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Bárány T., Izer A, Menyhárd A.

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# Reprocessability and melting behavior of self-reinforced composites based on PP homo and copolymers

Tamás BÁRÁNY[1*], András IZER[1], Alfréd MENYHÁRD[2]

[1] Department of Polymer Engineering, Budapest University of Technology and Economics, H-1111 Budapest, Műegyetem rkp. 3., Hungary.

[2] Department of Plastics and Rubber Technology, Budapest University of Technology and Economics, H-1111 Budapest, Műegyetem rkp. 3., Hungary.

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* Author to whom correspondence should be addressed,

E-mail: barany@pt.bme.hu

**Abstract**

In our present work the reprocessability of a self-reinforced PP composites (SRPPC) prepared by compression molding was studied. The composite materials (handled separately, based on the related matrix material) were ground, than extruded five times and injection molded after the first and fifth cycle in order to investigate the behavior of the material during reprocessing. As a reference, the matrices of the composites were also reprocessed and injection molded similarly to the composites. On the manufactured specimens static (tensile and flexural) and dynamic mechanical tests (Charpy) were performed. The melting and crystalline characteristics were studied by Differential Scanning Calorimetry (DSC). The probable decomposition caused by multiple extrusions was followed by the Melt Volume Rate (MVR). The results indicated that in the case of commercial materials there is no significant degradation even after multiple reprocessing cycles; therefore the reprocessability of SRPPC products has no hindrance. The presence of α-iPP reinforcement in the rPP based composites after reprocessing results in increased inclination for crystallization and consequently leads to improved mechanical stiffness compared to rPP neat matrices

Keywords: PP, reprocessing, self-reinforced composite, β-PP

## Introduction

The recyclability of polymers and their blends and composites has become very important issue nowadays. Its importance is represented by the European Union directive on the end-of-life of vehicles (ELV) [1]. In the automotive industry polypropylene (PP) is one of the most used polymeric materials. PP has excellent price/performance ratio, it has to be filled and/or reinforced in order to compete with engineering plastics. Glass fiber (GF) is commonly used to improve the properties of PP. Nevertheless, the recycling of the PP-GF composites is difficult. In order to fulfill the aforementioned directives, two possibilities are open: to improve the recycling technologies in order to be able to recycle more difficult systems or to develop materials which can be reprocessed easily. For the latter a very promising alternative is to develop self-reinforced polymer composites (SRPC – the reinforcement and the matrix belong to the same polymer family but differing in their melting temperatures), because these composites may be used after reprocessing as a one component material. SRPCs have been prepared form many polymers, although PP has become widespread in SRPC applications yet. Any kind of polymer is available for producing SRPCs, which can be processed in fibrous form.

To produce SRPCs suitable processing window (melting temperature difference between matrix and reinforcement) is necessary. It can be obtained by moulding the polymer fibres themselves (first only the skin of the fibre melts to become matrix and the core (reinforcement) remains oriented), called hot compaction, as developed by Ward and Hine [2-4]. This is a really one-component system, so it can be reprocessed without any difficulties. The processing window during composite manufacturing can further be increased by selecting matrix materials having lower melting temperature than the reinforcement (but differing only slightly in the chemical composition – random PP copolymer (rPP)). This is the basis of the consolidation of coextruded tapes, developed by the group of Peijs [5-7]. This combination of two PP materials can also be processed by film-stacking method [8-15]. As a result of reprocessing of these two-component systems an iPP/rPP blend is obtained, so its homogeneity and morphology is to be determined [16]. Since PP is a polymorphic polymer, it has three crystalline modifications: monoclinic (α), trigonal (β), and orthorhombic (γ) forms [17-19]. Commercial grades of PP crystallize essentially in the α-form [20]), however β-iPP can be prepared easily in the presence of highly active β-nucleating agents [20, 21]. The melting temperature of β-form is lower than that of the commercial α-iPP, which provides possibility to broaden the processing window of self-reinforced PP composite (SRPPC) products, if α-iPP is embedded as reinforcement in β-iPP matrix. This kind of composite is

really a homocomposite, because of the reinforcement and the matrix differ only in their crystalline forms [13, 14, 22, 23] (Note that analogous approach was also used for polyamide (PA6) based self-reinforced composites [24]). Therefore the expected recyclability of these composites shall be similar to the hot compacted composites.

The reprocessability of polypropylenes was studied by several authors. After several injection molding cycles, the reprocessing may resulted in chain scission, which is mainly caused by heat loading and mechanical shearing [25]. However, no significant change in the molecular architecture occurs during several reprocessing cycles of well stabilized commercial PP grades. With increasing number of cycles or with increasing reprocessing temperature, only a little reduction can be observed in the average molecular mass [26, 27]. Many studies have been reported no change or only a slight decrease in the tensile strength and in molecular mass of the material [28-30]. The impact properties and the elongation at break are improved however, as a consequence of the decreasing stiffness after reprocessing. Additionally, the melt viscosity and the elongation at break also decrease [31].

After reprocessing of reinforced polypropylenes, the mechanical properties (impact strength, flexural strength and modulus) were decreasing due to mainly the fiber breakage. These phenomenons were investigated in the literature [25, 28, 31]. The rheological tests showed the decrease of the viscosity. This is caused by chain scissions and reinforcing fiber breakage induced by reprocessing and grinding.

Since the reprocessing of PP and its composites has been widely published but there are no studies for the reprocessing of SRPPCs according to the author's knowledge. Therefore the goal of this work is to study the reprocessability of the SRPPCs. The composite materials (handled separately based on the related matrix material) were ground, than extruded five times and the properties were studied after the first and fifth extrusion, respectively, in order to investigate the melting characteristics and mechanical properties of the samples during reprocessing. From the granulated materials different specimens were produced by injection molding and tested.

## Materials, their processing and testing

### Materials

Composite specimens prepared and tested previously and cutting scraps were used in our present study [13, 14]. The composite sheets are composed of a plain woven fabric as reinforcement (presented in our earlier work [14], composed of highly stretched split PP yarns) and three kinds of PP as matrix material (β form of isotactic PP homopolymer

(H388F); random PP copolymer (R351F), and β form of the latter. The non-nucleated PPs were provided by TVK Co. (Tiszaújváros, Hungary) and exhibited a melt flow index of 8 g(10min)$^{-1}$ (at 230°C and 2.16 kg of load). 0.15 wt% of calcium salt of suberic acid (Ca-sub) was introduced into PP as a selective β-nucleating agent in order to prepare β-nucleated samples [32]. The detailed processing method of all SRPPC specimens was described in our earlier work [13, 14]. Thin film was extruded as matrix material and a woven fabric composed of highly stretched PP tapes was applied as reinforcement. Table 1 contains the melting temperature of the components.

Composite sheets with a thickness of 2.5 mm and a nominal reinforcement (i.e. α-PP fabric) content of 50 wt% were produced by compression moulding of a film-stacked package at 7 different processing temperatures. They are selected at 5, 10, 15, 20, 25, 30 and 35°C above the relevant matrix melting temperature. For the latter the Differential Scanning Calorimetry (DSC) melting peak was considered (cf. Table 1). Due to the different processing temperatures, the consolidation quality of the resulting composites was also different. The tested (failed) specimens and cutting scrap were collected separately according to the matrix material.

**Grinding and extrusion**

The collected tested composite specimens and cutting scrap were ground with Plastics Machinery Granulatore GRS 152_302 grinder. The ground composite material was inhomogeneous; significant amount of reinforcement material from the poorly consolidated SRPPCs was debonding from the matrix. Therefore the particle size distribution was also inhomogeneous.

The ground material was extruded five times with a Brabender Plasti-Corder PL 2100 twin screw extruder and then granulated. The extrusion temperature were set to 190°C, 195°C, 200°C, 200°C from the feeder to the die. The screw revolution 8 min$^{-1}$ for the ground material (first cycle) and 25 min$^{-1}$ for the granulated material (2-5$^{th}$ cycle) and for matrix materials was set. To compare, the original matrix materials were also extruded five times.

**Injection molding**

From the granulated materials after the first and fifth extrusion different (tensile and Charpy) specimens were manufactured on an Arburg Allrounder 320C 600-250 injection molding machine. Table 2 contains the injection molding parameters. The temperature zones were set to 185°C, 195°C, 200°C, 205°C, 210°C from the feeder to the mold. The injection molded materials are listed in Table 3. Note that composite materials with random PP copolymer

matrix (regardless of its crystalline form) became a blend due to the reprocessing at higher temperature (considerably higher than the melting temperature of the reinforcement). However, referring to the initial materials, we will further designate it as composite.

**Specimens and their testing**

To obtain the flowability and the degradation, Melt Volume Rate (MVR) test were performed on the materials after each process cycle on CEAST Modular Melt Flow 7027.000 machine. The test parameters were the follow: load: 2160 g, holding time: 240 s, temperature: 230°C, sample mass: 8 g.

The crystallization and melting characteristics of the specimens were studied by DSC. The melting curves were recorded by a Pelkin Elmer DSC 7 apparatus. Samples having a mass of 3-5 mg were scanned at 10°Cmin$^{-1}$ heating ($v_h$) and cooling ($v_c$) rates. In order to erase thermal and mechanical history, the samples were heated up to 220°C and held there for 5 min. Since β-form of PP samples cooled to below 100°C, recrystallize into the α-form during the partial melting of the β-phase [33], the end temperature of cooling ($T_R$) was set to 100°C during non-isothermal crystallization. This setting prevents βα-recrystallization, so the polymorphic composition of the blends can be determined accurately from the melting curves [20, 21].

Static tensile tests were performed on dumbbell specimens (according to standard ISO 527:1999) using a Zwick Z020 universal testing machine with a crosshead speed of 5 mm min$^{-1}$.

Charpy impact tests were performed on Type 1 notched rectangular specimens (according to standard ISO 179:2001) on a Ceast Resil Impactor Junior P/N 6963.000 using the following settings: starting angle of the hammer: 150°; maximum energy: 15 J.

All mechanical tests were performed at room temperature and at least five specimens were tested.

**Results and Discussion**

**MVR**

The MVR of the materials processed once and five times can be seen on Figure 1. One can observe that MVR does not change significantly with increasing of the processing cycles. The more or less constant MVR values hint that the stability of the polymer is good and no thermal degradation occurs during the multiplied processing cycles. In case of ground SRPPCs tested prior to first extrusion, the flowability has considerably higher scatter than after extrusion. The reason is that the particle size of the ground material was inhomogeneous.

The fluidity of the extruded SRPPCs is lower than that of matrices, since the fluidity of the neat material of the reinforcing tape production (foil extrusion grade) is much lower.

