Phononic glass: A robust acoustic-absorption material

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In order to achieve strong wide band acoustic absorption under high hydrostatic pressure, an interpenetrating network structure is introduced into the locally resonant phononic crystal to fabricate a type of phononic composite material called "phononic glass." Underwater acoustic absorption coefficient measurements show that the material owns high underwater sound absorption coefficients over 0.9 in 12–30 kHz. Moreover, the quasi-static compressive behavior shows that the phononic glass has a compressive strength over 5 MPa which is crucial for underwater applications.
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I. INTRODUCTION

In the past decades, various types of acoustic-absorbing materials are extensively studied, such as the porous metals, polymers, and composite materials.[1–4] However, these materials usually exhibit a poor acoustic-absorbing property under high hydrostatic pressure. This key issue hinders the development of deep-water noise-shielding materials which have both commercial and military uses. An energy-dissipative mechanism is necessary to design acoustic-absorbing materials for reduction of the sound wave amplitude.[5] For instance, rubber and polyurethane (PU) are usually used as underwater acoustic absorbing materials, which employ intermolecular friction to dissipate incident sound energy. The elastic modulus of these materials plays an important role in sound absorbing capability. Under high hydrostatic pressure in deep water, the elastic modulus of polymer is increased which impairs the acoustic absorbing ability in most cases. Therefore, it is necessary to develop the sound absorbing materials which are robust under high hydrostatic pressure in deep water. In addition, wide band absorbance is required for most underwater applications. In this paper, a composite material called "phononic glass" is reported, which is based on a novel idea of introducing the local acoustic resonators into a random network material structure. This type of material possesses both high mechanical strength and strong underwater sound absorption ability in a wider frequency band.

Robust and wide band sound absorbing materials for deep-water usage have attracted extensive research interest. Besides rubber and PU, aluminum foam is another classical underwater sound absorbing material. However, it suffers from low mechanical strength and poor corrosion resistance for deep sea use. Sound absorbance of traditional underwater anechoic coatings is usually considered to result from the multiple scattering from isolated inclusions dispersed in a polymer matrix.[3,6–8] Its mechanical strength is still not high enough to meet the demand for deep sea use, although relatively wide band absorption can be realized to some extent by this method. In short, a new concept in material design needs to be introduced to break through the limits of traditional underwater acoustic absorbing materials.

In this respect, the pioneer work of locally resonant phononic crystal (LRPC) is noticed.[9] In the past decade, LRPC had excited great interest because it can exhibit a phononic bandgap with a lattice constant 2 orders of magnitude smaller than the relevant sonic wavelength.[10–13] A recent theoretical simulation indicated that enhanced sound absorption can be obtained for LRPC materials by taking viscoelastic deformation into account.[14–16] It indicates that the LRPC can also be employed to design new acoustic absorbing materials. However, the LRPC concept was established on the sonic crystal which only exhibited a narrow acoustic bandgap around a specific frequency and poor mechanical property under high hydrostatic pressure. It is reasonable to deduce that the material containing a broad size distribution and multiple morphologies of LRPC-like resonant units might be used to broaden the acoustic absorbing band. However, the problem cannot be so simply resolved because LRPC units with broad size distribution are hard to be fabricated in practice and are not cost-efficient for industrialization.

Phononic glass is composed of a metal skeleton and PU infillings of an interpenetrating network structure. It was found that phononic glass exhibits a glass-like acoustic bandgap feature against the narrow characterized acoustic bandgap of phononic crystal. It can be regarded as multi-sized LRPC units dispersed and physically connected in a network matrix. The physical connection among LRPC units is helpful to excite more acoustic absorbing modes in phononic glass. In another aspect, the metal skeleton filled with PU polymers can provide higher mechanical strength to withstand hydrostatic pressure.

