Interpenetrating polymer network composites containing polyurethanes designed for vibration damping

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Abstract: The paper is a literature review concerning the interpenetrating polymer network (IPN) composites containing polyurethanes that can be used for vibration damping in marine vehicles. The effects of filler introduction on damping, mechanical and thermal properties of IPN are compared. The addition of filler affects all relevant properties, in dependence on the filler type and quantity. Damping enhancement is achieved by introduction of each of studied fillers. The ability to optimize IPN structure and filler incorporation provides a way to obtain a material with desired features.

Keywords: polyurethanes, interpenetrating polymer network composites, vibration damping.

Kompozyty poliuretanowe z wzajemnie przenikającą się siecią polimerową stosowane do tłumienia drgań

Streszczenie: Artykuł stanowi przegląd literatury dotyczącej kompozytów poliuretanowych z wzajemnie przenikającą się siecią polimerową (IPN), wykorzystywanych do tłumienia drgań w pojazdach morskich. Porównano wpływ rodzaju i ilości wprowadzonego napełniacza na właściwości tłumiące, mechaniczne i termiczne IPN. Stwierdzono, że dodatek każdego z omawianych napełniaczy poprawia właściwości tłumiące. Optymalizacja struktury IPN i wprowadzenie napełniacza umożliwia otrzymywanie materiałów o pożądanych właściwościach.

Słowa kluczowe: poliuretany, kompozyty z wzajemnie przenikającą się siecią polimerową, tłumienie drgań.

Vibration and noise arise from operation of marine machinery and equipment such as prime movers, propellers, maneuvering devices, cargo handling, mooring and air conditioning systems [1]. In addition to negative influence on crew habitability, passenger comfort, and marine environment, excessive vibration can cause damage to the vibration source or surrounding structural parts [2, 3]. Therefore, damping of vibration is a critical problem in the ship and submarine design in order to improve structural stability, durability and performance [3]. Additionally, elimination of vibration in marine vehicles is an important acoustic stealth measure [4, 5].

One of the ways of vibration and noise reduction is use of damping materials [6, 7]. Good damping material has high damping capacity in the frequency and temperature ranges of interest, high mechanical strength and good thermal stability. Usage of polymer composite structures for ships and submarines relies on their advantages over metal constructions and ceramics which provide enhancement of the operational performance but at

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the same time reduce the ownership cost [8]. Mass reduction, corrosion resistance, design flexibility, elimination of expensive tooling, reduced acoustic and magnetic signatures of marine vehicles are some important composite characteristics for engineering of ship and submarine components such as engine end equipment foundations, turbine fan blades, anechoic tiles, masts and ventilation ducts.

The demand for high-performance polymer materials has posed a great challenge for scientists and engineers. The polyurethanes (PUR) and viscoelastic polymers are able to convert mechanical energy into heat [7]. Undesirable properties of pure PUR, a narrow range of mechanical energy absorption, low mechanical strength and poor heat resistance, can be improved by the preparation of interpenetrating polymer networks. Due to a complex microphase structure of interpenetrating polymer networks (IPN) a broad transition region could be obtained and good damping ability achieved [9—11]. The use of fillers leads to additional improvement of the damping properties since friction between polymer chain and filler and friction between two fillers contribute to damping [12].

This paper presents an overview of recent studies of IPN composites containing PUR designed for vibration damping.

INTERPENETRATING POLYMER NETWORKS CONTAINING POLYURETHANES

The physical and mechanical properties of PUR elastomers are related to phase separation of hard from soft blocks, which depends on the solubility parameter of the blocks, crystallinity of phases, temperature and sample thermal history [13, 14]. Hence, the degree of phase separation, structural and dynamical heterogeneity and morphology of the resulting phases (size, shape, orientation and domain associations) have been widely investigated. Their dependence on factors such as chemical composition, degree of crystallinity, molecular masses and the ratio of the segments, introduction and structure of the side chains or functional groups, preparation method and sample storage have been the subjects of numerous studies.

PUR can be used as damping and/or underwater acoustic absorption material [15—19]. The ability to absorb mechanical energy and transform it into heat is the highest around the glass transition temperature (T_g), where the rate (frequency) of coordinated molecular chain segment motion is equal to the rate (frequency) of the mechanical action [20]. Therefore, damping capacity is related to motional heterogeneity, which can be tuned by varying soft and hard segments composition and mutual ratio and introduction of functional groups and side chains [15, 21—26].

