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## Impact of ionic liquids on the processing and photo-actuation behavior of SBR composites containing graphene nanoplatelets

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## ABSTRACT

The effects of butylpyridinium bromide-based ionic liquids (ILs) on the vulcanization kinetics, mechanical properties and photo-actuation ability of styrene-butadiene rubber (SBR) composites filled with graphene nanoplatelets (GnPs) were investigated. Two different ILs, 1-butylpyridinium bromide (BBP) and 4-methyl-1-butylpyridinium bromide (BMBP), were introduced into rubber compounds. The methodology of IL incorporation into the rubber compounds was studied as well. The ILs were added either directly as a single component or after immobilization onto the GnPs surface. High values of the modulus and tensile strength, which are important mechanical properties, were observed for the composites filled with immobilized BMBP. Freely dispersed BBP resulted in composites with the fastest scorch time, highest thermal and electrical conductivity, and best photo-actuation response of 0.230 mm.

## 1. Introduction

Polymer nanocomposite-based photo-actuators are a class of smart materials that are able to change their shape reversibly upon external light stimulation [1–3]. The stretched polymer chains form coils upon light exposure but regain their original shape when the stimulus is turned off. Polymer photo-actuators have been utilized in applications such as robots, light-driven motors, valves [4] or pens for molecular printing [5–7]. Elastomers, when used in these applications, are usually formed by mixing an elastomeric polymer matrix and a filler. The fillers in these materials should not only provide good mechanical properties but also a high absorption of light and facilitate heat transport in the nanocomposites. Carbon black, carbon nanotubes and graphite allotropes have been investigated due to their ability to absorb the near infrared light and convert it to heat [8–12]. A plate-like character of the filler shape has been found to be most beneficial for the actuation

response, as well as the dispersion and distribution of the filler particles within the polymer matrix [13,14]. Both thermoplastic elastomers and crosslinked polymer matrices have been extensively studied [15–17]. Although thermoplastic elastomers enable repeated material processing, these materials are prone to a loss of shape when the temperature generated in the sample exceeds the melting point of the physical crosslinking points [14]. In contrast, chemically crosslinked structures provide a more stable shape and allow for material utilization at higher light intensities. Among the spectrum of crosslinked matrices, poly (dimethyl siloxane) (PDMS) is preferred as a polymer matrix due to its large tensile strain. However, the modulus and tensile strain of PDMS are lower than those of vulcanized rubbers, such as natural rubber or styrene-butadiene rubber (SBR). SBR still arouses unflagging interest among scientists, since 70% of its global production is used in manufacturing tires due to SBRs inherent properties of abrasion resistance, which improves tire performance. The elastomer also offers

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excellent water resistance, good physical properties and easy processing. Thus, it is also used for production of footwear soles and heels, seals, membranes, hose and rolls [18]. In addition, vulcanized elastomers have beneficial processing techniques. Compared to the crosslinking of liquid DMS monomers, rubbers are processed by melt mixing techniques, including two-roll milling. These techniques are considered more environmentally friendly than other techniques, and products can be prepared in large scale.

Photo-mechanical actuators based on vulcanized rubber have not been studied before. Therefore, in this study, sulfur-vulcanized composites of SBR elastomers were investigated. The SBR composites were filled with graphene nanoplatelets and enriched with ionic liquids. Ionic liquids are organic salts with melting points below 100 °C consisting of organic cations and organic or inorganic anions [19]. Ionic liquids have been recognized as promising additives in rubbers vulcanized by SBR. It has been shown that ILs can enhance the efficiency of vulcanization, playing the role of a coagent of the vulcanization process [20]. ILs even enable the replacement of the traditional vulcanization accelerator (2-mercaptobenzothiazole), which is known for its allergenic effects, without detrimental effects on the mechanical properties of vulcanizates [21]. Vulcanization accelerators are insoluble in rubbers, and thus, crosslinking occurs at the interphase of the two-phase system, and ILs have been observed to further accelerate this process [21]. ILs also contribute to the dispersion and distribution of carbon-based fillers due to their conjugated structure [22], and their high ionic conductivity [23], they have been used for the preparation of conductive composites [24] or ionomers [25]. These features of ILs suggest that their utilization in graphene/rubber vulcanizates can result in better particle dispersion and distribution, which are important parameters for a good photo-mechanical response [2], improved vulcanization kinetics and enhanced mechanical properties of composites.

In this study, SBR rubber was filled with graphene platelets, and two different ionic liquids, 1-butylpyridinium bromide (BBP) or 4-methyl-1-butylpyridinium bromide (BMBP) were used as additives. These ILs differ in the methyl pendant group, which is located in the *para* position toward the nitrogen cation. This methyl group can improve the interactions with the aliphatic parts of SBR rubber chains. However, this methyl group can prevent heat and charge transport and affect the vulcanization kinetics as well as the mechanical properties and photo-actuation response of these composites. Moreover, the ILs were introduced in two different ways, either as a single component or *via* immobilization onto the graphene nanoplatelets through noncovalent interactions. The vulcanization kinetic parameters; thermal, electrical and mechanical properties; and photo-actuation ability were studied for systems with various filler loadings, types of ILs, and methods of IL introduction.