II. EXPERIMENTAL PROCEDURE AND MEASUREMENTS

The LRPC was produced by periodically embedding metallic balls coated with a compliant polymer layer into a stiff polymer matrix. The elastic modulus ratios of the metallic ball to the compliant and stiff polymers play an important role in adjusting the LRPC property. As an analogy to the

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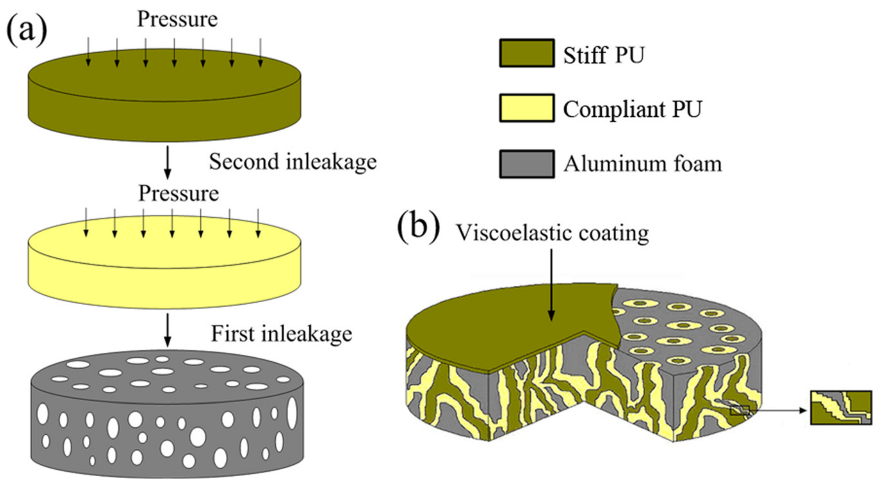

FIG. 1. (Color online) A schematic of synthesis and structure of phononic glass. (a) Synthesis scheme. (b) The internal structure of phononic glass. The stiff PU was chosen as the viscoelastic coating of characteristic impedance matching with water.

LRPC structure, a thin layer of compliant polymer was coated on the metal foam skeleton of the phononic glass, and then the stiff polymer was filled into the pores. According to the LRPC formation mechanism, three types of materials with different elastic modulus were employed to produce the phononic glass material. They are metal foam, compliant PU, and stiff PU. The aluminum foam, which the average pore size is 4–mm in diameter and the porosity is 81%, i.e., the relative density is about 0.19, was chosen as the metal skeleton. In order to reduce the sound reflection,[17] the incident sound wave should be introduced to the most extent into the acoustic-absorbing material by matching the acoustic impedance of stiff PU with water. The concrete procedure of material synthesis is described as follows. First, the uncured compliant PU was infiltrated into the aluminum foam to form a thin coating layer with thickness of about 0.3–0.6 mm along the interior wall of the porous aluminum. Secondly, the uncured stiff PU was filled into the compliant PU coated aluminum foam after totally curing this compliant PU layer. Finally, the stiff PU was totally dried. The densities of the compliant and stiff PUs are taken as 898 and 1076 kg/m^3, respectively. The shore A hardness values of the compliant PU and stiff PU are 43 and 77, respectively.

The scanning electron microscopy (SEM) images of the phononic glass were obtained by a LEO-1530-FESEM scanning electron microscope. The pulse tube method is often used to measure the underwater sound reflection coefficient in the field of water acoustics (Chinese National Standard GB/T 5266-2006), while the sound absorption coefficient can be deduced from the reflection coefficient.[4] The corresponding data in this paper were measured in the air back mode at the Institute of Acoustics of Chinese Academy of Sciences. For the sake of comparison, all testing samples were fabricated into the same dimensions with a diameter of 56 mm and a thickness of 10 mm. The quasi-static compression tests were conducted by a CMT4305 mechanical testing apparatus with a crosshead speed of 1.2 mm/min, i.e., a strain rate of 10^{-3} s^{-1}. The cylindrical specimens were cut from the ingots to dimensions with a diameter of 35 mm and a thickness of 20 mm.

III. RESULTS AND DISCUSSION

A schematic of the synthesis route and the material structure is shown in Figs. 1(a) and 1(b). To achieve PUs with different elastic modulus, the amount of the chain extender which can determine the stiff segment content in a PU macromolecule was adjusted in compliant and stiff PUs synthesis procedures. The volume ratio of stiff PU, compliant PU, and aluminum is 2:2:1. Figures 2(a) and 2(b) show the cross section photo of the sample and the stiff-compliant-stiff triple layer SEM image. It should be noticed that the composite materials and biomaterials[18–21] of interpenetrating network microstructures are usually found to exhibit unexpected merit due to the cooperative interaction among their component materials. The interpenetrating network structure in phononic glass can be considered to be constructed in a relatively large scale of millimeter-sized building blocks to work cooperatively for sound wave absorption.

