Article

Experimental Study on Properties of Ultrasonic Coupling Agent with Graphene in NDT

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Abstract: An ultrasonic coupling agent, as an acoustic medium between the ultrasonic probe and the surface of the specimens, is indispensable in Nondestructive Testing (NDT). Whether it is liquid, air, or solid coupling agent, the problem of improving the efficiency of ultrasonic propagation in a coupling agent is one worth studying. Glycerol and hydrogels are two common liquid coupling agents in NDT. This study intended to investigate the effect of graphene addition on the performance of these coupling agents in NDT. Firstly, based on the theory of acoustic impedance matching, the authors established an index system to evaluate the performance of ultrasonic coupling agent by experiments. Secondly, hydrogel–graphene and glycerol–graphene composite coupling agents were prepared by adding three-dimensional graphene structure powders with mass fraction of 0.25%, 0.5%, 0.75%, and 1% to CG-98 hydrogel coupling agent and HG-99 glycerol coupling agent, respectively. Corresponding experiments were conducted on these composite coupling agents. Peak-to-peak value, attenuation coefficient, and energy value of first echo are calculated at different frequencies. The experimental results showed that graphene can significantly improve the ultrasonic propagation performance of hydrogel and glycerin coupling agents. In addition, when the mass fraction of graphene added was 0.75%, the coupling agent had the best performance. Finally, we measured the acoustic impedance values of the composite couplings with different graphene contents to demonstrate the reliability of the experimental results.

Keywords: graphene; ultrasonic coupling agent; time-domain analysis; acoustic impedance

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1. Introduction

In recent years, ultrasonic nondestructive testing technology has been widely used in industrial testing, and has become an important means in the field of nondestructive testing. In ultrasonic testing, the selection of a coupling agent is very important [1]. These coupling agents improve detection accuracy in three main ways [2]. First, when the ultrasonic coupler is filled between the surface of the specimen and the surface in contact with the probe, the ultrasonic coupler removes air and fills uneven pits and gaps so that the transducer is in close contact with the surface of the specimen. Secondly, the ultrasonic coupling agent can reduce the acoustic impedance difference between the surface of the ultrasonic probe and the surface of the specimen, and reduce the ultrasonic energy reflection loss. Therefore, the energy transmission loss between the probe and the specimen is small. Third, the ultrasonic coupling agent as a “lubricant” can reduce the friction between the surface of the ultrasonic probe and the specimen, so that the ultrasonic probe flexible slides along the surface. Therefore, well-performing coupling agents can ensure that the ultrasonic wave propagates in the measured part more effectively, and enough sound intensity transmittance is obtained, so as to achieve the purpose of optimizing the detection.

The coupling effect is also influenced by the properties of coupling agent materials. Bhadwal et al. [3] used a thin foil of annealed silver as a filler material/coupling agent at each component interface. Preliminary high temperature tests indicated that this coupling method was suitable for high temperature and achieved signal quality comparable to that at room temperature with ultrasonic gel. K. Dugmore et al. [4] compared the coupling effects of several commonly used coupling agents, such as glycerin, petroleum jelly, and 'Kendon Laboratories CC' Ultra-Gel. C. Li and E. Nordlund [5] argued that liquid coupling (grease, epoxy resin and water) was better than that of metal foil (aluminum), and analyzed the effect of coupling agent on ultrasonic propagation of longitudinal wave and shear wave. J. F. Couvreur [6] analyzed the wave velocity and attenuation as functions of stress and coupling agent. Differences were more notable on attenuations, which were equally evaluated in time and frequency domains. Lead foil, grease, and honey scored the best among metal foils and visco-liquid compounds. B. Piwakowski [7] raised the selection of the best combination wedge/coupling, performed by searching separately for the best wedge material and the best coupling material. Three wedge materials and five coupling agents were tested. Among the three tested wedge materials, Teflon was found to be the best. The investigation on the coupling agent showed that the gel type materials were the best solutions. The 'thick' materials displaying higher viscosity were found as the worst. Zhang et al. [2] proposed a new method to improve the sound transmission performance of lipid-like coupling agent to form a new grease-like coupling agent by combining different metal ions such as Li^+ , Na^+ , and Ca^{2+} with base grease. The experimental results showed that compared to other oil-based coupling agents, except for slightly delayed peak response time, lithium-based coupling agents had higher energy transfer efficiency of pulse signal and better pulse vibration waveform, which could improve energy transmission performance and greatly improve the sensitivity of ultrasonic flaw detection.