## 2. Materials and methods

### 2.1. Materials

Styrene-butadiene rubber (SBR), trade name KER 1500, with a styrene content of 23 wt. % was supplied by Synthos S.A., Oswiecim, Poland. The Mooney viscosity of SBR was ML 1 + 4 (100 °C) 45–55. SBR was vulcanized with a conventional curing system containing sulfur (Siarkopol, Tarnobrzeg, Poland), 1,3-diphenylguanidine (DPG) and 2,2'-dibenzothiazyl disulfide (MBTS) (both provided by Akrochem Co., Akron, USA). A graphene nanoplatelet powder with an average particle diameter of less than 2 microns and an average surface area of 300 m<sup>2</sup> g<sup>-1</sup>, 5 at. % of oxygen, in high purity form was purchased from XG Sciences Inc., Lansing, USA. Pyridinium-based ILs, namely, 1-butylpyridinium bromide (BBP) and 4-methyl-1-butylpyridinium bromide (BMBP), were supplied by IoLiTec Ionic Liquids Technologies GmbH, Heilbronn, Germany. Pure acetone (99.5 %) was obtained from POCH S. A., Gliwice, Poland.

### 2.2. Preparation of IL-modified graphene nanoplatelets (GnPs/IL)

The ionic liquids were immobilized on the GnPs surface prior to blending with SBR. The details of the procedure can be found in the SI.

### 2.3. Preparation of rubber mixes and vulcanizates

The SBR compounds were first melt blended on a two-roll mill and then vulcanized. The description of the processes can be found in SI. The list of SBR compounds and their composition can be found in Table S1.

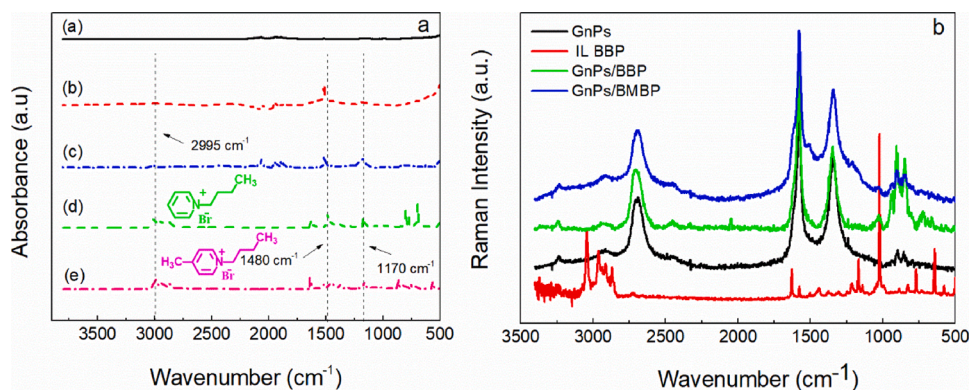
### 2.4. Methods

The presence of ILs on the GnPs surface was proved by Fourier transform infrared spectroscopy (FTIR), while the amount of organic content was determined by thermogravimetric analysis (TGA). The methods used for characterization of modified GnPs as well as SBR compounds are listed in SI. The curing characterization of SBR compounds was studied by rheology measurement, and the mechanical properties were studied by tensile tests. The particle dispersion and distribution were investigated by scanning electron microscopy (SEM). The effect of ILs on thermal and electrical conductivity was determined. The photo-actuation ability was evaluated by using thermomechanical analyzer [26]. The detailed description of the methodology used is listed in the SI.

## 3. Results and discussion

Successful modification of the GnPs in solution with both ILs – 1-butylpyridinium bromide (BBP) and 4-methyl-1-butylpyridinium bromide (BMBP) – was confirmed using FTIR spectroscopy. As shown in Fig. 1a, neat GnPs did not show any significant absorption bands because of the lack of terminal oxygen-containing groups and the absence of modifying agents. On the other hand, the GnPs modified with BBP exhibited a small peak at 2995 cm<sup>-1</sup> corresponding to NH— stretching from an amine salt, a sharp peak at 1480 cm<sup>-1</sup> can be attributed to the pyridine ring and the peak at approximately 1197 cm<sup>-1</sup> attributed to CN— bond stretching. A very similar situation was observed in the spectra of GnPs modified with BMBP due to their similarity in chemical structure. In this case, the absorption peaks in the GnPs/IL particle spectrum (Fig. 1b,c) were slightly shifted toward higher wavenumbers than those of neat ILs (Fig. 1d,e).

This phenomenon suggests intermolecular interactions between the molecules. They could be generated between the cations in the pyridine nitrogen center and the electron cloud around the GnPs (cation -  $\pi$  interactions). The interactions could also be generated by the oxygen-containing groups present on the graphene surface [27]. Hydrogen bonding interactions occur between the bromide anion and the hydroxyl groups on the GnPs surface, similar to what has been described in the literature [22]. The functional groups containing hydrogen (including hydroxyl groups) are naturally present on the graphene surface and are located at the edges of particles. Their amount depends on the particle size. The GnPs used in this study contain only 5 at. % of oxygen within all functional groups. Therefore, these interactions are considered to be minor and hardly detectable by FTIR or Raman spectroscopy (Fig. 1a and b). On the other hand, the Raman spectroscopy revealed that the intermolecular interactions are predominantly provided through cation- $\pi$  interactions. The strong band at around 1050 cm<sup>-1</sup> related to in-plane = C—H vibrations of mono-substituted or di-substituted benzene ring is obvious. This spectral feature is missing in the spectrum of neat graphene. Moreover, in the case of neat graphene, bands with small intensities can be observed in the region from 1000 cm<sup>-1</sup> to 800 cm<sup>-1</sup>. These bands belong to impurities at the ends of the graphene sheets and are related to out-of-plane = C—H vibrations (the amount of impurities is ~5 % in accordance with the supplier datasheet), and in this region, mentioned bands are highly stressed by the addition of IL to graphene



**Fig. 1.** a) FTIR spectra of pristine GnPs (a), modified with BBP (b), BMBP (c) and neat compounds of BBP (d) and BMBP (e). b) Raman spectra of BBP ionic liquid, neat GnPs and GnPs modified with BBP and BMBP (ILs immobilized on the GnPs surface).

sheets. This can be explained by the sensitivity of out-of-plane = C–H vibrations to the degree of face-to-face  $\pi$ - $\pi$  stacking whereas the ILs create such states and thus cause higher delocalization and higher polarizability. Similar behavior was already observed and described for a perylene system [28]. The interactions of ionic liquids and graphene nanoplattlets are shown in Scheme 1.