## Melting and crystallization characteristics

### Effect of blending

The rPP-based SRPPC is a "two-component" system, because the copolymer matrix contains ca. 3-5 wt% ethylene co-monomers distributed randomly. As a result of the melt reprocessing it is important to know the resulting structure of iPP/rPP blend. The melting curves of α-PP (the neat material of α-PP reinforcement) and α-rPP matrices and the blend of α-rPP matrix and α-PP tapes (α-PP composite based sample) are displayed in Figure 2. The calorimetric traces indicate clearly that one peak appears during the melting and crystallization of composite materials indicating the miscibility of iPP and rPP in the reprocessed α-PP composite based sample. The melting peak at 164 °C relates to α-PP and, α-rPP matrix melts in the vicinity of 145°C. The melting of the blend consist of these two PPs (ca. in 50-50 wt%) is around at 155°C and its melting is reflected by one melting peak. The iPP component has advantageous effect on the crystallization of rPP. iPP and rPP are miscible polymer pair in molten state and the crystallization of the components take place simultaneously. Consequently, only one crystallization peak can be observed during the crystallization of iPP/rPP blends and the higher the iPP content is, the higher is the peak temperature of crystallization.

The melting and crystallization characteristic of β-rPP composite based material after reprocessing is demonstrated in Figure 3. The peak temperature of crystallization ($T_{cp}$) of the α-reinforcement, the β-matrix and the composites is in the vicinity of 112, 104 and 114°C respectively. The $T_{cp}$ of β-rPP matrix is significantly higher than that of the non-nucleated rPP matrix (see in Figure 2a), which unambiguously indicates the presence of the highly active nucleating agent. The presence of Ca-sub results in even higher $T_{cp}$ of the β-rPP composite as well compared to the α-PP reinforcement. The β-nucleated matrix and composites have complicated melting peaks. The double melting peak at lower temperatures refers to the β-form, however the melting peak duplication hints at structural instability [34]. A perfection process within the unstable β-phase results in a ββ'-recrystallization. Similar characteristics are represented by the melting trace of β-rPP composite based specimen, although a peak is shifted toward higher temperatures, because of the presence of iPP in the composites. The doubled melting peaks at 142 and 148°C can be attributed to the ββ'-recrystallization as well, but it has to pointed out that the intensities of the melting peaks of β-form are less pronounced

because of the decreased amount of the nucleating agent. The β-rPP matrix contains 0.15 wt% Ca-sub, while the content of β-nucleating agent in the β-rPP composite based material is only the half of that in the β-rPP matrix.

**The effects of number of reprocessing**

Figure 4 shows the melting and cooling curves of β-PP matrix and β-PP composite based material after reprocessing once and five times. When comparing samples reprocessed once and 5 times one can see that there was no considerable difference due to the cyclic reprocessing for both matrix and composite based specimen.

The β-nucleating agent is still efficient even after five reprocessing steps, because of its good thermal stability, although during injection moulding the related standard for PP was followed, parameters of which did not support the formation of the β-crystals. The crystallization temperatures are at around of 120 °C, which is characteristic for nucleated iPP. Pronounced βα-recrystallization process is reflected on the melting traces (see in Figure 4b) indicating the presence of the β-form. The βα-recrystallization can be eliminated if the samples are not cooled below 100 °C [33]. The melting traces recorded after limited recooling step are represented in Figure 5. One can see that the β-nucleated matrix materials crystallized fully in β-form, but the composites also contain significant amount of α-form due to considerably lower β-nucleating agent content. The reprocessability of the β-nucleating agent is shown only on β-PP composite based samples due to the higher β-content of the matrix. The same tendency can be observed in the case of β-rPP materials. The melting curves do not indicate any degradation after multiplied processing in good agreement with MVR data.

**Mechanical properties**

**Static tensile tests**

The tensile strength (a) and modulus (b) values of iPP and rPP based systems are shown in Figures 6 and 7, respectively. When comparing the results of once and five times processed materials, the mechanical properties of the specimens do not modify considerably, which indicates that no pronounced degradation take place during multiplied reprocessing. Comparing the non-nucleated and β-nucleated matrix materials (both homo- and copolymer), it can be stated that the β-nucleation resulted in a slight decrement in tensile strength and in the Young's modulus, because of the presence of β-form. It should be noted that the β-nucleated matrix material have the lowest stiffness and tensile strength and the properties of the composite based ones is closer to that of the non-nucleated matrix material in the case of

iPP based materials. The relatively large stiffness and tensile strength of the α-PP composite based specimens can be explained by the large α-PP content of the samples (See in Figure 5). This is also true when comparing α- and β-rPP SRPPC based materials. The modulus and tensile strength of rPP composite based specimen (iPP/rPP blends) were significantly higher than those of related matrix material, because of the presence of iPP phase.

From the result it can be stated further that there was no considerable change in the tensile properties if comparing the material processed once and five times.

**Charpy impact tests**

Figure 8 shows the Charpy impact strength results for iPP (a) and rPP (b) based systems. Based on the results it can be seen that the β-modification has improved the Charpy impact strength in case of matrix materials. The increasing of impact resistance is more pronounced in the case of homopolymer [20], because the rPP contains elastomers (PE), which improves the impact resistance as well. In the case of β-PP composites efficiency of the lower amount of β-nucleating agent may be more sensitive for the processing parameters, so the impact resistance did not increase significantly. In the case of rPP composite based samples the lower toughness is resulted by the approximately 50 wt% of α-iPP content (see Figure 8a). It can also be concluded from the test results, that with increasing number of reprocessing, the Charpy impact strength did not decrease considerably.

**Conclusion**

The goal of this paper was to study reprocessability and the melting behavior of the self-reinforced PP composite composed of fabric, woven from highly stretched split PP yarns as reinforcement and α and β crystal forms of isotactic PP homopolymer and random PP copolymer as matrix materials. From the granulated materials different specimens were produced by injection molding and they were tested (MVR, DSC, tensile, Charpy).

Based on the results the following conclusions can be drawn:

- self-reinforced PP composites can well be reprocessed without any loss of properties related to the properties of the matrix materials, thereby it is an excellent candidate for upcycling,
- the cyclic reprocessing (once and five times) did not result in considerable deterioration of the mechanical properties,

- based on the DSC results it can be stated that the used β-nucleating agent (Ca-sub) is still efficient after even five reprocessing cycles. However, it is necessary to apply suitable settings of injection molding.
- the random PP copolymer based SRPPCs (reinforced with PP homopolymer) became a one-phase blend after reprocessing. Their melting temperature and mechanical properties are between those of random copolymer and homopolymer.

**Acknowledgements**

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**References**

[1] Directive 2000/53/EC of European Parliament and of the Council of 18 September 2000.
[2] Ward IM. Developments in oriented polymers, 1970-2004. Plast. Rubber Compos. 2004;33:189-94
[3] Ward IM. Hine PJ. Novel composites by hot compaction of fibers. Polym. Eng. Sci. 1997;37:1809-14
[4] Ward IM. Hine PJ. The science and technology of hot compaction. Polymer 2004;45:1413-27
[5] Alcock B. Cabrera NO. Barkoula NM. Reynolds CT. Govaert LE. Peijs T. The effect of temperature and strain rate on the mechanical properties of highly oriented polypropylene tapes and all-polypropylene composites. Compos. Sci. Technol. 2007;67:2061-70
[6] Alcock B. Cabrera NO. Barkoula NM. Spoelstra AB. Loos J. Peijs T. The mechanical properties of woven tape all-polypropylene composites. Compos. Pt. A-Appl. Sci. Manuf. 2007;38:147-61
[7] Alcock B. Cabrera NO. Barkoula NM. Wang Z. Peijs T. The effect of temperature and strain rate on the impact performance of recyclable all-polypropylene composites. Compos. Pt. B-Eng. 2008;39:537-47
[8] Houshyar S. Shanks RA. Hodzic A. Influence of different woven geometry in poly(propylene) woven composites. Macromol. Mater. Eng. 2005;290:45-52
[9] Houshyar S. Shanks RA. Hodzic A. The effect of fiber concentration on mechanical and thermal proverties of fiber-reinforced polypropylene composites. J. Appl. Polym. Sci. 2005;96:2260-72
[10] Houshyar S. Shanks RA. Morphology, thermal and mechanical properties of poly(propylene) fibre-matrix composites. Macromol. Mater. Eng. 2003;288:599-606
[11] Bárány T. Izer A. Czigány T. On consolidation of self-reinforced polypropylene composites. Plast. Rubber Compos. 2006;35:375-9
[12] Bárány T. Izer A. Czigány T. High performance self-reinforced polypropylene composites. Mater Sci Forum 2007;567-538:121-8
[13] Bárány T. Izer A. Karger-Kocsis J. Impact resistance of all-polypropylene composites composed of alpha and beta modifications Polym Test 2009;28:176-82

[14] Izer A. Bárány T. Varga J. Development of woven fabric reinforced all-polypropylene composites with beta nucleated homo- and copolymer matrices. Compos Sci Technol 2009;69:2185-92
[15] Houshyar S. Shanks RA. Hodzic A. Modelling of polypropylene fibre-matrix composites using finite element analysis. Express Polym. Lett. 2009;3:2-12
[16] Menyhárd A. Varga J. Liber Á. Belina G. Polymer blends based on the beta-modification of polypropylene. Eur. Polym. J. 2005;41:669-77
[17] Padden Jr. FJ. Keith HD. Spherulitic Crystallization in Polypropylene. J Appl Phys 1959;30:1479-84
[18] Varga J. Ehrenstein GW. Schlarb AK. Vibration welding of alpha and beta isotactic polypropylenes: Mechanical properties and structure. Express Polym. Lett. 2008;2:148-56
[19] Varga J. Beta-modification of isotactic polypropylene: Preparation, structure, processing, properties, and application. J Macromol Sci Phys 2002;41:1121-71
[20] Menyhárd A. Varga J. Molnár G. Comparison of different beta-nucleators for isotactic polypropylene, characterisation by DSC and temperature-modulated DSC (TMDSC) measurements. J Therm Anal Calorim 2006;83:625-30
[21] Abraham TN. Wanjale SD. Bárány T. Karger-Kocsis J. Tensile mechanical and perforation impact behavior of all-PP composites containing random PP copolymer as matrix and stretched PP homopolymer as reinforcement: Effect of [beta] nucleation of the matrix. Compos. Pt. A-Appl. Sci. Manuf. 2009;40:662-8
[22] Bárány T. Karger-Kocsis J. Czigány T. Development and characterization of self-reinforced poly(propylene) composites: carded mat reinforcement. Polym. Advan. Technol. 2006;17:818-24
[23] Bhattacharyya D. Maitrot P. Fakirov S. Polyamide 6 single polymer composites. Express Polym. Lett. 2009;3:525-32
[24] Martins MH. De Paoli MA. Polypropylene compounding with post-consumer material: II. Reprocessing. Polym Degrad Stabil 2002;78:491-5
[25] González-González VA. Neira-Velázquez G. Angulo-Sánchez JL. Polypropylene chain scissions and molecular weight changes in multiple extrusion. Polym Degrad Stabil 1998;60:33-42
[26] Xiang Q. Xanthos M. Mitra S. Patel SH. Guo J. Effects of melt reprocessing on volatile emissions and structural/rheological changes of unstabilized polypropylene. Polym. Degrad. Stabil. 2002;77:93-102
[27] Incarnato L. Scarfato P. Acierno D. Rheological and mechanical properties of recycled polypropylene. Polym. Eng. Sci. 1999;39:749-55
[28] Ramírez-Vargas E. Navarro-Rodríguez D. Blanqueto-Menchaca AI. Huerta-Martínez BM. Palacios-Mezta M. Degradation effects on the rheological and mechanical properties of multi-extruded blends of impact-modified polypropylene and poly(ethylene-co-vinyl acetate). Polym Degrad Stabil 2004;86:301-7
[29] Rust N. Ferg EE. Masalova I. A degradation study of isotactic virgin and recycled polypropylene used in lead acid battery casings. Polym Test 2006;25:130-9
[30] da Costa HM. Ramos VD. de Oliveira MG. Degradation of polypropylene (PP) during multiple extrusions: Thermal analysis, mechanical properties and analysis of variance. Polym. Test 2007;26:676-84
[31] Varga J. Mudra I. Ehrenstein GW. Highly active thermally stable beta-nucleating agents for isotactic polypropylene. J. Appl. Polym. Sci. 1999;74:2357-68
[32] Varga J. Melting memory effect of the beta-modification of polypropylene. J Therm Anal 1986;31:165-72