Damping capacity can be determined from dynamic mechanical properties. For two damping methods interesting from the engineering point of view, values of the loss modulus (E'') and the loss factor (tan δ) are important. An extensional damping requires high E'' value, while constrained layer damping requires high tan δ value [20]. In order to be efficient in wide temperature and frequency ranges encountered in real damping application, damping material should exhibit a high tan δ value (> 0.3) over the temperature range of at least 60—80 °C.

PUR often exhibit an insufficiently broad tan δ peak. Furthermore, usage of PUR in structural materials can be hindered by poor thermal stability and mechanical properties. In order to extend the temperature range with sufficiently high damping peaks and to obtain material with adequate properties, polyurethanes can be combined with polymers possessing high modulus and strength through preparation of interpenetrating polymer networks [10, 11].

The changes in the physical and mechanical properties of networks compared to the pure components depend on the degree of phase separation and morphology. The degree of phase separation in IPN depends on numerous factors such as the miscibility, mutual ratio, crystallinity and T_g of the components, the degree of cross-linking of individual networks and interconnection of networks, method and conditions (temperature, pressure) of preparation and the rate of forming [12].

In order to investigate the effect of blending on damping ability of PUR, IPN with various polymer components such as epoxy resin (ER) [11—13, 27—30] vinyl ester resin (VE) [11, 13, 31—33], unsaturated polyester resin [34], acrylates [11, 13, 35—38], poly(vinyl chloride) (PVC) [11—13, 39] and polystyrene [10, 11, 13, 40] have been extensively prepared and characterized.

The extent of the miscibility determines damping properties. Namely, a certain level of heterogeneity in segmental dynamics, which depend on the microstructure, is required for the efficient damping. Therefore, better damping properties can be obtained by increasing or decreasing compatibility of IPN components. Thus, varying the composition of PUR can influence damping performance. For example, polyether type crosslinked epoxy/PUR IPN is much better damping material than polyester type crosslinked epoxy/PUR IPNs due to the lower compatibility of polyether type PUR with epoxy [27]. The isocyanate index of PUR component is also an important factor that can be altered in order to optimize compatibility. A study performed on PUR based on poly(tetramethylene glycol)/epoxy resin (PTMG/ER) graft IPNs showed that the damping temperature range became broader and maximum value of tan δ increased with decrease of the isocyanate index as a result of enhanced miscibility [28]. Furthermore, long and soft chains of PTMG enhance the internal friction and the entanglement with the ER chains. Modifying of the second polymer chain structure also provides a way to alter damping properties. For instance, a choice of co-monomer of vinyl ester resin affects tan δ profile of PUR/VE

resin simultaneous polymer networks [30]. The introduction of acrylic esters instead of styrene as VE resin co-monomer leads to better compatibility and consequently to the improvement of damping properties. Similarly, IPNs of PUR—poly(methyl methacrylate) with incorporated glycidyl methacrylate or 2-hydroxyethyl methacrylate have higher and broader tan δ peaks as a result of enhanced mixing of two phases [35]. An additional option for altering the degree of phase separation and motional heterogeneity of IPNs is introduction of complementary functional groups in the backbone of network components in order to increase non-covalent interaction [36—38]. The component ratio is another parameter which has to be probed in order to create a material with the best properties. The optimal ratio of polyurethane and other polymer component depends on their types, molecular structure, and it is different for various properties and thus can vary significantly. For example, regarding damping properties, it varies from 40 to 85 wt % of PUR in IPNs with ER component [27—29]. Incorporation of the dangling chains or graded length side chains into one of the network components can increase the intramolecular friction and molecular relaxation, leading to the improvement of IPN damping properties if suitable type and length of chain has been chosen [13, 30]. Internetwork grafting reduces phase separation and offers opportunity to achieve broad and high transition zone by selecting proper grafting agent and adjusting its concentration [10, 11]. The usage of compatibilizers could improve interphase mixing and interfacial adhesion of incompatible mixtures and therefore expand the damping temperature range. The improvement depends on compatibilizer nature and quantity [11, 40]. Damping capacity also can be modified by IPN preparation method and conditions. For example, a study performed on simultaneous and gradiental interpenetrating polymer networks based on PUR and VE resin showed that controlling the techniques of gradient IPN renders possibility to manufacture noise and vibration damping materials [32].