Figure 3(a) shows that strong underwater acoustic absorbance with the absorbing coefficients over 0.9 can be achieved in a wide frequency range, while sound absorbing coefficients for each component material are much lower than 0.9. Figure 3(b) illustrates that a simple Reuss layered composite fabricated by two types of PU and aluminum slice in accordance with the volume ratio in the phononic glass

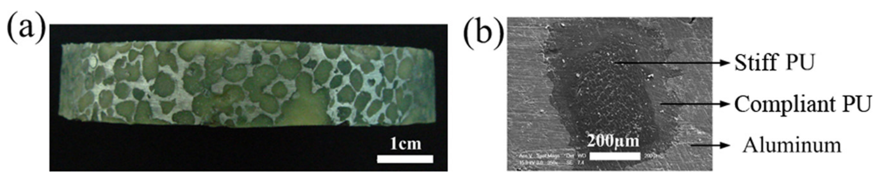

FIG. 2. (Color online) Optical and SEM images of a typical phononic glass sample. (a) The optical photo shows the cross-section view of the phononic glass sample in which the interpenetrating network structure can be clearly identified. (b) The SEM image shows a stiff-compliant-stiff multilayer structure around one LRPC-like unit in phononic glass.

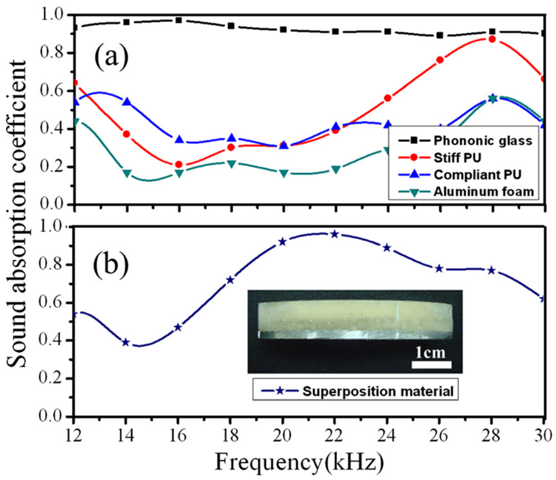

FIG. 3. (Color online) A comparison of underwater absorption coefficients for different materials that were fabricated with the same dimensions with a diameter of 56 mm and a thickness of 10 mm in 12–30 kHz and a model calculation of phononic glass. (a) The phononic glass' own sound absorbing coefficient over 0.9 in 12–30 kHz, while that could not be achieved by component materials. (b) The simple Reuss layered composite has a high underwater sound absorption coefficient in just 22 kHz.

have sound absorption coefficients over 0.9 just in a narrow sound bandgap. These results gave a solid hint on achieving unique properties of the new material due to network structural design. Component materials (Aluminum foams and PU) and a simple Reuss layered composite are representative of the traditional underwater acoustic absorbing materials. Their sound absorbing behaviors have been well studied in the literatures.[22–26] The underwater acoustic absorption capability of the phononic glass is much better than that of component materials from the measured spectrum. Its strong acoustic absorption characteristic is not originated from its component or simple linear superposition of the component materials. It is reasonable to deduce that the combination of LRPC structure units and the cooperative effect from the interpenetrating network play an important role in achieving a strong wide band acoustic absorbing material.

As mentioned previously, phononic glass can be considered as an interpenetrating network structure which contains inverse LRPC sound absorbing units. That LRPC can exhibit effective negative elastic density at a certain frequency is due to the existence of sound resonators dispersed in the matrix. The physical essence of these metamaterials with negative density originated from the elastic density match among component materials and the geometrical shape structure of a sound resonator.[9,27,28] The locally resonant property of LRPC is independent of the periodic arrangement of resonators in the matrix.[9] The phononic glass possesses inverse LRPC structure units, as shown in Fig. 2(b). A stiff-compliant-stiff triple layer structure in LRPC is preserved in phononic glass which might guarantee the sound resonant feature. If viscoelastic scattering in materials is considered, the large viscoelastic energy dissipation was expected at locally resonant frequency.[14–16] It is reasonable to deduce that the strong sound absorbance partially resulted from this energy dissipation mechanism, even if not totally. Subsequently, a direct deduction of the above hypothesis is that the resonant bandgaps can be used to estimate the frequency at which strong absorbance could occur. In the geometrical point of view, resonators in the phononic glass have a broad size distribution and various shapes. This can be invoked to explain the presence of the broadened peak in the sound absorption spectrum. It is well known that an acoustic wave could be largely attenuated by the multiple scattering effect.[3,29] In phononic glass, resonant scatters are substantively and closely connected by interpenetrating networks against discrete and sparse distribution of resonators in LRPC. Multiple scattering effects, in most cases, can be greatly strengthened by this type of close network linkage.