[33] Horváth Z. Sajó I. Stoll K. Menyhárd A. Varga J. The effect of molecular mass on the crystallization tendency and supermolecular structure of the polymorphic modifications of isotactic polypropylene. Express Polym Lett 2010;4:101-4

## Legend of figures

**Figure 1.** The MVR index in the case of PP homopolymer (a), and random PP copolymer (b) systems

**Figure 2.** The cooling (1st cool - a) and melting curves (2nd heat - b) of the α-PP, α-rPP matrix and the reprocessed α-rPP composite

**Figure 3.** The cooling (1st cool - a) and melting curves (2nd heat - b) of the α-PP, β-rPP matrix and the reprocessed β-rPP composite

**Figure 4.** The cooling (1st cool - a) and melting curves (2nd heat - b) of the β-PP materials reprocessed once and five times

**Figure 5.** The melting curves (3rd heat) of the β-PP materials reprocessed once and five times

**Figure 6.** Tensile strength (a) and modulus (b) of the PP homopolymer based systems

**Figure 7.** Tensile strength (a) and modulus (b) of the random PP copolymer based systems

**Figure 8.** Charpy impact strength of the PP homopolymer (a) and random PP copolymer (b) based systems

## Legend of tables

**Table 1.** The melting temperature of the materials

**Table 2.** The injection molding parameters

**Table 3.** The injection molded materials

**Authors' Response to Reviewers' Comments**
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Answers to reviewers´ comments

The authors want to thank the reviewers´ comments which were very useful to improve the quality of the manuscript.

Answers to Reviewers' comments:

English wording needs considerable smoothing

**English wording has been improved.**

all abbreviations should be disclosed two-times, in the abstract and in the main text, respectively

**According to the reviewer's suggestion, it has been corrected.**

it was a pity that authors did not make any polaroptical investigations!

**Since the POM would not provide any additional information concerning the topic of our manuscript, the authors do not want to include micrographs into the paper in order not to exceed the 15 page limitation of the Journal.**

the DSC traces are treated only "qualitateively" - so their amount may be reduced

**In this work, 16 different materials were investigated. The authors demonstrated the changes qualitatively by DSC traces as the reviewer had pointed out. However, we think that the introduced curves are essentially important to represent the concept of the manuscript. Therefore, no changes were made at this point in order to keep the flawless introduction of the most important results.**

Abstract: last sentence is unclear - what was advantageous?

**Probable this sentence was written ambiguously. It has been corrected as follows: "The presence of α-iPP reinforcement in the rPP based composites after reprocessing results in increased inclination for crystallization and consequently leads to improved mechanical stiffness compared to rPP neat matrices."**

Introduction: last sentence in penultimate paragraph - chain scission of the the matrix polymer (should be extended. Refs. 23-24 (p.4 top) are not in line with the corresponding citations.

**According to the reviewer's suggestion, the references have been corrected.**

section 1.1. paragraph above table 1: "7 processing temperaures": so the scrap fractions themselve had different thermal history?

**The polymer composites were manufactured at 7 different processing temperatures (to achieve different consolidation quality). Regardless of the consolidation quality the composite materials (separated according to only the matrix materials) were reprocessed and investigated together.**

section 1.3: Table 2 may be delated and the related info inserted in the text

**Table 2 contains too many data, so to insert them in the text, their perspicuity would lose.**

section 1.6. last sentence on page 8: the last two sentences are fully obscure

**It has been corrected as follows: "iPP and rPP are miscible polymer pair in molten state and the crystallization of the components take place simultaneously. Consequently, only one crystallization peak can be observed during the crystallization of iPP/rPP blends and the higher the iPP content is, the higher is the peak temperature of crystallization."**

Editorial remarks

Our Journal prefers the term of "mass" instead of "weight", so please substitute weight by mass throughout the manuscript. (Example: sample mass, mass/% or mass loss etc.)

**According to the editor's suggestion, it has been corrected**

Figure 1a

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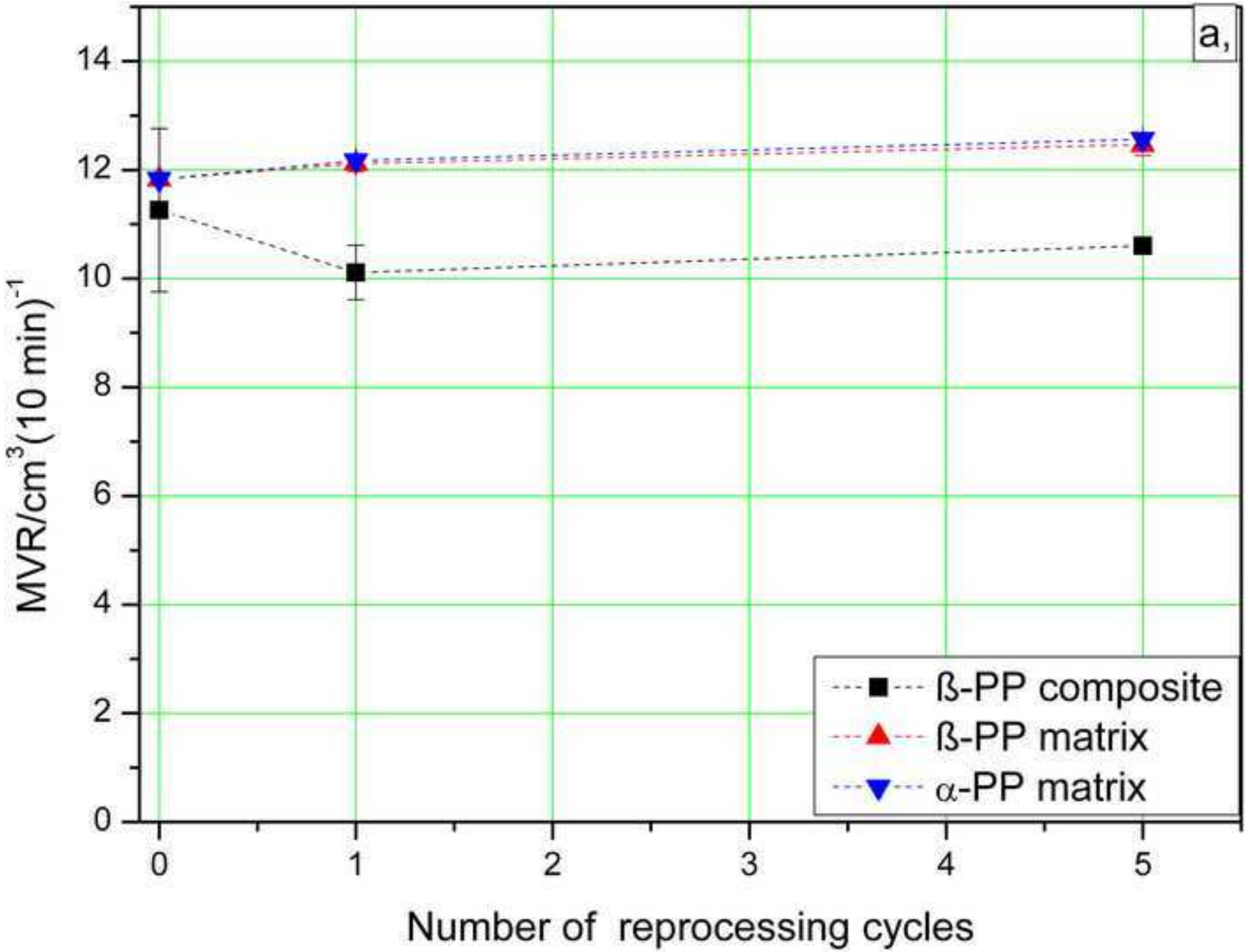