Graphene is a single carbon layer consisting of sp^2 hybrid carbon atoms, and its thickness is the diameter of a single carbon atom. Carbons atoms are connected by σ and π bonds [8]. Due to its unique structure, graphene has excellent mechanical and electrical properties [9,10]. As a two-dimensional carbon nanomaterial with a dense single-atom structure, graphene has attracted extensive attention due to its unique properties [11,12]. However, graphene has poor processing performance, and is easy to aggregate; it is not widely used in practical engineering. At present, three-dimension structured graphene has good thermal stability, mechanical strength, and other characteristics. Besides, three-dimensional graphene is an attractive structure that effectively prevents two-dimensional graphene from aggregation or accumulation. It not only maintains the intrinsic properties of graphene, but also has a unique self-supporting structure and porosity. Water-based polymer gels are a new three-dimension functional polymer material, which can swell in water but cannot dissolve in water. Hydrogels not only perform good swelling property, but also have ideal mechanical strength. Moreover, graphene is incorporated into water-based polymer gels to enhance the mechanical strength of the obtained hydrogels. Moghaddam M B et al. [13] determined the rheological properties of graphene–glycerin nanofluids at different mass fractions (0.25–2%) and temperatures (20–60 °C). The experimental results showed that the viscosity of graphene–glycerin nanofluids increased with the increase in mass fraction and decreased with the increase in temperature. At the same time, graphene–glycerin nanofluids were found to have strong shear thinning behavior. At 20 °C, 6.32 s^{-1} shear rates, the viscosity of glycerol was increased by 401.49% with the addition of 2% graphene. Moghaddam M B et al. [14] calculated the solubility parameters of pure graphene in pure glycerin. Then he found that graphene could form stable dispersion in glycerin, and the dispersion coefficient of graphene/glycerin nanofluids decreased with the increase in graphene layers. Graphene can improve the properties of polymer hydrogels. Liangfei et al. [15] underlined that graphene be incorporated into the mixed solution of sodium alginate and CaCO_3 to enhance the mechanical strength of the hydrogel.

In this study, different proportions of graphene were added to hydrogel and glycerin coupling agents to prepare a hydrogel–graphene composite coupling agent and a glycerin–

graphene composite coupling agent. An experimental platform for ultrasonic testing was built to study the performance changes of different composite couplings after graphene was added.

2. Principle and Performance Index of an Ultrasonic Coupling Agent

2.1. Principle of Acoustic Impedance

The principle of application of ultrasonic coupling agent is the theory of acoustic impedance. When the acoustic impedance of the specimen matches the acoustic impedance of the coupling agent better, the smaller the sound energy loss is, and the more ultrasonic waves can enter the tested part successfully. The principle of acoustic impedance matching is shown in Figure 1.

Figure 1. Principle of acoustic impedance matching.

The acoustic impedance of the probe surfaces is Z_1 , the acoustic impedance of the coupling agent in thin layer medium is Z_2 , and the acoustic impedance of the tested part is Z_3 , and the corresponding wavelengths of ultrasonic waves in the three media are $\lambda_1, \lambda_2, \lambda_3$ respectively. The transmission coefficient of ultrasonic intensity t_I is expressed by the ratio of transmitted ultrasonic intensity I_T to incident ultrasonic intensity I_i :

$$t_I = \frac{I_t}{I_i} = \frac{4Z_1Z_3}{(Z_1 + Z_3)^2 \cos^2\left(\frac{2\pi}{\lambda_2}d\right) + \left(Z_2 + \frac{Z_1Z_3}{Z_2}\right)^2 \sin^2\left(\frac{2\pi}{\lambda_2}d\right)} \quad (1)$$

In order to maximize t_I , the derivative of Equation (1) with respect to Z_2 was obtained, and set it as 0, and you get the following formula:

$$Z_2 = \sqrt{Z_1Z_3} \quad (2)$$

The equations above indicate that the ultrasonic wave enters the tested part completely and there is no loss in the coupling agent thin layer medium. Therefore, the closer the square root value of the acoustic impedance product between the sensor and the specimens tested is to the acoustic impedance value of the coupling agent, the better the ultrasonic propagation efficiency will be. The corresponding acoustic impedance value Z is expressed as:

$$Z = \rho C \quad (3)$$

where ρ is the material density, while C is the speed of ultrasonic wave in material.

Based on physical principles, the acoustic impedance value Z and the speed value C are determined by material properties, independent of other conditions. In this study, the experimental system established was employed to obtain the acoustic impedance values of the sensors and the specimens. According to Equation (2), it is necessary to change the acoustic impedance value of the coupling agent to approximate the square root of product of the acoustic impedance of the sensor and the specimens. Only in this way can the transmission coefficient of sound intensity be maximized. Corresponding high transmittance of ultrasonic wave was obtained in this way. In general, the acoustic impedance value of the coupling agent is changed by adding graphene into the coupling agent, in which Equation (2) was satisfied.