The content of immobilized ILs was estimated by using thermogravimetric analysis. Fig. 2 shows the weight loss of GnPs/BBP and GnPs/BMBP, which corresponded to 15 wt. % and 25 wt. %, respectively. With respect to their molar masses, the molar concentrations of immobilized BBP and BMBP had very similar values of 7 % and 9 %, respectively.

GnPs modification in solution allows for an effective immobilization of ILs on the particle surface. However, from a technological point of view, the simplest method relies on introducing the GnPs and the IL directly into the bulk composition during preparation of the rubber mixture by melt mixing. In this case, the ILs are freely dispersed within the rubber matrix. This approach was applied as an alternative strategy in this study. It was expected that the ILs and GnPs introduced into rubber compounds as free compounds could more easily interact with all the components of rubber compared to ILs already immobilized on the GnPs surface. Similarly, as with the GnPs, the ILs can interact with benzene rings of SBR and/or the accelerators DPG and MBTS. Their chemical structures are shown in Scheme 2.

The effect of the ILs on the curing kinetics of rubber mixtures was investigated by rheometrical measurements that allow determination of the parameters of the vulcanization process. Also, the shape of the obtained curve shows that neither chain scission nor crosslink breakage was dominant during heating because reversion did not occur, Fig. 3. The addition of neat GnPs did not affect the curing kinetics significantly compared to the neat SBR in the studied concentration range. The torque was relatively unaffected, or slightly decreased, in the case of BMBP at a

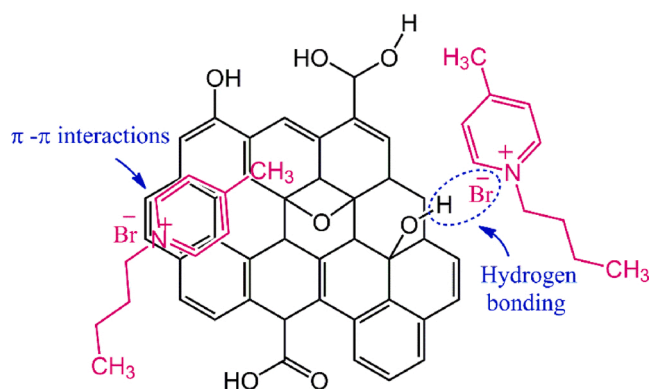
low filler content. To the contrary with 10 phr of filler content, a slight increase of torque was observed in this case. Except for the sample with 1 phr of filler's concentration, the BBP increased the torque regardless of the way of incorporation into the vulcanizate. It can be concluded the BBP affects the vulcanization process more significantly.

To quantify the effects, the vulcanization parameters such as the minimum torque ( $ML$ ), torque increment ( $\Delta M$ ), scorch and optimal curing time ( $TC2$  and  $TC90$ , respectively) were determined. The comparison of the vulcanization parameters is shown in Fig. 4(a–d).  $ML$  correlates with the viscosity of rubber mixtures, parameter  $\Delta M$  reflects elastomers' crosslinking density, while the scorch time  $TC2$  and the optimal vulcanization time  $TC90$  mainly correlate with the processing of the rubber [29].

The presence of fillers enhanced the  $ML$  of the SBR compounds. The most significant increase was observed when freely dispersed BMBP was used (Fig. 4a). Thus, the presence of a methyl pendant group in the BMBP structure may result in interactions with the aliphatic parts of SBR chains, thus hindering SBR chain movement. The highest torque increment ( $\Delta M$ ) was observed for compounds containing freely dispersed BBP (BBP1) at lower filler content (1 phr to 5 phr), as well as immobilized on GnPs surface (BBP2) at higher filler content (7 phr and 10 phr). BMBP2 did not result in a similar improvement, probably because restrictions in chain mobility influenced the crosslinking process. The scorch time is an induction period when the activator complex is formed. This complex reacts with molecular sulfur, which results in the formation of various polysulfide chains. The effects of the ILs on  $TC2$  are apparent at 3 and 5 phr, and is even more pronounced at higher concentrations. The ILs freely dispersed within rubber compounds significantly decreased their scorch time, –BBP was more active. These results confirm that the investigated ILs interact with the accelerators and sulfur during the curing process.

The addition of neat GnPs led to an enhancement of scorch time ( $TC2$ , Fig. 4c). This phenomenon may be due to the presence of active centers on the GnPs surface, which favor the absorption of curing system compounds. Therefore, GnPs at higher filler loading can be considered as a deactivator in the process of sulfur vulcanization [30]. The ILs reduce the direct adsorption of the curing system onto the GnPs surface, which makes it less accessible and thus reduces its efficiency as a deactivator. The shorter  $TC2$  for the system with ILs can also be related to a higher thermal conductivity promoting the vulcanization process and/or accelerating the effect of the IL itself on the rate of sulfur vulcanization of the rubber matrix. Here, BBP was more effective than BMBP, where the methyl group partially prevented the crosslinking process.