FILLER EFFECTS ON DAMPING PROPERTIES OF IPN CONTAINING PUR

The use of fillers is one of the methods used to modify the properties of IPN [29, 31, 34, 41—54]. A number of fillers that exhibit some desirable features such as high aspect ratio and surface area, high elastic modulus, high thermal and oxidative stability, low price, low toxicity, have been introduced into the IPN and their effect on damping properties was examined. The results are summarized in Table 1.

T a b l e 1. Damping properties of IPN and composites

IPN composition	Filler	Optimal filler content, wt %	Loss factor		The width of the damping temperature range, °C	
			IPN	composite	IPN	composite
40PUR/60VE [31]	kaolin	10[a]	no data	no data	no data	no data
85PUR/15ER [29]	silica	7	—[b]	—[b]	—[b]	—[b]
70PUR/30PEMA [41]	silica	10	0.38[c]	0.54[c]	132	improved
70PUR/30ER [42]	carbon fiber	5	0.37[c]	0.65[c]	32	49
70PUR/30ER [42]	carbon fiber, nano-sized silica	5, 3	0.37[c]	0.73[c]	32	53
70PUR/30ER [43]	carbon fiber, hollow glass bead	5, 3	0.37[c]	0.72[c]	32	49
50 polyester type PUR-ER / 50 polyether type PUR [44]	zinc oxide whiskers	3	1.10[d]	1.44[d]	179	169
60PUR/40P(MMA-BMA) [45]	graphite	5	0.31[d]	0.53[d]	40	109
50PUR/50ER [46]	potassium titanate whiskers	3	1.06[b]	1.26[b]	47	47
50PUR/50ER [47]	calcium sulfate whiskers	3	1.06[c]	0.94[c]	47	52
50PUR/50ER [48]	hydroxyl-terminated polydimethyl-siloxane	15	1.06[c]	0.94[c]	47	53
50PUR/50ER [49]	hydroxyl-terminated polydimethyl-siloxane, carbon nanotubes	15, 0.5	1.06[c]	1.12[c]	47	61
50PUR/50ER [50]	carbon nanotubes	0.1	1.06[c]	1.04[c]	47	49
50PUR/50ER [51]	hydroxy-terminated liquid nitrile rubber	5	1.06[c]	1.11[c]	47	49
50PUR/50ER [52]	montmorillonite	3	1.06[c]	1.13[c]	47	53
50PUR/50ER [53]	aramid fiber	7	1.06[c]	0.93[c]	47	50
70PUR/30UPR [34]	BaTiO$_3$ fiber or nanopowder	70	—[b]	—[b]	—[b]	—[b]
PUR/ER/UPR [54]	mica	8	—[b]	0.92	—[b]	65

[a] Data shown for only one filler content.   [b] Due to graphical presentation of data it is not possible to determine the exact value.
[c] Measured at 10 Hz.   [d] Measured at 1 Hz.

One of the fillers that have been investigated is kaolin [31]. Simultaneous IPN composites based on blocked PUR and VE with various component ratios have been prepared and their mechanical and damping properties investigated. The effect of kaolin on tan δ was investigated for composite with 10 wt % of kaolin added to network containing 40 wt % of PUR and 60 wt % of VE. The tan δ peaks became broader and higher with introduction of filler. Mechanical properties were investigated for samples with 15 wt % of PUR containing various quantity of kaolin (5, 10, 15, 20, 25 and 30 wt %). The tensile strength, flexural strength, tensile modulus, and flexural modulus of composites increase to the maximum value for kaolin content between 20 and 25 wt % and then they decrease. Hardness increases and impact strength of IPN decreases with increasing filler content.

