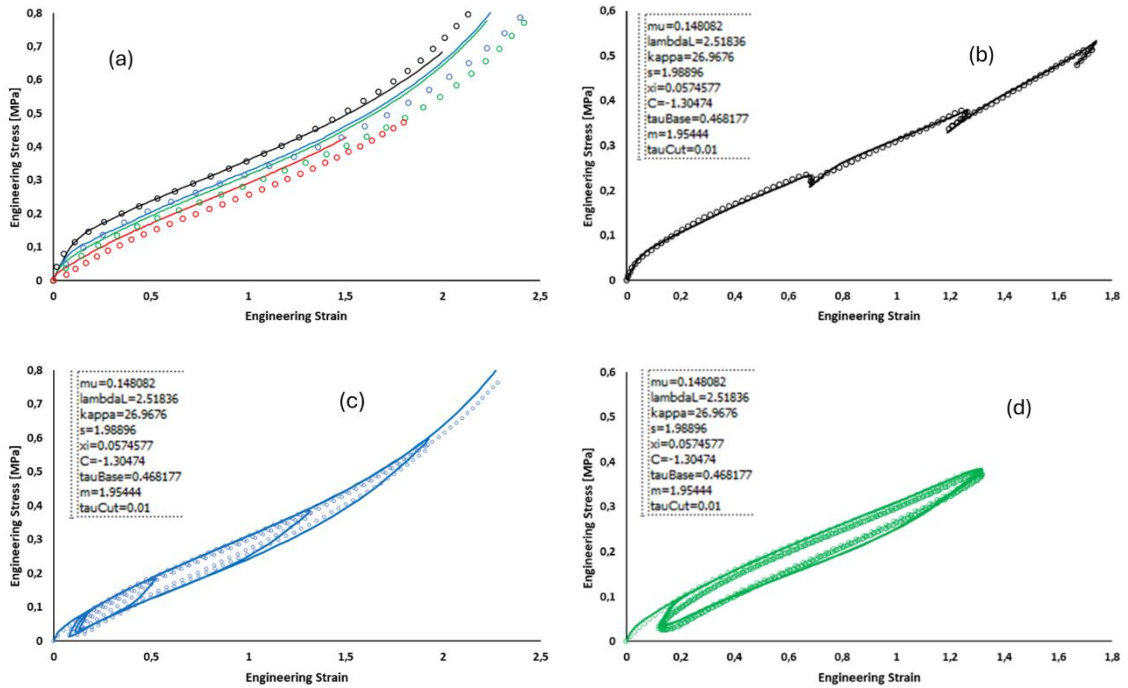


Fig. 6. Fitting of experimental data with BB model a) Tensile test, b) step relaxation c) Step cyclic, d) Repeated cyclic

Finally, a finite element model including BB constitutive law was also implemented in the commercial code ABAQUS rel. 2022 for validation purposes. Model predictions were compared with the results of a biaxial test experiment carried out in a previous study (Aguilar Coello (2023)) on cruciform specimens. As shown in Fig. 7a,b), where taking advantage of symmetries only a quarter of the sample is considered, the model correctly predicts the evolution the reaction force as a function of the time. On a side note, previous biaxial tests did not highlight anisotropy in the planar directions (i.e. perpendicular to printing direction).

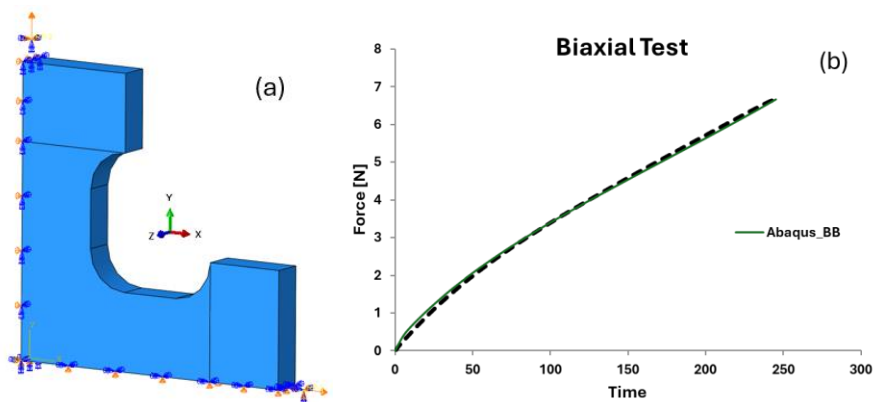


Fig. 7.a) FEM model of biaxial test on TB+, b) Comparison between model predictions and experimental data for biaxial test

4. Conclusions

The set of experimental tests carried out on TB+ showed that the material has a nonlinear stress-strain response influenced by the strain rate. By increasing the strain rate the material response is globally stiffer, but the ultimate strain seems to increase. Step-relaxation test highlighted some dependence of the relaxation process on the applied strain level, suggesting the presence of nonlinear viscous flow. Cyclic tests showed mild softening, presence of a Mullins' effect and of residual strain. Overall, this complex behavior could be satisfactorily represented by means of BB model. FE implementation of the model also provided good results when compared with previous equibiaxial tests. Future developments will involve usage of the BB model to verify the possibility to improve the accuracy of fracture process simulations by XFEM enrichment, as previously done in a previous work (see Aguilar Coello (2023) but with a different constitutive model.

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