The optimal curing time  $TC90$  is a period necessary to reach 90 % of maximal torque, and serves as time used for vulcanization of rubber mixtures. The shorter the  $TC90$ , the more efficient the crosslinking system. At lower filler content the differences in  $TC90$  were not as



**Scheme 1.** Interactions of ionic liquid BMBP with graphene nanoplattlet.

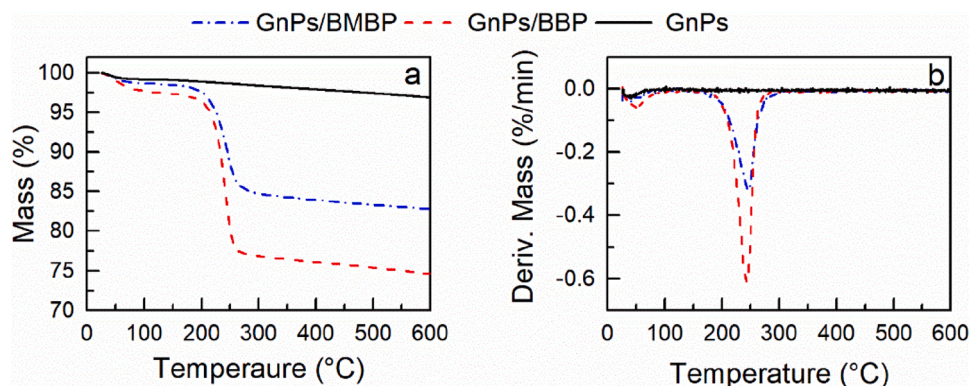
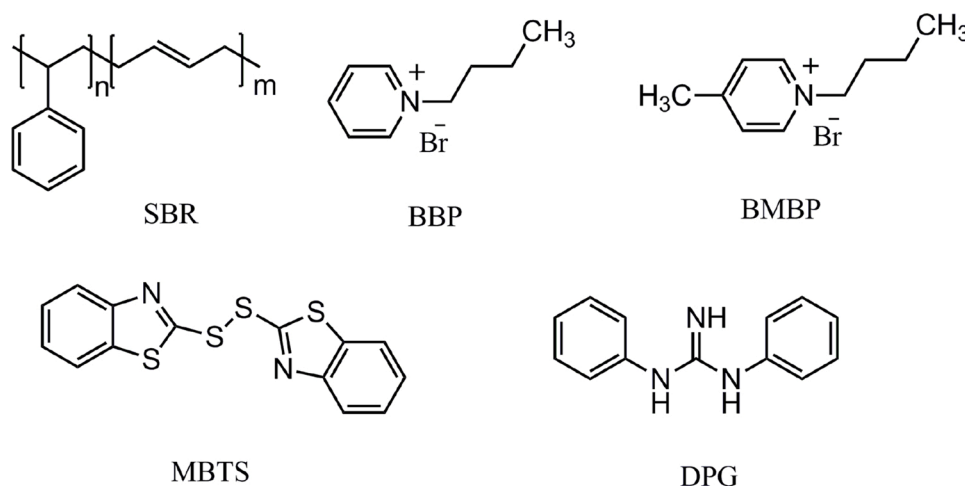


Fig. 2. Thermogravimetric analysis of neat GnPs and GnPs modified with ionic liquids from solution (a) and the derivation of the TGA curves (b).



Scheme 2. Chemical structures of the SBR matrix, ionic liquids butyl pyridinium bromide (BBP), 4-methyl-1-butylpyridinium bromide (BMBP), accelerators 2,2'-dibenzothiazyl disulfide (MBTS) and 1,3-diphenylguanidine (DPG).

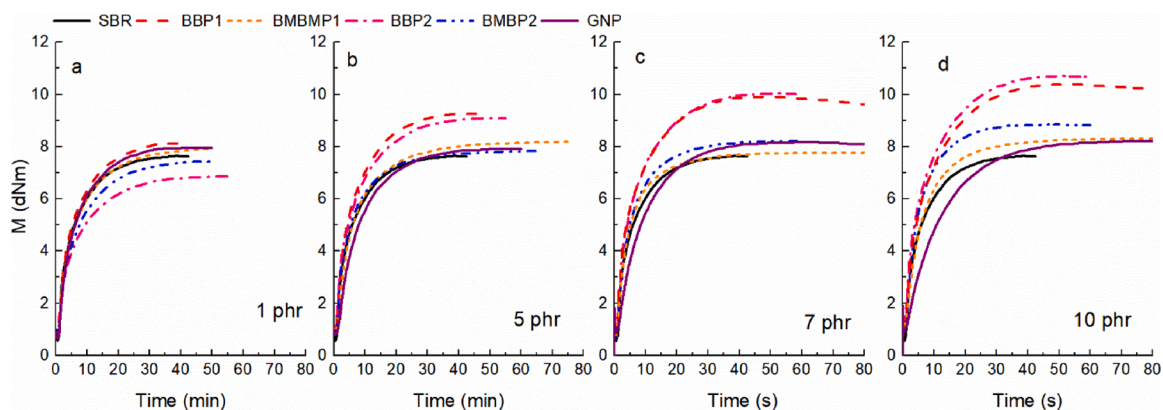


Fig. 3. Vulcanization curves of the investigated SBR mixtures with different filler contents at 160 °C; (a) 1 phr of GnPs; (b) 5 phr of GnPs; (c) 7 phr of GnPs; (d) 10 phr of GnPs (1 refers to freely dispersed ILs, 2 refers to ILs immobilized on the GnPs surface).

apparent. The  $TC_{90}$  of all the filled rubber compounds ranged from 18 to 30 minutes (Fig. 4d). The freely dispersed BBP led to the fastest formation of chemical crosslinks and the highest density of the formed network. At higher filler loading (7 and 10 phr) a significant increase of  $TC_{90}$  was observed for neat GnPs. That means the active sites on neat GnPs surface captured crosslinking compounds leading to the decrease of their concentration that was taking part in the crosslinking process.