The lumped-mass method is a common approach to study a bandgap in LRPC.[30–33] In previous works, it had been demonstrated that the lumped-mass method can be used to study the acoustic absorbing bandgap of the material consisting of LRPC units.[34] Therefore, to elucidate the physical effect of the new material, the lumped-mass method is employed to estimate possible sound resonant bandgaps in phononic glass. A resonant unit structure [indicated by the solid square in Fig. 4(a)] can be abstracted as a cylindrical core with two coaxial coating layers as shown in Fig. 4(b). For convenience of calculation, the fitted radius of core cylinder and two coaxial layers were $r_1 = 0.75$ mm, $r_2 = 1.1$ mm, and $r_3 = 1.25$ mm in light of the volume ratio of the three components and the average sizes of different unit cells. One unit can be further simplified by the spring-mass model,[10,12,32,33] as shown in Fig. 4(d).

Two vibration blocks with masses m_1 and m_2 are connected by a virtual spring with stiffness coefficient k. Basically speaking, m_1 is the mass of the central stiff-PU core and m_2 is the mass of the outer aluminum shell. The main effect of the inner compliant-PU layer is to provide a type of pliable connection between the stiff-PU core and the aluminum shell. Therefore, the inner compliant-PU layer can be described as a virtual spring. However, its mass had to be assigned to the surrounding medium. The mass of part a in Fig. 4(d) is added into mass m_1 in the calculation. The treatment for part b is a little bit cumbersome.

An appropriate fraction of part b mass was added into m_2, while the rest of the part b mass is assigned to the stiff-PU core m_1. m_1 and m_2 can be calculated through the following formula:

$$m_1 = \rho_{H-\mathrm{PU}} \cdot \pi r_1^2 + m_b \cdot \frac{1}{1 + \dfrac{m_b + \rho_{H-\mathrm{PU}} \cdot \pi r_1^2}{\rho_{S-\mathrm{PU}} \cdot \pi(r_2^2 - r_1^2) + \rho_{Al} \cdot \pi(r_3^2 - r_2^2)}}, \tag{1}$$

$$m_2 = \rho_{Al} \cdot \pi(r_3^2 - r_2^2) + m_a + m_b \cdot \frac{\dfrac{m_b + \rho_{H-\mathrm{PU}} \cdot \pi r_1^2}{\rho_{S-\mathrm{PU}} \cdot \pi(r_2^2 - r_1^2) + \rho_{Al} \cdot \pi(r_3^2 - r_2^2)}}{1 + \dfrac{m_b + \rho_{H-\mathrm{PU}} \cdot \pi r_1^2}{\rho_{S-\mathrm{PU}} \cdot \pi(r_2^2 - r_1^2) + \rho_{Al} \cdot \pi(r_3^2 - r_2^2)}}, \tag{2}$$

where m_a and m_b are masses of parts a and b. They can be calculated through the similar formula in Ref. 33. The stiffness k depends not only on the elastic modulus of the compliant PU layer, but also on the geometrical shape of its part b.