Figure 1b

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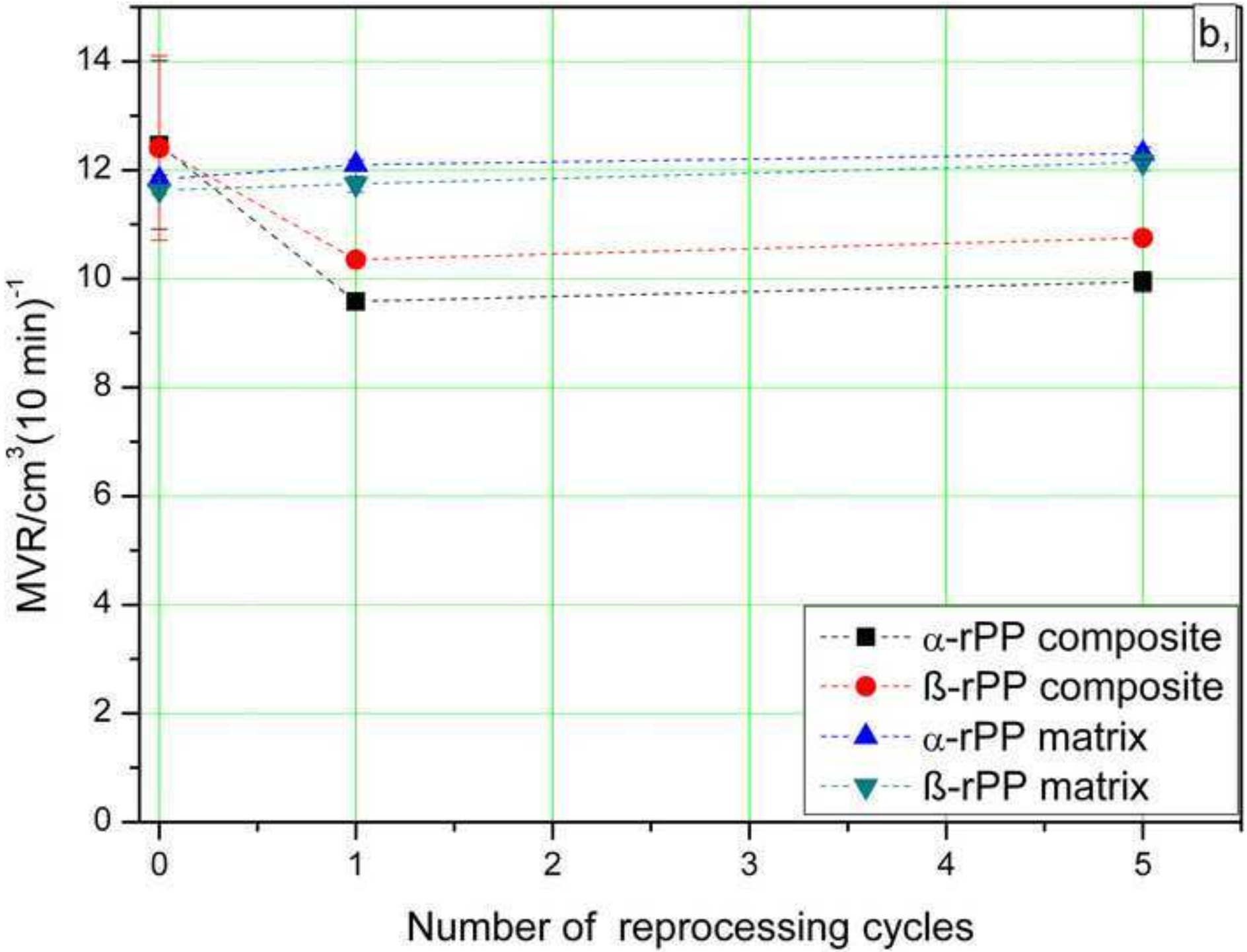

Figure 2a

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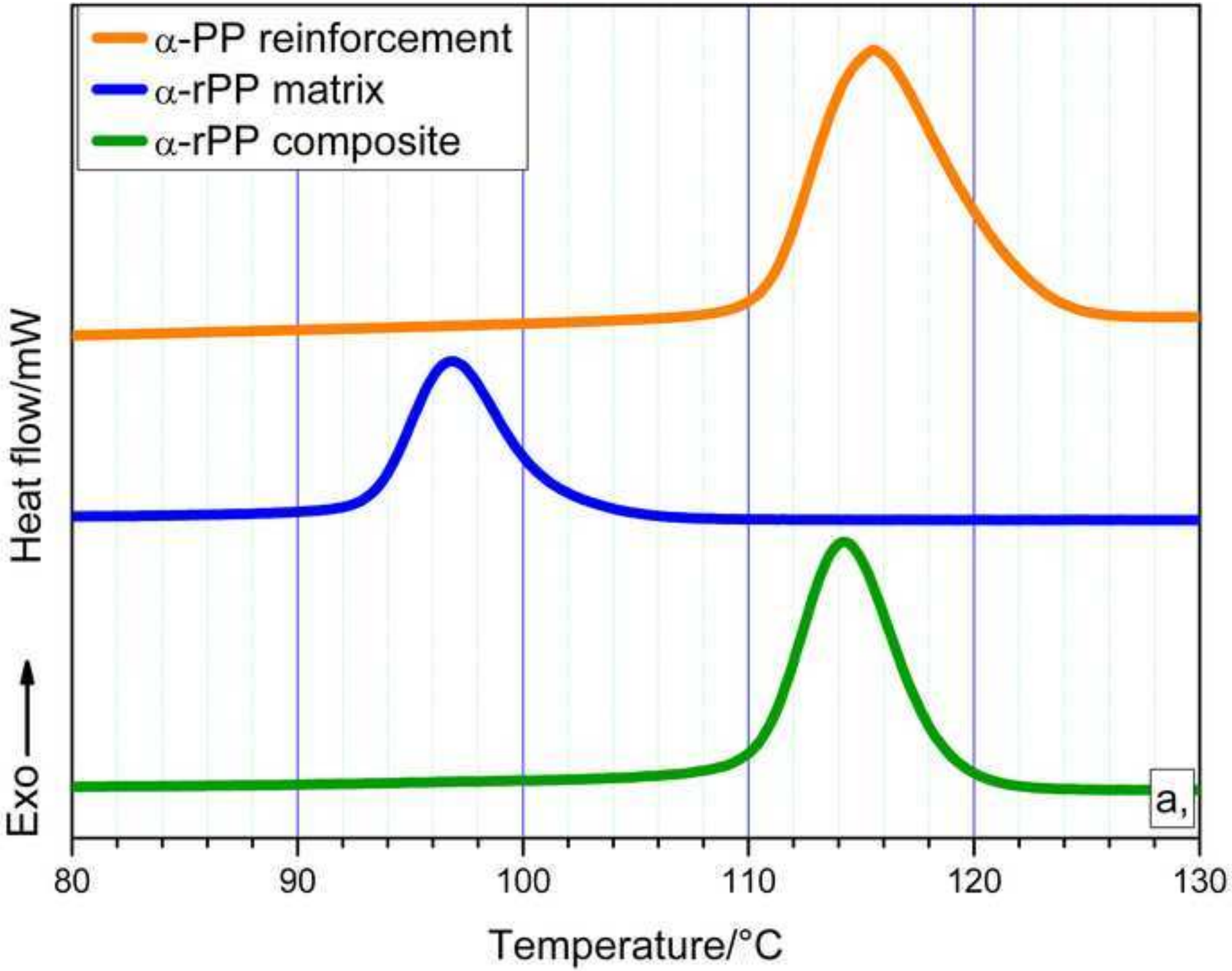

Figure 2b

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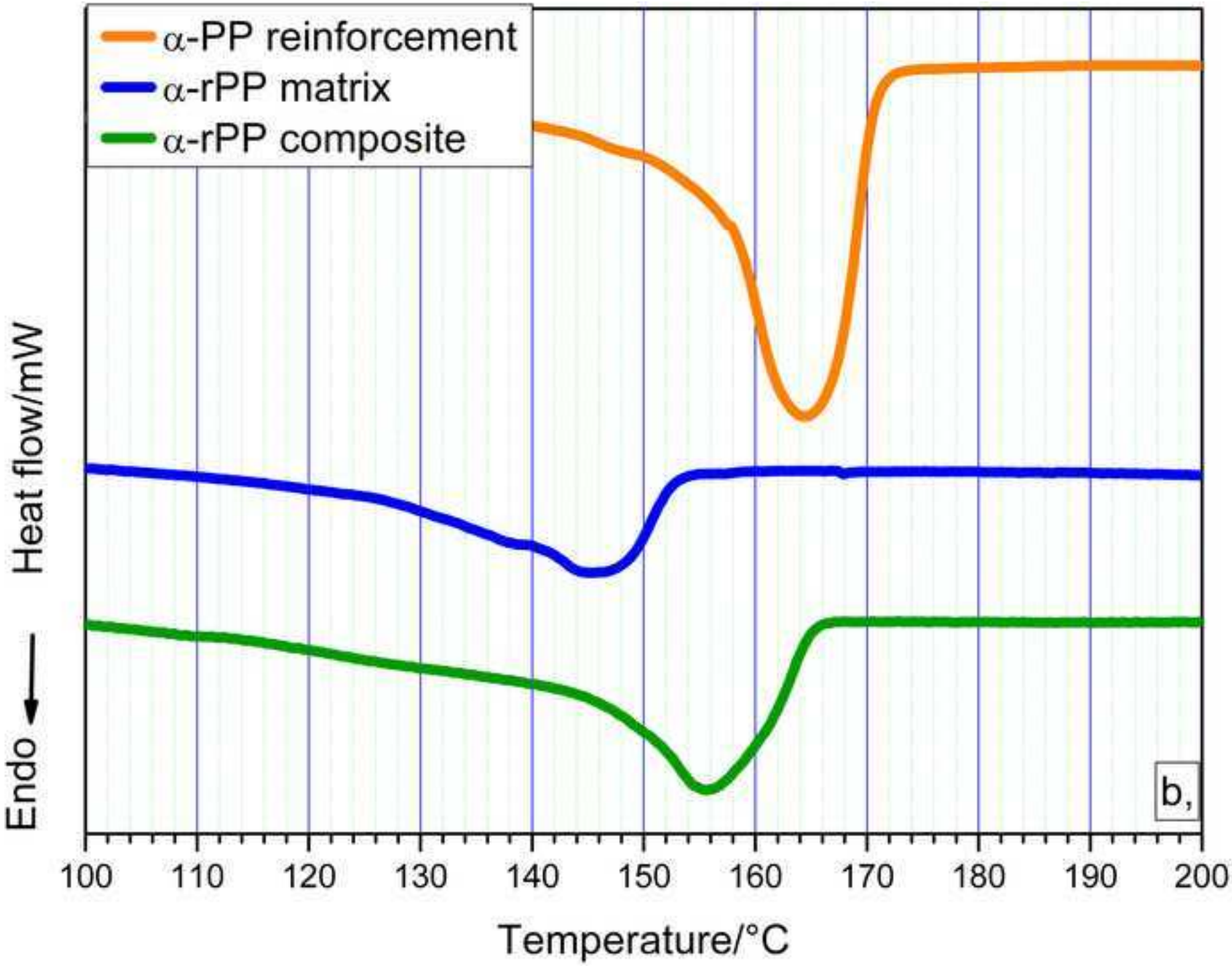