The static mechanical properties of the vulcanized rubber composites were characterized by tensile tests. The values of the modulus at 100 %

elongation ( $M_{100}$ ), tensile strength ( $TS$ ) and elongation at break ( $EB$ ) are shown in Fig. 5.

It is known that ILs enable stress transfer and thus affect the mechanical properties of rubber vulcanized compounds [31]. At lower filler content, below 5 phr, the highest modulus at an elongation of 100 % and tensile strength was obtained for a system with BMBP2, where the IL was immobilized on the GnPs surface. The values of the 100 % modulus were 0.8 MPa and 1.2 MPa, and the tensile strength values were 4.5 MPa and 6 MPa for composites filled with neat GnPs and BMBP immobilized on

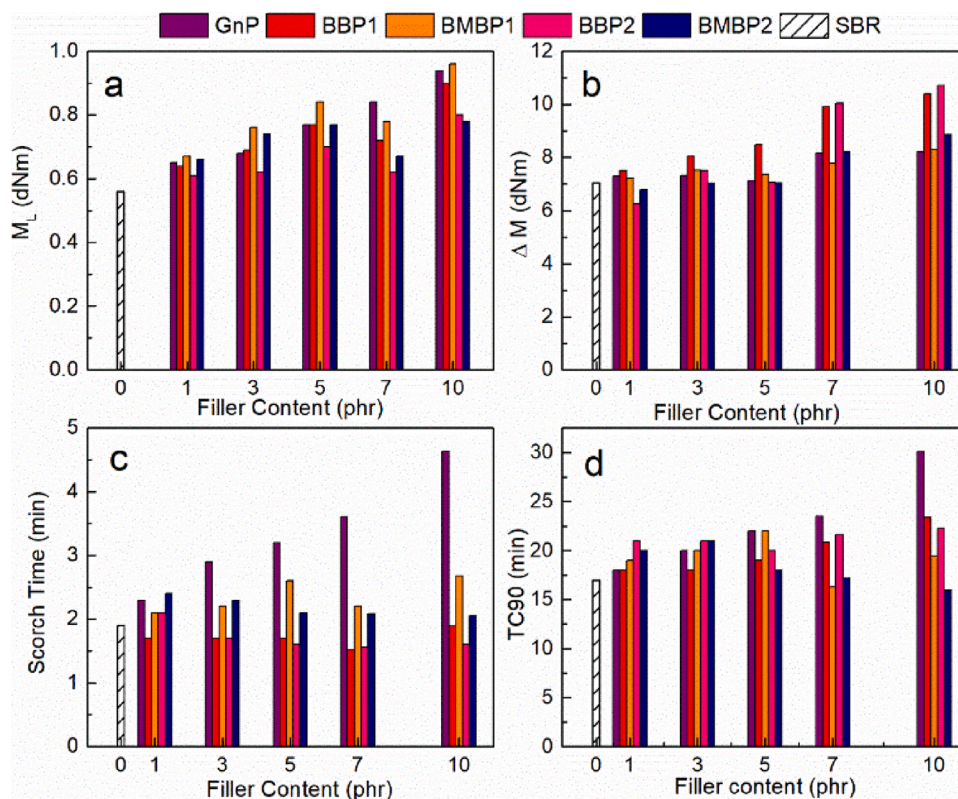


Fig. 4. Curing characteristics of SBR compounds, both, unfilled and filled with untreated and modified GnPs: (a) minimum torque ( $M_L$ ), (b) torque increment during vulcanization ( $\Delta M$ ), (c) scorch time ( $TC_2$ ), and (d) optimal curing time ( $TC_{90}$ ).

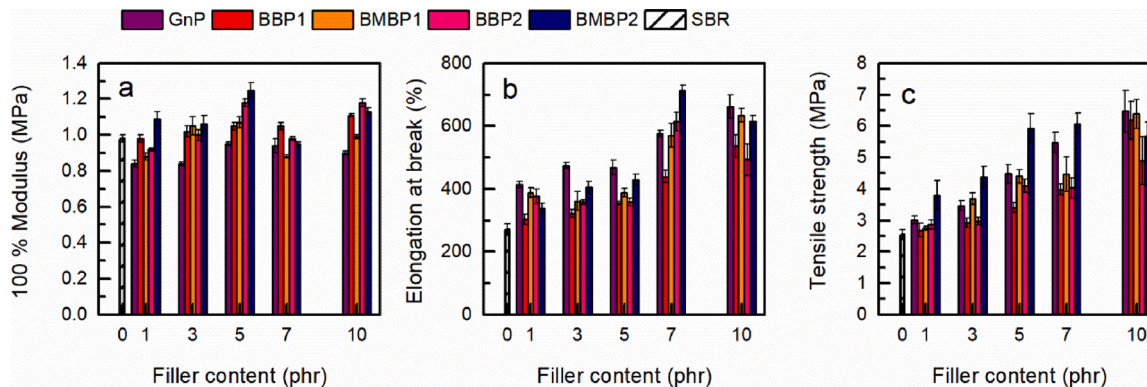


Fig. 5. Mechanical properties of SBR vulcanizates versus the filler content: (a) stress at 100 % elongation of SBR vulcanizates ( $M_{100}$ ); (b) tensile strength of SBR vulcanizates ( $TS$ ); (c) elongation at break of SBR vulcanizates ( $EB$ ).

the GnPs surface, respectively. This effect is related to the effective stress transfer in the interfacial region, but can also be related to a better dispersion of the GnPs in presence of ILs. A similar trend was reported in SBR system fillers with IL-modified carbon nanotubes [31]. The increase in the elongation at break was caused by the plasticizing effect of the neat GnPs that facilitated polymer chain movement. Graphitic fillers were found to disturb the physical crosslinking sites of styrenic-based elastomers [9,14], facilitating chain movement. A similar effect could occur in this system of GnPs-filled SBR composites. The elongation at break increased in all composites at high filler composites (7 and 10 phr). When composites filled with freely dispersed IL were compared, BBP1 led to lower elongations and lower tensile strengths compared to BMBP1. The same trend was observed for immobilized ILs, in systems filled with BBP2 and BMBP2.