Silica (SiO_2) is suitable filler since its small and spherical particles with high surface have unique surface properties. Among studied IPN of PUR and ER with various component mass ratios (PUR:ER of 90:10, 85:15 or 65:35) sample with 85 wt % of PUR had the highest damping value and was chosen for preparation of composites with 3, 7 or 10 wt % of SiO_2 [29]. Introduction of SiO_2 contributes to compatibility and leads to improved damping. The broadest glass transition region was observed for nanocomposite with 7 wt % of SiO_2. Various amounts of SiO_2 (2.5, 5.0, 7.5 or 10.0 wt %) were also added to PUR composite with poly(ethyl methacrylate) (PEMA) with 70:30 components mass ratio 70PUR/30PEMA in order to investigate damping characteristics of composites [41]. Values of tan δ of these IPN do not vary significantly and they are higher than 0.3 spanning a temperature range of 132 °C. All composites exhibit higher tan δ peak and broader damping temperature range than IPN. Higher SiO_2 content results with better damping properties. Among investigated mechanical properties tensile strength and elastic modulus of composites increase with higher silica content, and elongation at break decreases with increased load of silica. Addition of nano-sized SiO_2 in combination with short carbon fibers (SCF) provides improvement of damping properties [42]. SCF possess high tensile strength, Young's modulus and thermal conductivity. The samples of PUR/ER IPN with 70 wt % of PUR content were loaded with SCF in order to obtain composites with mass contents of SCF 3, 5, 7 or 10 wt %. The tensile strength of the PUR/ER IPN composites was improved by SCF (up to content of 7 wt %, then decreases), while the impact strength deteriorates. Composite with 5 wt % of SCF was selected for incorporation of 1, 3 or 5 wt % of SiO_2. The tensile strength first increased and then slightly decreased with increasing nano-sized SiO_2 content. When the content of nano-sized SiO_2 was 3 wt %, the composites showed the best tensile strength. The impact strength improved for the sample with 1 wt % of SiO_2 and deteriorated for 3 and 5 wt %.

SCF were also tested in combination with micro hollow glass beads (HGB), characterized by relatively low density, good heat resistance, and sound insulation properties [43]. Composites with contents of SCF 5, 10, 15, 20, 25 or 30 wt % were prepared by combining PUR/ER IPN with 70 wt % of PUR. The tensile strength of the PUR/ER IPN composites was improved by SCF. To the composite with 5 wt % of SCF there were added HGB in the amount of 1, 3 or 5 wt %. Combination of SCF and HGB leads to improvement of damping properties, thermal stability and tensile strength, but the impact strength deteriorates.

Another type of filler investigated was tetrapod-shaped zinc oxide (T-ZnO) whisker [44]. Polyester type PUR crosslinked ER/polyether type PUR graft IPN with various component ratios were probed in order to find an optimal composition regarding damping capacity. IPN with equal mass ratio of polyester type PUR-ER and polyether type PUR was selected to examine the effect of T-ZnO on damping, thermal and mechanical properties. The values of tan δ of all studied composites (with 1, 3, 5 or 7 wt % of T-ZnO) are higher than that of unfiled IPN. After the initial increase with increasing amount of filler, tan δ decreases with the further increase of filler and the optimal content of filler is 3 wt %. The value of tensile strength also initially increases, reaching the maximum for composite with 5 wt % of T-ZnO and decreases with addition of more filler. The thermal stability was improved by the incorporation of the filler.

The effect of the incorporation of graphite on the damping properties of PUR/poly(methyl methacrylate-butyl methacrylate) [PUR/P(MMA-BMA)] semi-IPN was investigated [45]. The proportion and particle size of graphite were varied in order to examine their influence on tan δ, damping temperature range and frequency range. The composites had improved the damping performance, tensile strength and hardness in comparison to pure IPN. Two optimal contents were observed: sample with 5 wt % of graphite exhibited the best damping capacity, while composite with 7 wt % of graphite had the widest frequency range with the highest tan δ value of 0.65. The best damping properties were achieved with the finest graphite particles.

The IPN containing urethane elastomers with natural oil polyols can be used as damping materials [46—53]. Their synthesis and tailoring of properties are important from an environmental point of view since castor oil and other vegetable oils are rich and renewable resources and products of agriculture. A series of fillers were used to test effects on damping, mechanical and thermal properties of castor oil-based PUR/EP resin interpenetrating polymer networks with equal mass of network components. Composites with various content of potassium titanate whiskers (PTW) (1, 3, 5 or 7 wt %), calcium sulphate whiskers (CSW) (1, 3, 5 or 7 wt %), hydroxyl-terminated poly(dimethylsiloxane) (HTPDMS) (5, 10, 15 or 20 wt %), carbon nanotubes (CNT) (0.1, 0.3, 0.5 or 0.7 wt %), hydroxy-terminated liquid nitrile rubber (HTLN) (5, 10, 15 or 20 wt %), montmorillonite (MMT) (1, 3, 5 or 7 wt %) and aramid fiber (AR) (1, 3, 5 or 7 wt %)