Figure 3a

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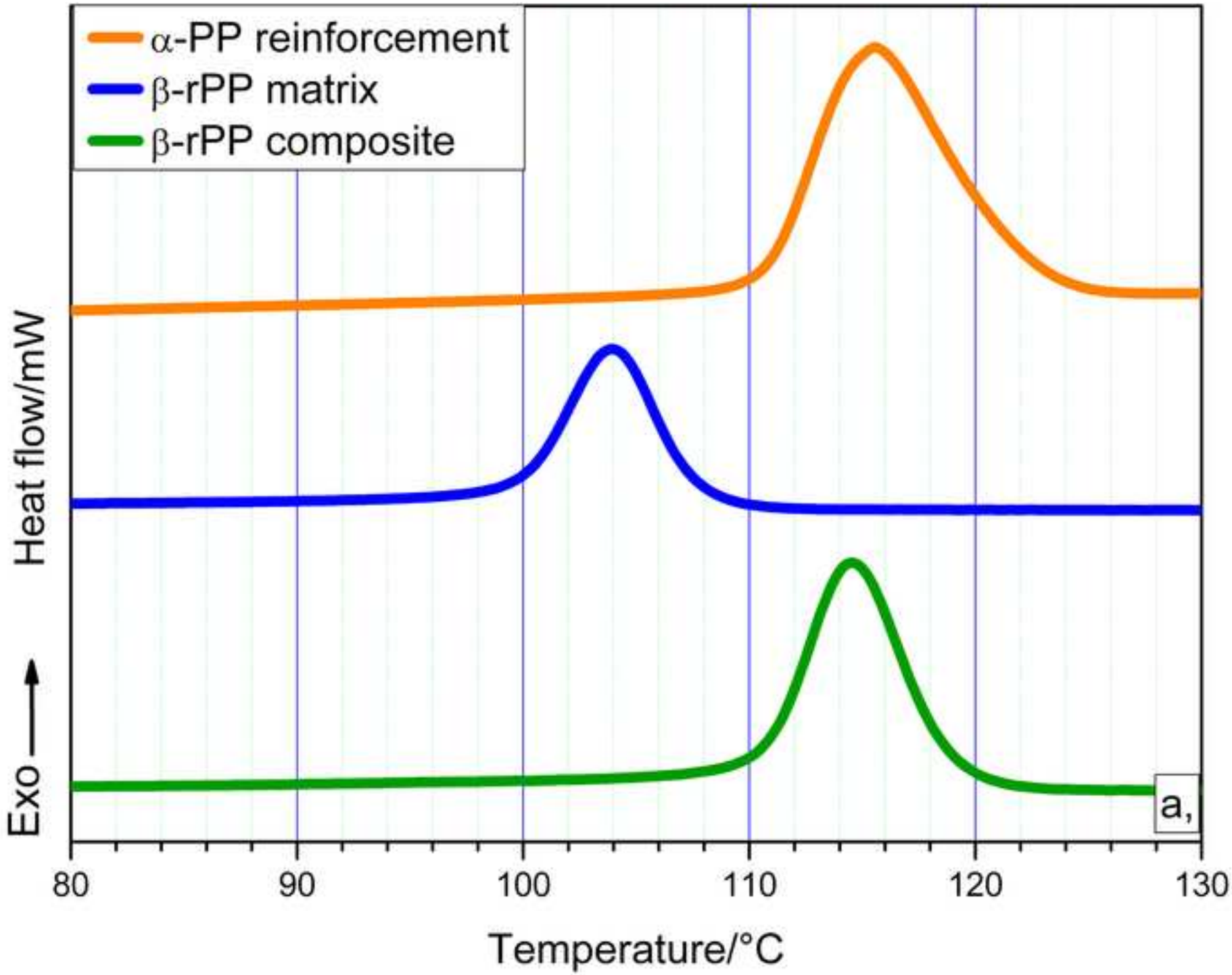

Figure 3b

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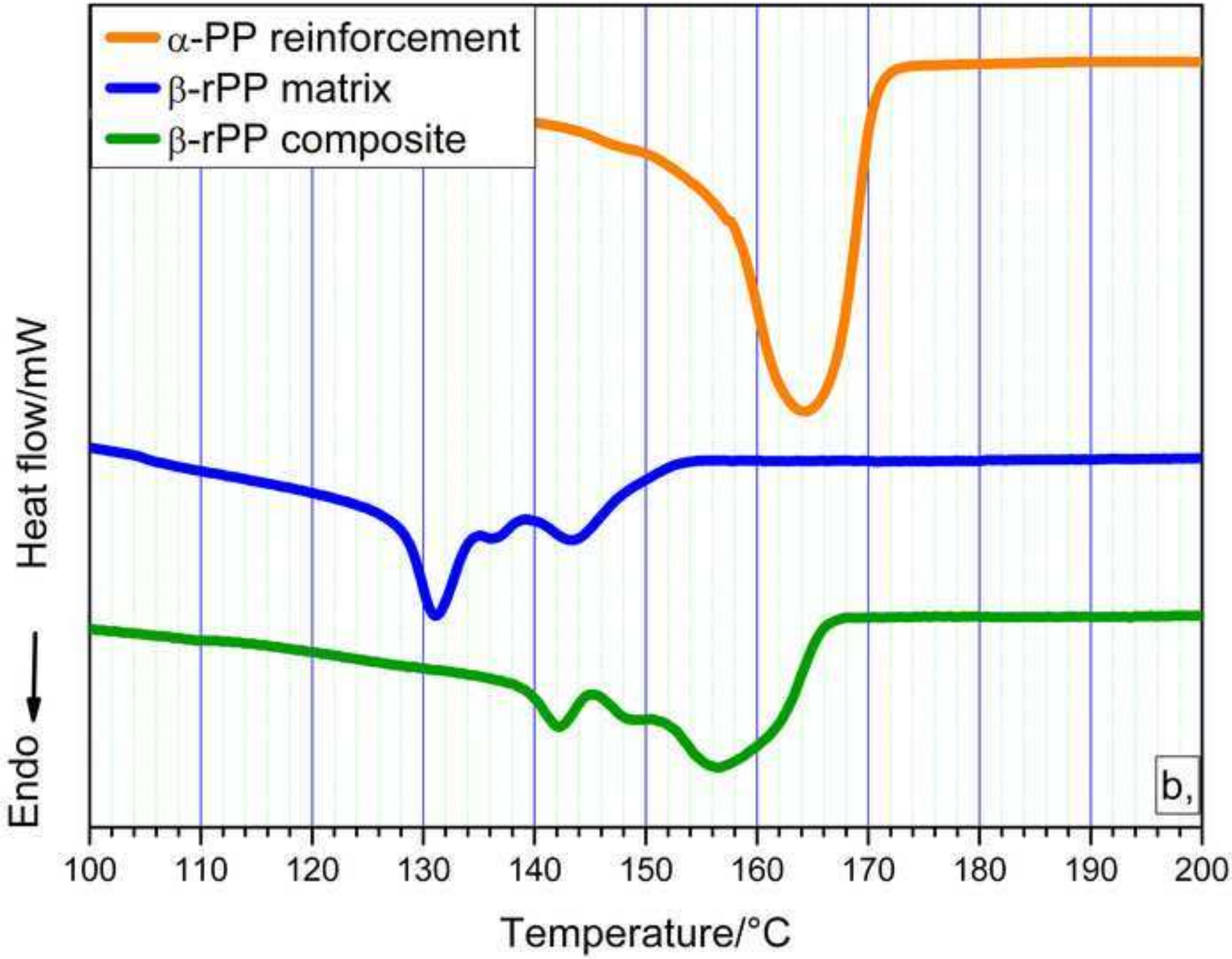

Figure 4a

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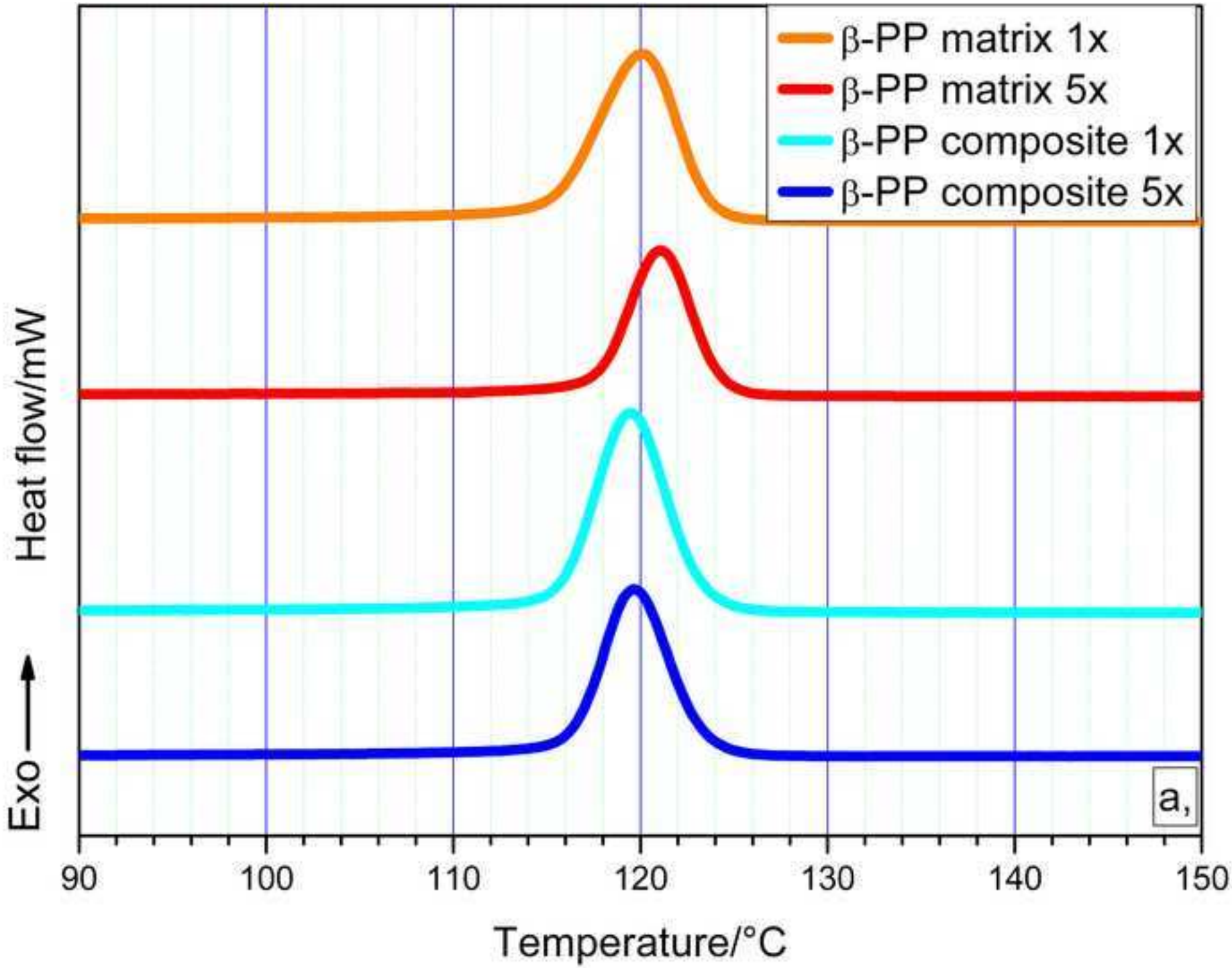