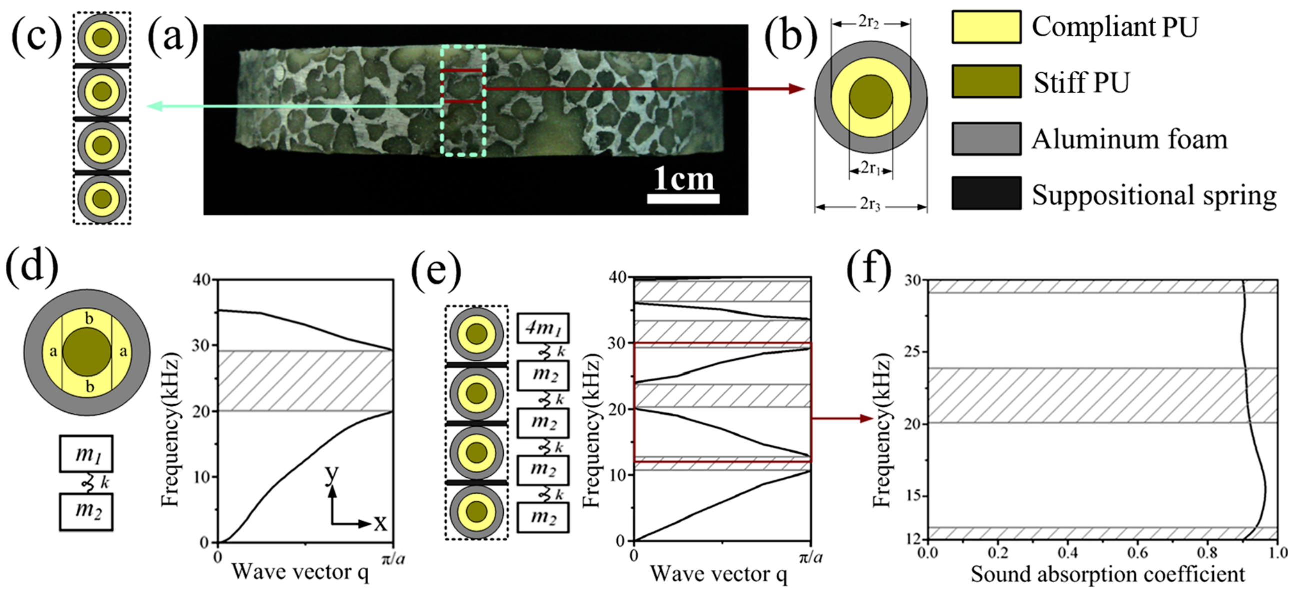

FIG. 4. (Color online) The model simplification process of phononic glass and spectral characteristics. (a) The phononic glass has an internal random interpenetrating network phase structure. (b) A unit cell of this random structure could be approximately described as coaxial cylindrical inclusions with two coaxial coating layers. (c) Along the normal incidence direction of sound wave, a phononic glass sample of 1 cm thickness can be predigested as four vertical units connected by virtual spring. (d) The vibration mode of a unit of phononic glass can be predigested as a double-oscillator model because it is similar to that of the vibration mode of LRPC. (e) Along the normal incidence *y* direction of an acoustic wave, a fourfold approximate unit of phononic glass can also be predigested as a fivefold-oscillator model. Four vertical units connected by a virtual spring have four relevant bandgaps of 10–40 kHz. (f) Four sound bandgaps ignoring viscoelastic scattering corresponded with measured absorbing spectrum.

In terms of physics, only the mass of part b [shown in Fig. 4(d)] took part in the resonant reflection. Therefore, the stiffness k can be calculated by the similar formula in Ref. 33. The calculated vibration frequency spectra are shown in the right panel of Fig. 4(d). The gray hatched area indicates the phononic bandgap which is induced by the locally resonant unit. It can be seen that the bandgap can only cover a narrow frequency range from 20–29 kHz. This is a natural result because only the vibration mode from an individual unit is considered in the calculation. In fact, strong connections among locally resonant units exist in our samples. Neglecting the influence of these physical connections on the resonant vibration mode is unreasonable. To estimate this influence, it is assumed that LRPC units are also linked by the virtual springs of stiffness k in the incident acoustic wave direction. For the normal incident sound wave, the sample of thickness 1 cm contains four LRPC units in the thickness direction, as shown in Fig. 4(c) and the left panel of Fig. 4(e). The central panel of Fig. 4(e) shows the corresponding mass-spring model which contains five mass blocks and four virtual springs. The topmost mass block is assigned 4 times the mass of m_1 under the consideration that the central stiff PU cores are rigidly connected and form a stiff PU network matrix in the sample. Therefore, a simple linear superposition for the core mass m_1 is used.

Here, the same virtual spring as that in the calculation of an individual LRPC unit vibration mode is also employed to estimate the cooperative vibration among LRPC units. This is due to the consideration that the compliant PU layer is uniformly coated on the aluminum skeleton and formed a compliant PU network structure. Therefore, a similar compliant PU layer connects the LRPC units as it works in the interior of an individual LRPC unit.