The dispersion and distribution of the filler was evaluated by SEM.

The SBR composites loaded with the lowest (1 phr, left column) or the highest (5 phr, right column) amount of filler were analyzed. The SBR filled with 7 phr of GnPs, BBP1 and BMBP1, is shown in Fig.S2. Generally, the micrographs reveal a homogenous distribution of the particles in the rubber matrix which is typical for the two-roll mill processing technique [32]. The neat GnPs formed agglomerates (Fig. 6a, b, Fig. S2a, d). The freely dispersed ILs did not improve the particle dispersion (Fig. 6b,c and Fig. 6g,h, Fig. S2). While the addition of BBP immobilized on the GnPs surface led to a slight improvement in the filler dispersion (Fig. 6e,f), a significant effect was observed in the composite of the GnPs with immobilized BMBP (Fig. 6i,j). These observations correlate well with the tensile test results.

The effect of the incorporation of pristine GnPs and GnPs with BMBP and BBP on the thermal and electrical conductivity of the SBR vulcanizates was investigated (Fig. 7). The electrical conductivity increased

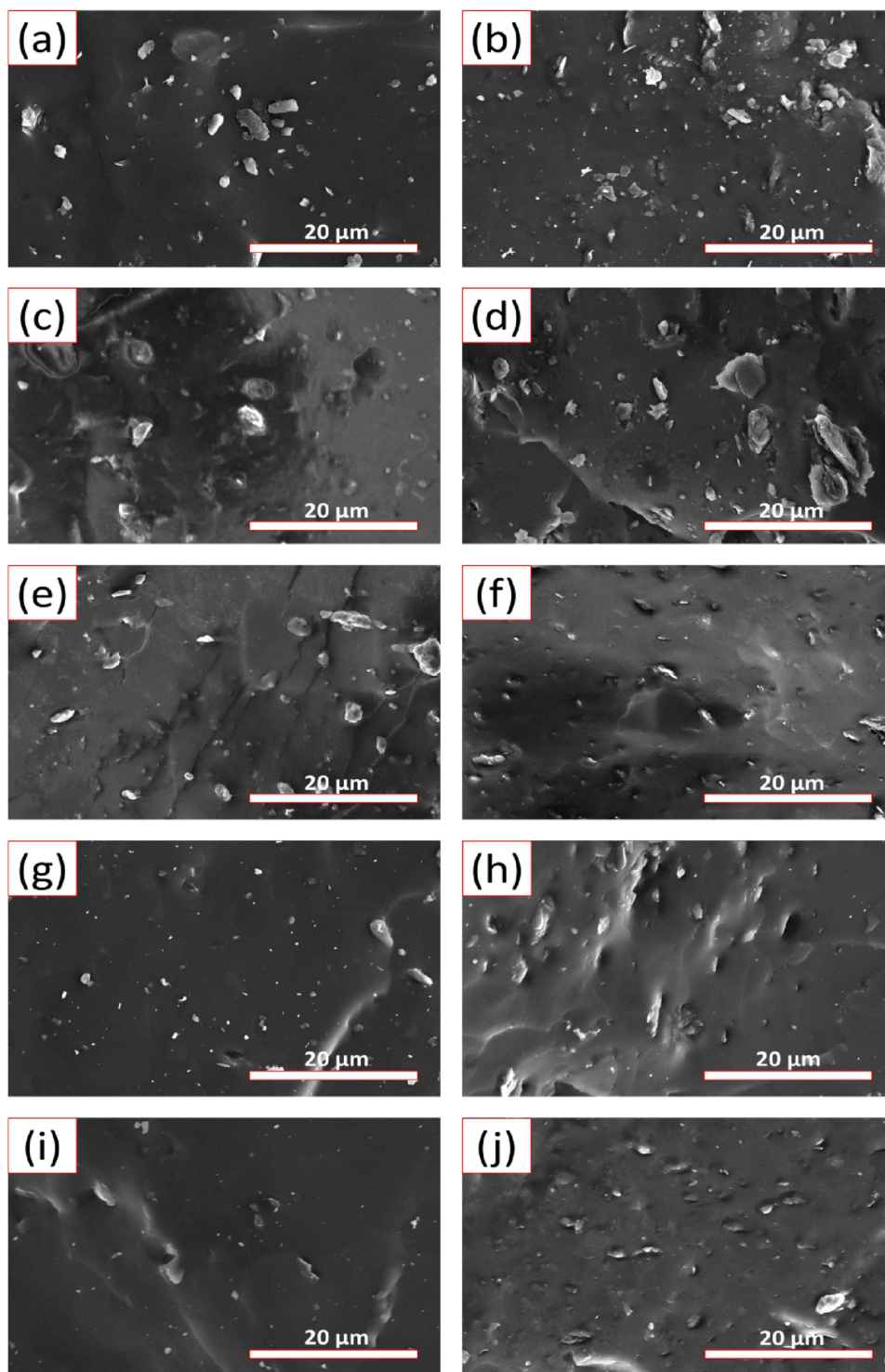


Fig. 6. SEM micrographs of vulcanizates filled with GnPs; 1 phr of GnPs: (a) neat GnPs (c) with BBP1 (g) BBP2 (e) with BMBP1 (i) BMBP2; 5 phr of GnPs: (b) neat, (d) with BBP1 (h) BBP2 (f) with BMBP1 (j) BMBP2.