Figure 4b

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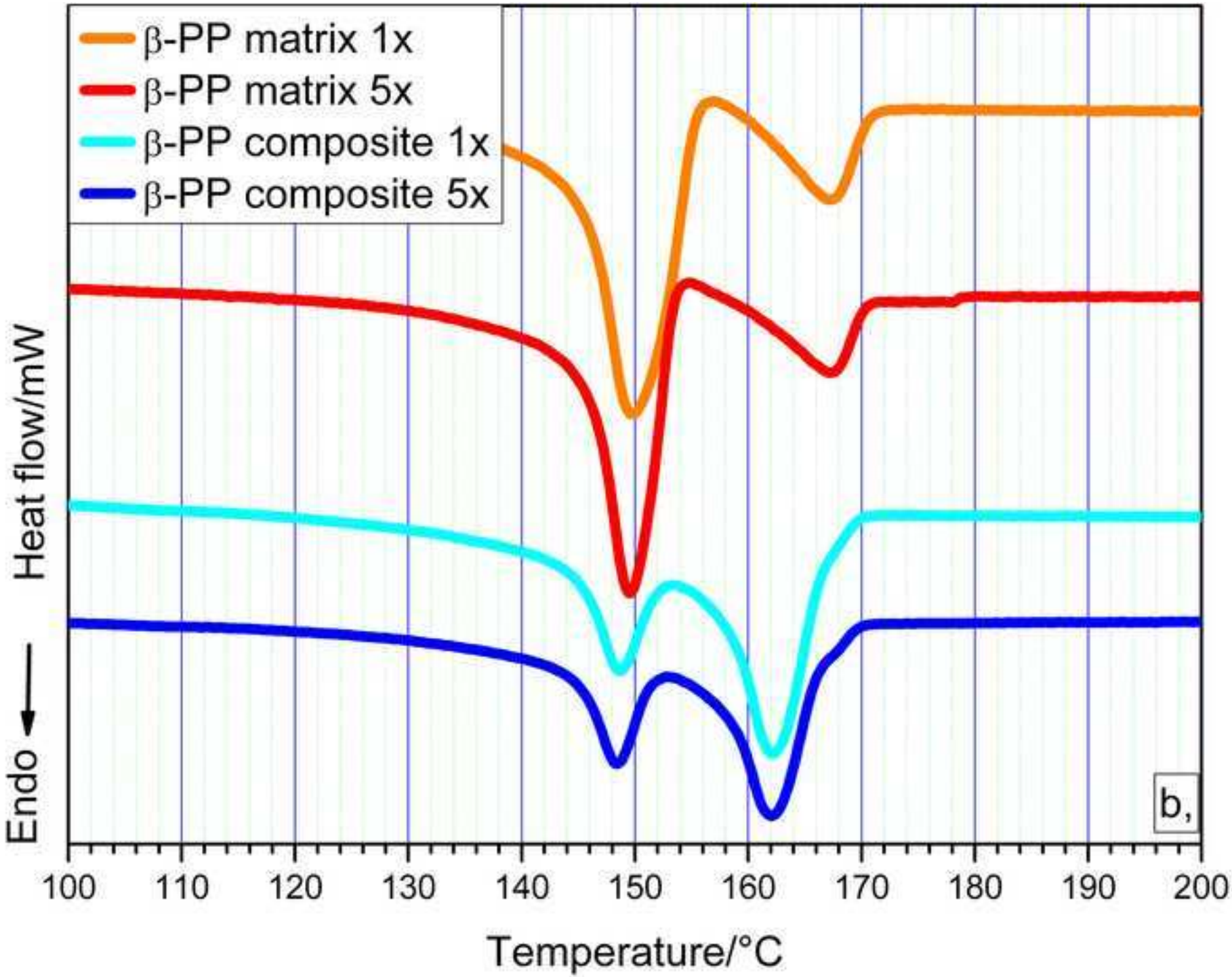

Figure 5

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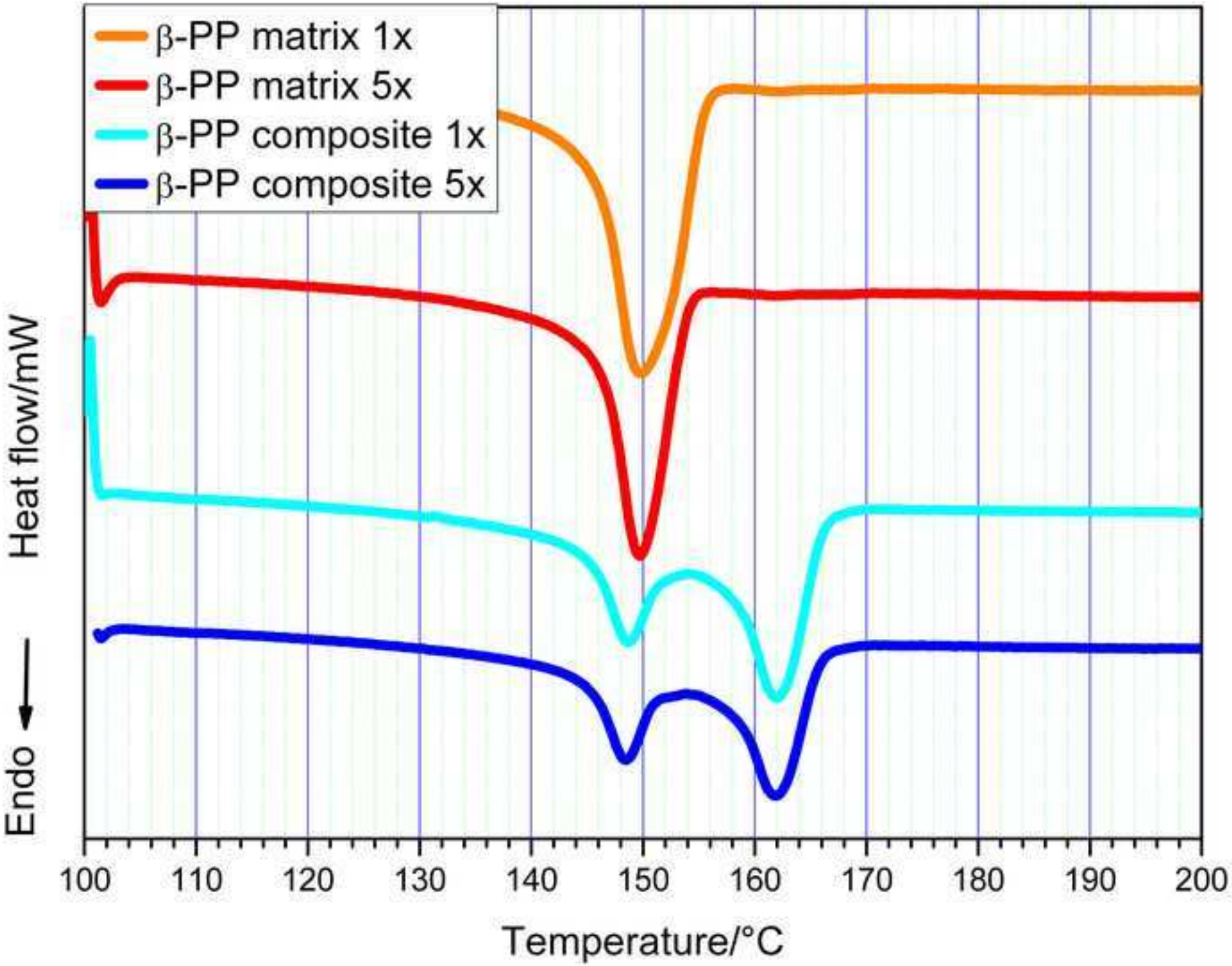

**Figure 6a**

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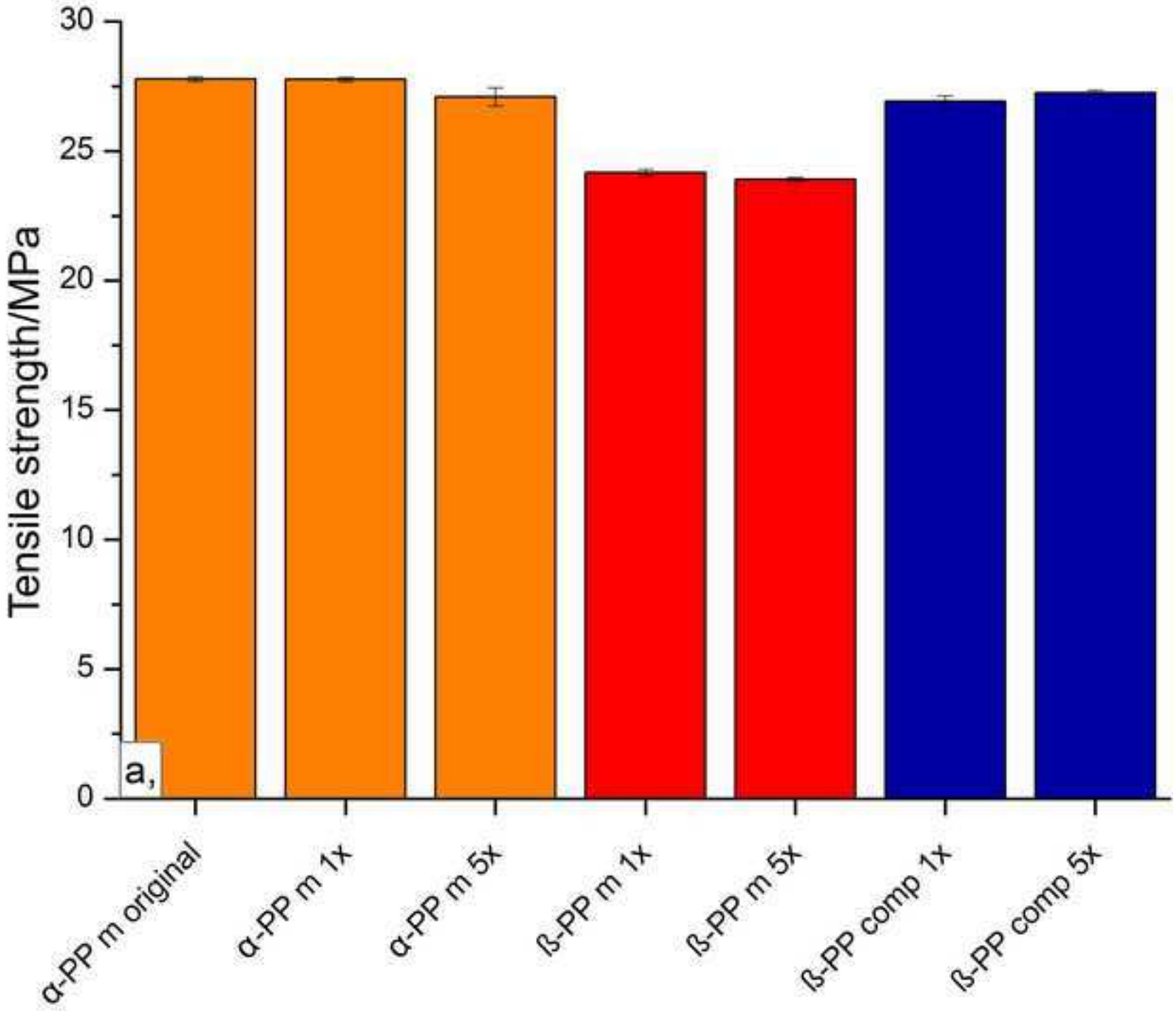