The right panel of Fig. 4(e) shows the corresponding calculated resonant bandgaps. An obvious feature is that multiple resonant bandgaps appeared and covered different frequency ranges instead of a single bandgap in Fig. 4(d). When the solid square in Fig. 4(e) is enlarged, it can be seen that bandgaps of four units covered the acoustic absorbing spectrum of the phononic glass from 12–30 kHz, as shown in Fig. 4(f). This spectral feature implies that the resonant sound reflection would happen in a wider frequency range if the connections between the individual units of the phononic glass are taken into account. From Fig. 4(f) it can deduce that the interpenetrating network structures in the present samples produced strong connections between LRPC units and resulted in a broad strong absorption in the acoustic absorption curve shown in Fig. 3(a). Because broad size distribution and various shapes of locally resonant units are not considered in the calculation, it is reasonable to be understood that the calculated bandgaps cannot cover the full strong-sound-absorbing frequency range as in the experimental sound-absorbing spectrum.

The phononic glass not only has strong absorption properties but also has excellent compression performance due to its unique interpenetrating network phases. Figure 5 exhibits a comparison of compression resistance experimental results of materials. The mechanical compression test was performed up to failure when the sample was loaded for the first time. The phononic glass shows compressive strength over 5 MPa, while that could not be achieved by PU and aluminum foam. This result demonstrated that the new material is a robust underwater acoustic-absorption material. The compressive strength of this composite material originated from the collective effects achieved by structural design. The interpenetrating network structure unified strong wide band

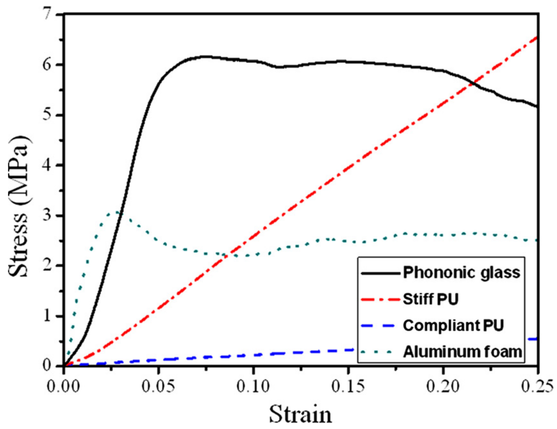

FIG. 5. (Color online) Compressive stress-strain curves of phononic glass and its component materials. The compression resistance of phononic glass was much better than its components.

sound absorption capability and high mechanical strength in the phononic glass, while it appears to be conflicting in normal circumstances. Two types of PU and aluminum foam show separately the typical behavior of elastomer and lower relative density metallic foams. For stiff and compliant PUs, the stress slowly and monotonously increases with increasing strain and no yielding point is observed. It means that stiff and compliant PUs have incompressible property. The compressive stress-strain curve of the aluminum foam is characterized by three distinct regions, i.e., linear elastic region, plateau region, and densification region. However, the phononic glass had a completely different stress-strain behavior from PU and the aluminum foam. Its anti-compression capacity is much better than that of aluminum foam. It should be attributed to the interactions between the PU and the cell wall of aluminum foam. In the initial stage of compression, the phononic glass experiences short term nonlinear elastic deformation which was affected by the thin stiff PU coating. With the pressure increase, curves of the phononic glass and aluminum foam tend to be parallel. In this stage, the stress dominantly arises in the cell wall of the aluminum foam due to a much higher strength of phononic glass over that of PU. Thus, the response at this stage reflected the aluminum foam's behavior. When the force approaches the yield point of foam aluminum, the phononic glass is able to keep the compression performance as normal because two types of PU impose a resistance to the deformation of the cell wall once the interstices are closed. At this time, lateral deformation in two types of PU occurred under the compressive stress. This lateral deformation was also hindered by the cell wall, which conversely enhanced the resistance to the bending of the cell wall. Accordingly, the densification process was delayed until the compressive stress was large enough to break the cell wall to allow two types of PU to flow out.[35]

IV. CONCLUSIONS

Based on the theory of LRPC and interpenetrating network structure, the phononic glass with a high performance for underwater sound absorption had been developed. Theoretical and experimental results revealed an excellent sound absorption capability due to the combination of LRPC structure units and cooperative effect from the interpenetrating network. The lumped-mass method can be employed to study its anomalous sound absorbing mechanism. Moreover, phononic glass showed better compression resistance. This paper gives a clue to the material design through a combination of functional structure units and cooperative effects from the interpenetrating network. This original design concept can also be extended to other application areas.

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