with filler loading (Fig. 7a). At higher filler content, 7 phr and 10 phr, the presence of ILs increased the electrical conductivity, especially those with freely dispersed BBP1 and BMBP1. However, the values suggest the particles do not form a percolation pathway, even at 10 phr content. The addition of freely dispersed BBP into rubber compounds resulted also in the highest thermal conductivity (Fig. 7b). The results correlate with the conclusions reached from curing kinetics (Fig. 4), torque increment (Fig. 4b) and scorch time (Fig. 4c).

The photo-actuation ability was tested by using a TMA analyzer

device to monitor the sample length change upon illumination. Fig. 8 shows the sample change in length ( $\Delta L$ ) as a function of time during illumination. The neat GnPs resulted in significantly higher changes than the neat SBR (Fig. 8a). The photo-actuation ability of composites containing BBP is shown in Fig. 8b. The utilization of BBP as a free additive (BBP1) resulted in higher actuation changes. A similar trend was observed in the case of BMBP (Fig. 8c). It should also be noted that all the SBR composites maintained a stable baseline during irradiation.

Fig. 9a shows the  $\Delta L$  values as a function of the filler content for

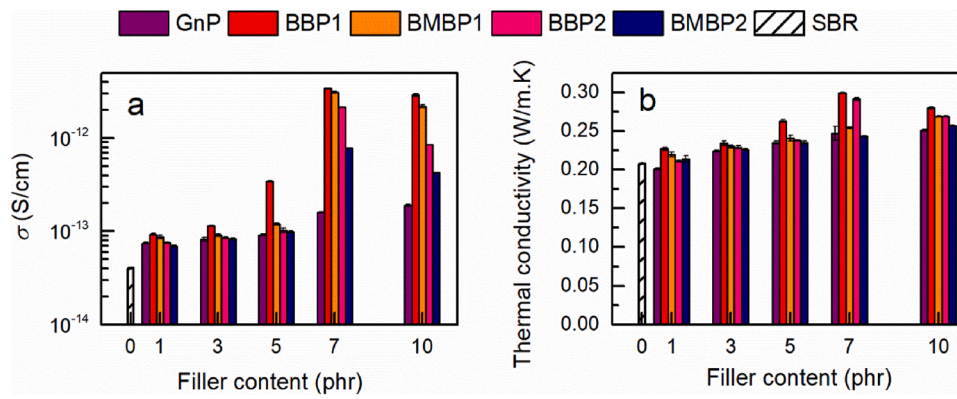


Fig. 7. Thermal conductivity (a) and electrical conductivity (b) of SBR composites versus the filler content.

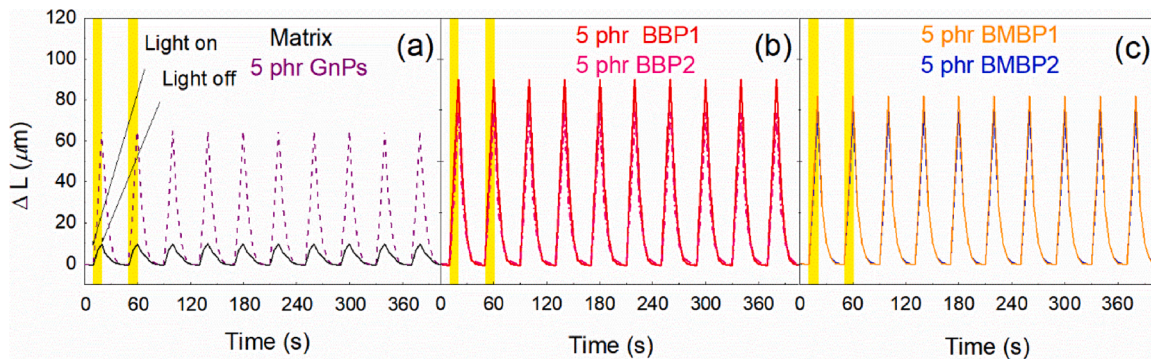


Fig. 8. Change in length ( $\Delta L$ ) as a function of time during the illumination of neat SBR and GnPs (a), composites with GnPs and BBP (b) and composites with GnPs and BMBP (c). The samples contain 5 phr of filler, and the light source intensity was 6 mW.

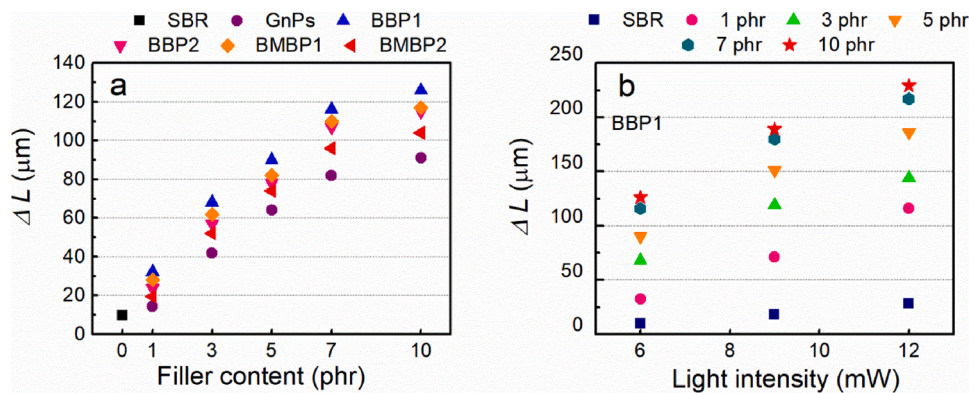


Fig. 9. Dependence of  $\Delta L$  on different filler contents; the light source intensity was 6 mW (a). The dependence of  $\Delta L$  of SBR composites filled with GnPs and freely dispersed BBP on various light intensities (b). The error bars of the individual measurements did not exceed the size of the symbols.