**Figure 6b**

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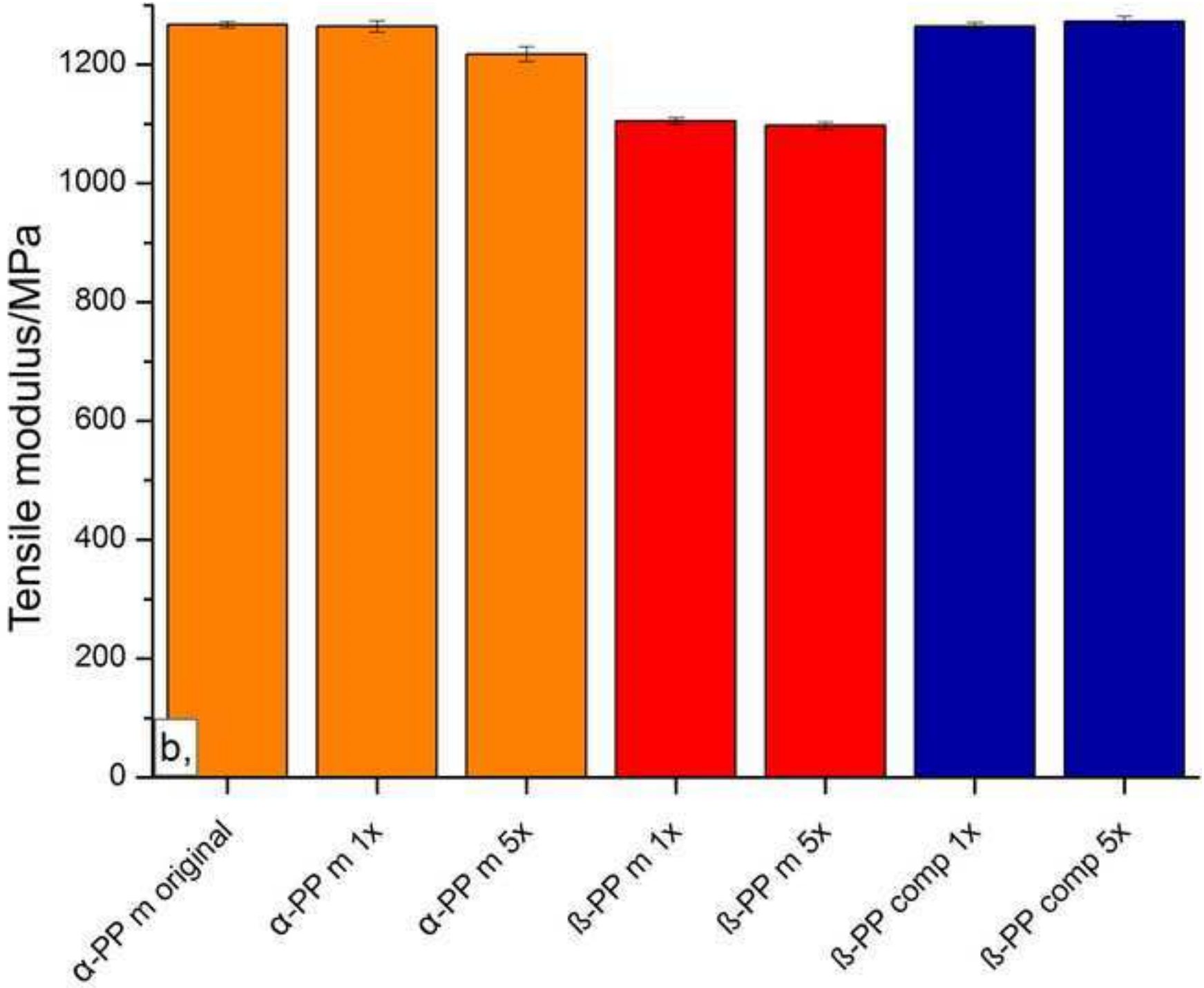

Figure 7a

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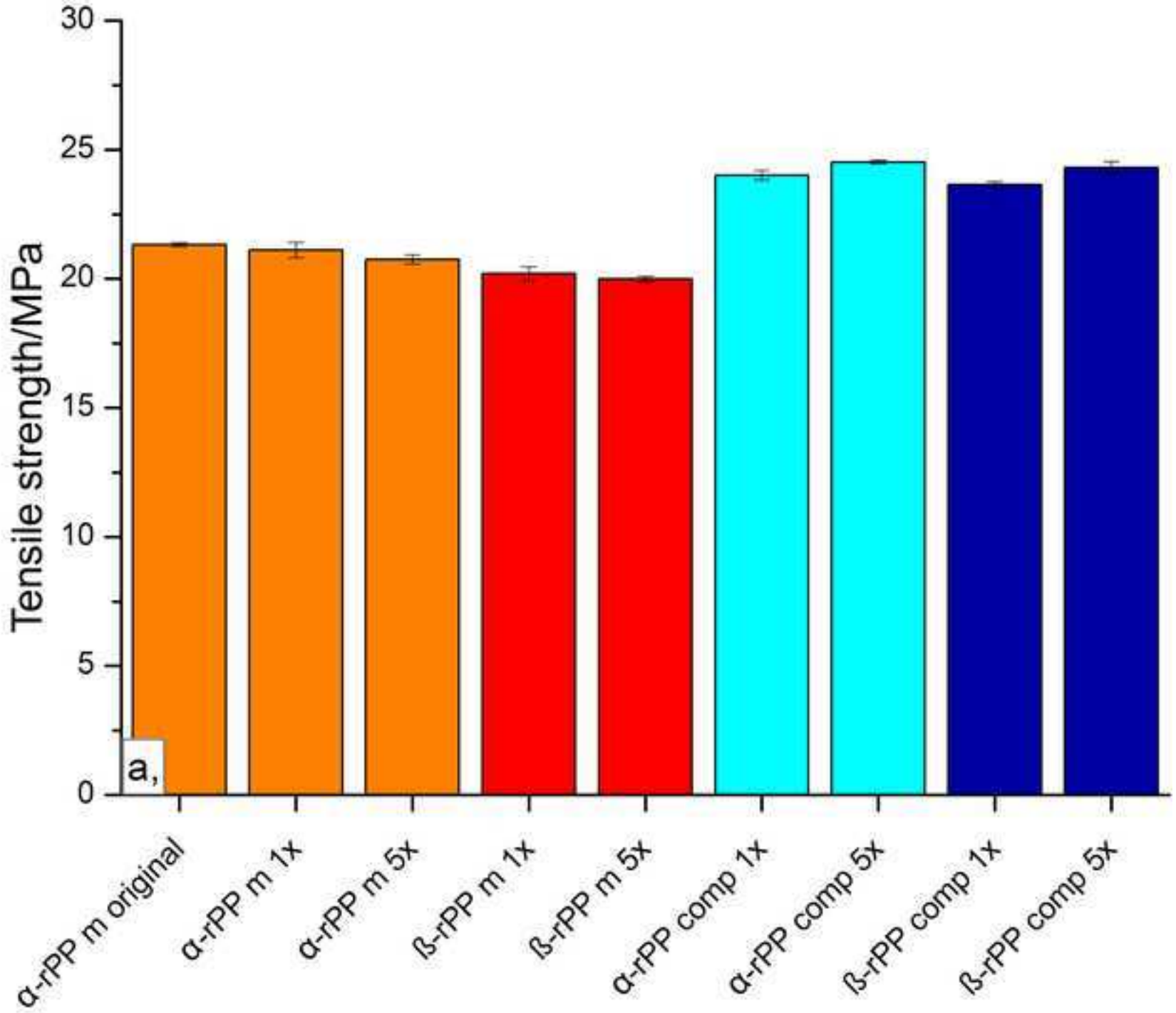

Figure 7b

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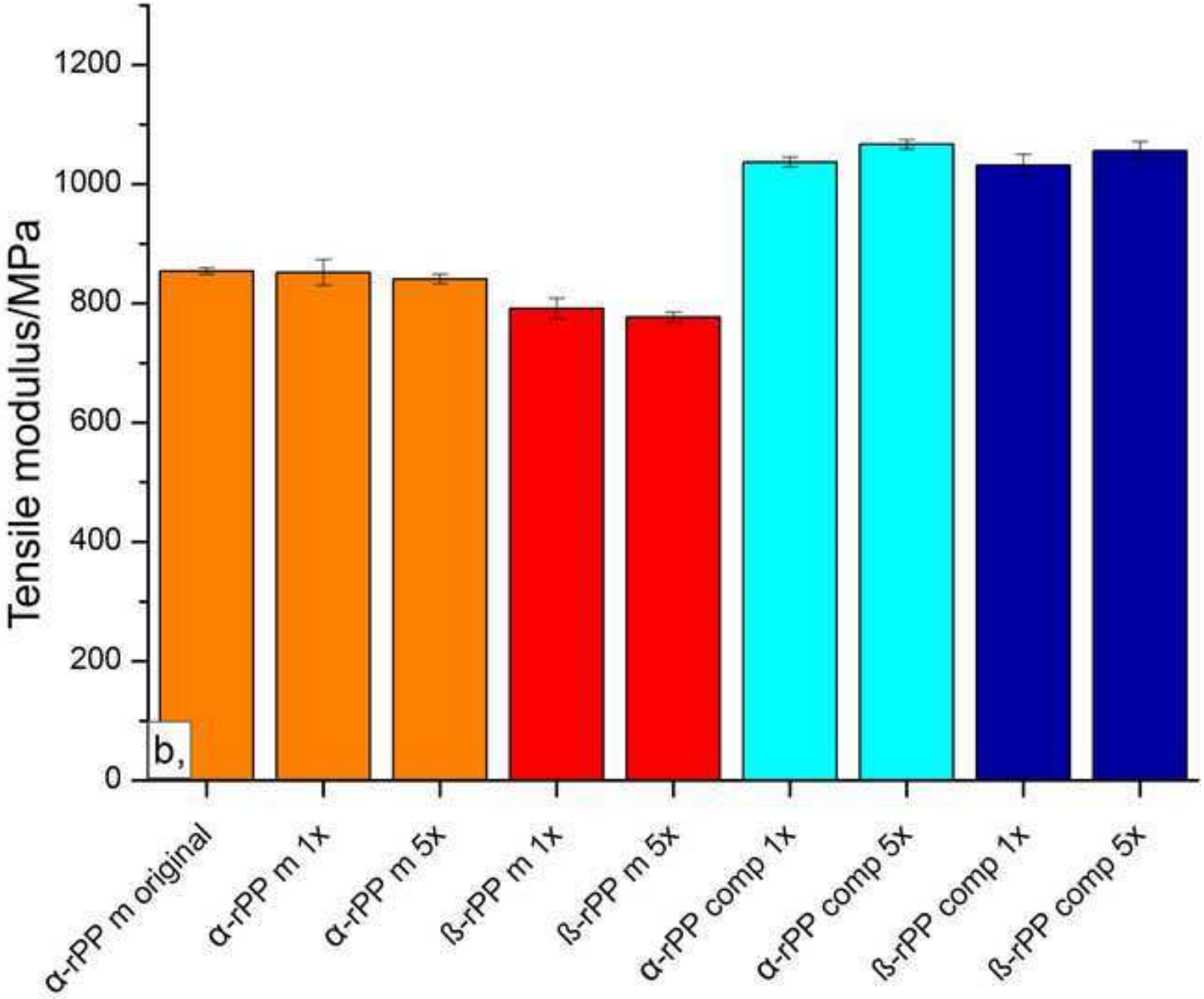