were prepared and their properties were investigated [46—53]. Modification of properties depends on the filler type and content. Damping properties (maximum value of tan δ and/or damping temperature range) of pure IPNs were improved with all investigated fillers. The composites showed better damping properties under higher frequencies. The incorporation of filler results with improved tensile strength except in the case of HTPDMS, where after initial improvement, the tensile strength decreases with increase of filler quantity and becomes lower than that of pure IPN [48]. The impact strength was impaired with addition of CSW [47] and combination of HTPDMS and CNT above a certain amount of CNT [49]. Impact strengths of all other investigated composites increase in comparison to the IPNs. The thermal stability was better in all cases except composites with carbon nanotubes where the thermal decomposition temperature decreased slightly. Regarding effect of the filler content on tensile and impact strengths, tan δ and damping temperature range showed different trends with increasing filler contents, depending on the filler type. For example, the maximum value of tan δ increases with increase of AR content [53], while in the case of MMT it increases until certain composition (3 wt %), and decreases with further addition of MMT [52].

The fillers with dielectric and/or piezoelectric properties can transform mechanical into electric energy and finally to heat and contribute to damping. The effect of addition of various types (fiber or nanopowder) and amounts of barium titanate ($BaTiO_3$) to IPN containing PUR and unsaturated polyester resin (UPR) was investigated [34]. The influence of polymer components mass ratio (PUR:UPR of 50:50, 60:40 or 70:30) on damping behavior was studied. Based on these results IPN with 70:30 components mass ratio was selected for incorporation of $BaTiO_3$ fiber or nanopowder. The filler improves damping properties but fiber type $BaTiO_3$ more than nanopowder. A further improvement can be achieved by the polarization treatment due to synergistic effects caused by elastomeric damping, interfacial frictional damping and piezoelectric damping mechanisms. For the polarized filled IPN, the values of tan δ are higher than 0.3 for a temperature range of above 100 °C. Dielectric and damping properties in IPN are correlated, and study of dielectric constant and dielectric loss can provide information on damping properties of IPN-based composites.

The IPN composites containing PUR can be used as underwater acoustic absorption material. The underwater acoustic stealth performance of IPN with various content of PUR, ER, UPR and poly(dimethylsiloxane) (PDMS) depends on IPN microstructure [54]. The best results are obtained for IPNs with nanometer grade phase size and continuous phase boundary. Among tested fillers which were added to obtain composites with 8 wt % of mica, microballoon or nanosized SiO_2 the best performance parameters were achieved with mica.

A survey of published results shows that preparation of composites leads to the improvement of damping performance of IPN. Comparison of literature data reveals that the impact of filler introduction on all relevant properties (damping, mechanical and thermal) depends not only on the filler chemical structure, but also on particle size, shape and amount. Since damping capacity improves due to internal friction, which increases with augmented interface area, fillers with higher specific surface exhibit better properties. The changes in interface design imposed by the filler introduction initially contribute to damping ability. However, filler particles also influence phase continuity and available free volume leading to diminished mobility of molecular chains. Therefore, after certain filler concentration, in dependence on IPN and filler, further addition of filler leads to deterioration of properties. Hence, it is necessary to find optimal filler content. Polymer composites are complex systems where a number of interactions exist and changes of mechanical and thermal properties also may show different trends regarding filler concentration. Optimal values of various properties are mostly achieved at various compositions, and it is not possible to optimize material regarding all important properties.

CONCLUSIONS

Preparation of IPN composites containing PUR is a route to obtain materials for vibration and noise damping in marine vehicles. Their application relies on properties that depend on microstructure, which can be altered by numerous means, offering a way for preparation of versatile materials tailored for particular usage. Possible utilization of reusable materials and materials produced partly from renewable resources could contribute to eco-efficiency of maritime transport.

An addition of fillers affects damping properties, thermal stability and mechanical properties of IPN. The fillers improve damping capacity, depending on their chemical structure, content, particle size and shape. Mechanical and thermal properties can be improved or impaired by selected filler, since introduced interactions contribute to various properties differently.

In order to predict and control macroscopic properties of the materials, further investigations of filler-matrix interaction and dynamics of IPN composites on the molecular level are necessary. Various experimental techniques need to be used to provide more insight into complex polymer chain interactions, microphase structure and resulting properties.

Before the widespread use in shipbuilding, in addition to the cost-effectiveness, the new materials have to meet requirements of the ship safety standards. Therefore, determination of failure modes under blast, shock, collision and fire events are required.

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