various types of ILs and fillers. The actuation ability was higher as the filler content increased and was the highest for ILs freely dispersed within the polymer matrix. In general, actuation is affected by the polymer chain dynamics, heat absorption and transport ability [16,33, 34]. These properties were reflected in the tensile tests and thermal conductivity measurements. Actuation was evaluated in the low stress-strain mode, i.e., when a force of 0.03 N was applied, which resulted in a maximum pre-strain of 10 %. The tests were performed at low strain; thus, the performance can be correlated with the trends obtained for the modulus 100 %. The modulus 100 % refers to the stress generated in a material at 100 % strain. The highest generated stress indicates a better confinement of the rubber chains in the composite system. In the case of the IL-enriched composites shown in Fig. 5a, the

freely dispersed BBP and BMBP showed the lowest moduli, while the highest moduli were found for BMBP immobilized on the GnPs surface. Here, the chain end mobility defined the possible actuation compression.

The composite based on neat GnPs showed an even lower modulus than the composites with freely dispersed ILs; however, their actuation ability was lower. This performance can be related to heat absorption and thermal conductivity. In case of the thermal conductivity (Fig. 7a), the highest values were obtained with freely dispersed ILs, and the actuation ability of these IL-enriched composites followed the same trend as the thermal conductivity. The neat SBR exhibited a lower thermal conductivity, which supported the results of the lower photo-actuation response. The explanation for the poor photo-actuation of

GnPs-enriched samples seems to be related to light absorption. The ILs probably contributed to the light absorption of the samples. Photo-absorbers have been successfully used to facilitate the absorption of light and promote photo-actuation [35]. It can be expected that ILs played a similar role in this study. Further experiments are necessary to confirm this hypothesis.

The composite with freely dispersed BBP was further tested in terms of its maximal photo-actuation response. Fig. 9b shows the dependence of the actuation stroke on various light intensities. At 12 mW, the maximal actuation stroke was 0.230 mm for SBR filled with 10 phr of GnPs and freely dispersed BBP. This high a value has not been reported to date. Compared to other crosslinked composites tested with the same equipment following the same protocol, a  $\Delta L$  of 0.040 mm at 12 mW was observed in a PDMS matrix filled with graphene oxide grafted with poly(glycidyl methacrylate) brushes [33], 0.037 mm was obtained for a system based on PDMS and graphene oxide grafted with poly(butyl methacrylate) [34], and 0.075 mm was obtained for a PMDS filler with graphene oxide grafted with poly(2-(trimethylsilyloxy)ethyl methacrylate) [36]. In addition to maximal actuation stroke, the increase in filler content from 7 to 10 phr did not lead to a linear increase in actuation stroke, Fig. 9b. The trends in both Fig.9a and Fig.9b suggest that the system approached maximal actuation strokes with 10 phr concentration, since the plateau region can be observed. Therefore, the higher concentrations were not investigated.

The photo-actuation constraint of 0.230 mm corresponds to 2.3 %. Higher values can be expected with increasing prestrain. The small prestrain obtained by using a 0.03 N force was used to allow for a proper comparison with other systems, which were tested following the same protocol. Among those, the SBR filled with freely dispersed GnPs and BBP showed the most promising results.

#### 4. Conclusion

Both types of butyl pyridinium ionic liquids were proven to affect the vulcanization kinetics of sulfur-vulcanized SBR rubber fillers with GnPs. The vulcanization kinetics determined from rheometric measurements revealed a higher activity for BBP, but the effect of immobilized BBP on the vulcanization kinetics was established at a higher filler content. The presence of a methyl pendant group and the immobilization of the ionic liquid on the GnPs surface provided the composite with the highest modulus and tensile strength, while freely dispersed BBP resulted in the composite with the highest thermal conductivity. This composite also demonstrated the best photo-actuation response of 0.230 mm. This high photo-actuation constraint has not been reported in crosslinked systems studied under similar conditions to date. These investigations may broaden the application of ionic liquids in the preparation of multi-functional rubber composites.

#### CRedit authorship contribution statement

**Magdalena Gaca:** Conceptualization, Methodology, Investigation, Resources, Writing - original draft, Writing - review & editing. **Marketa Ilcikova:** Conceptualization, Methodology, Investigation, Resources, Writing - original draft, Writing - review & editing, Visualization. **Miroslav Mrlik:** Conceptualization, Methodology, Formal analysis, Investigation, Visualization. **Martin Cvek:** Formal analysis, Investigation. **Cyril Vaulot:** Investigation. **Pavel Urbanek:** Investigation. **Robert Pietrasik:** Methodology, Formal analysis, Resources, Visualization. **Igor Krupa:** Methodology, Writing - review & editing. **Joanna Pietrasik:** Conceptualization, Methodology, Writing - original draft, Writing - review & editing, Supervision.

#### Declaration of Competing Interest

The authors declare no conflicts of interest.

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#### Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.snb.2020.129195>.

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