Figure 8a

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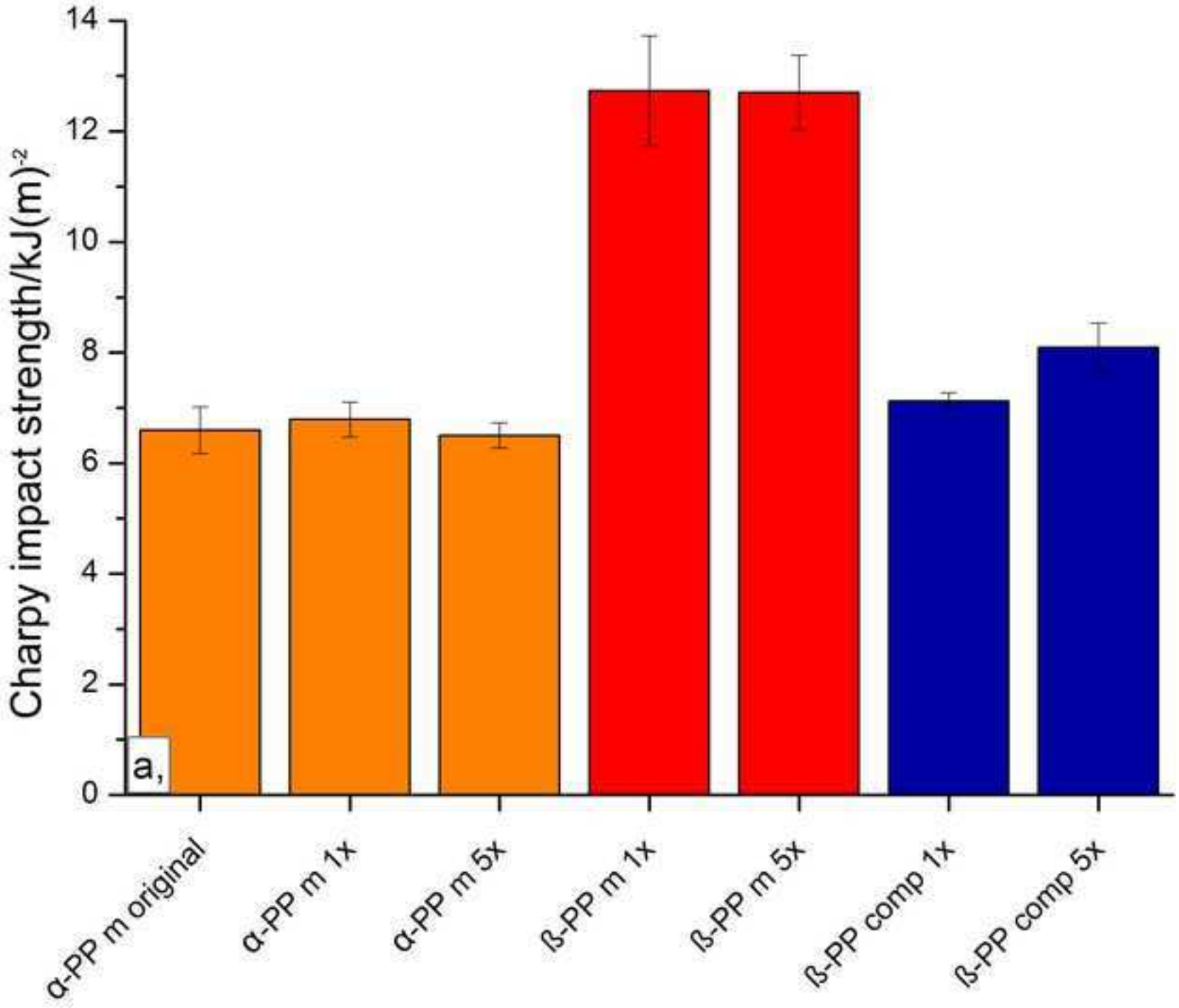

**Figure 8b**

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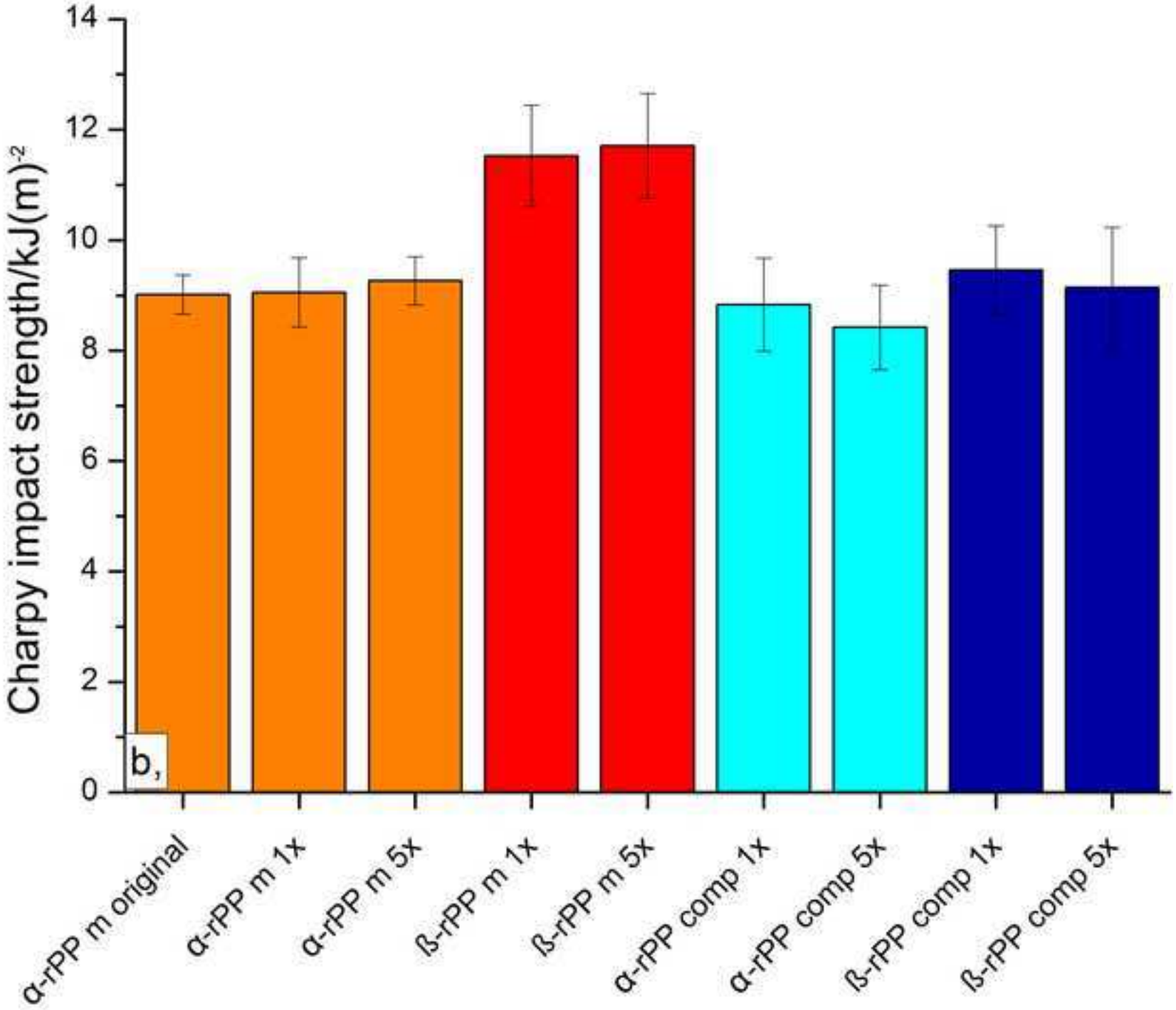


**Table 1**

Table 1

| Type | Melting temperature, $T_m$/°C |
|---|---|
| α-PP tape | 172.4 |
| α-PP | 164.4 |
| β-PP | 151.5 |
| α-rPP | 142.5 |
| β-rPP | 131.3 |

**Table 2**

Table 2

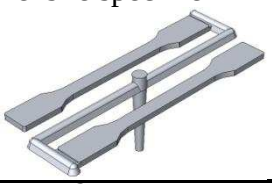
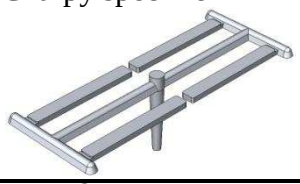
| Parameter | Tensile specimen  | Charpy specimen  |
|---|---|---|
| Injection volume | 44 $cm^3$ | 40 $cm^3$ |
| Injection rate | 50 $cm^3s^{-1}$ | 50 $cm^3s^{-1}$ |
| Switch over point | 12 $cm^3$ | 11 $cm^3$ |
| Holding pressure | 400 bar | 300 bar |
| Holding time | 20 sec | 20 sec |
| Screw speed | 15 m $min^{-1}$ | 15 m $min^{-1}$ |
| Cooling time | 15 sec | 15 sec |
| Decompression | 5 $cm^3$ | 5 $cm^3$ |
| Mould temperature | 50°C | 50°C |
| Real pressure | 540 bar | 440 bar |

**Table 3**

Table 3

| | α-PP matrix | β-PP matrix | α-rPP matrix | β-rPP matrix | β-PP composite | α-rPP composite | β-rPP composite |
|---|---|---|---|---|---|---|---|
| Neat material | x | | x | | | | |
| 1x extruded | x | x | x | x | x | x | x |
| 5x extruded | x | x | x | x | x | x | x |