Compared with the specimens tested, the density of the gel and glycerol coupling agents are relatively small, so the acoustic impedance value greatly differs from that of the specimens. Therefore, some energy will be reflected back in the ultrasonic propagation, resulting in a small transmission coefficient. When graphene powder is added to the coupling agent, the density of the composite coupling agent is increased; therefore, acoustic impedance matching between the coupling agent and the tested specimens was easy to satisfy Equation (2), so as to optimize the transmission effect of ultrasonic wave. In addition, the appropriate addition of graphene can increase the lubricity of the coupling agent [16], which can reduce the friction between the sensor and the specimen to increase the service life of the sensors.

2.2. Performance Evaluation Index

In this work, the performance of the composite coupling agent was evaluated by analyzing peak-to-peak value, attenuation coefficient, and energy value of the received signal in the time domain. Peak-to-peak value refers to the difference value between the highest and lowest value of a signal in the same period. Peak-to-peak values V_p are expressed by the following formula:

$$V_p = V_{\max} - V_{\min} \quad (4)$$

$V_{\max}$ and $V_{\min}$ denote the maximum and minimum amplitude values of the first echo received in the time domain, respectively.

The attenuation of ultrasonic wave is more sensitive to material properties [17]. The phenomenon of energy loss will occur when sound waves propagate through a medium or across media's boundaries [18]. In addition, the ultrasonic wave velocity is directly linked to the mechanical properties of an elastic medium: Young's modulus (E), Poisson's ratio (ν), and density (ρ) [19]. When the propagation medium changes, it will cause energy loss and attenuation. The main attenuation of ultrasonic wave originates from absorption and scattering, and depends on the properties and thickness of the medium [20]. The ultrasonic attenuation coefficient of single-layer steel can be expressed as:

$$\alpha = -\frac{10}{d(n-m)} \lg \frac{P_n}{P_m} \text{ (dB/mm)} \quad (5)$$

where d value is the thickness of the steel plate; P_n and P_m denote echo sound pressure amplitude at the m times and n times, respectively. In this paper, the amplitude of the first and second echo was selected to calculate P_n and P_m , namely $n = 1$ and $m = 2$.

In this study, the energy value E is used to indicate the amount of energy passing through the specimen. The energy calculation formula [13] of the signal can be expressed as:

$$E = \int_0^t A^2 dt \quad (6)$$

where A is the peak value of the primary echo signal at time t .

In the time domain diagram, the amplitude square of the ultrasonic primary echo signal is integrated, and the obtained integral value is taken as the energy value of the ultrasonic primary echo signal.

In addition, the pulse method is used to measure ultrasonic wave velocity in the coupling agent. Ultrasonic sound velocity can be obtained by the following formula:

$$C = \frac{L}{\Delta t} \quad (7)$$

where Δt is the time between the excitation signal and the received signal observed on the oscilloscope, L is the thickness of the coupling agent. In this work, $L = 20$ cm.

According to Equation (3), the acoustic impedance of the composite coupler was calculated. The acoustic impedance of the coupling agent was obtained when graphene

was added to the coupling agent or not; the influence of graphene on the performance of the coupling agent was easily obtained.

3. Experimental Section

3.1. Experimental Materials and Equipment

The raw materials were CG-98 hydrogel ultrasonic coupling agent (Hengcan, Jining, China), which consists of surfactants, rust inhibitors, softeners, and wetting agents; HG-99 glycerin ultrasonic coupling agent (Hengcan, Jining, China, ingredient: 95% glycerin); and a three-dimensional graphene structured powder (BET: 1810 m²/g, electric conductivity: 1.0 × 10³ S m^{−1}, producer: Graphene Research Center of Guangxi University)

The experimental equipment were two longitudinal wave sensors with central frequency of 5 MHz, RITEC-RAM-5000 snap-nonlinear high energy ultrasonic testing system, an oscilloscope (Tektronix MDO3022, Beaverton, OR, USA), a microscope (Axio Scope.A1 and Cam MRc 5, Oberkochen, Germany), a thermostatic magnetic stirrer (MYP11-2A), a specially processed ASTM 1020 steel specimen with surface roughness less than 5 µm [21] and dimensions of 150 mm × 30 mm × 20 mm, four plastic sheets with thickness of 0.2 mm, a rubber head dropper with volume of 2 mL, and a precision pallet balance (DH-JCS).

3.2. Preparation of the Graphene Composite Coupling Agent

First, 30 g glycerol coupling agent was placed in a beaker; then, a certain amount of weighed three-dimensional graphene was added. The coupling agent was stirred (600 RPM, 60 °C) for 30 min with a stirrer. Finally, it was left to stand in air for 8 h to eliminate bubbles and obtain the glycerol–graphene composite coupling agent. The same method was used to prepare two different compound couplings with different mass fractions.

3.3. The Experimental Method and Schematic Diagram of the Experimental Platform

Firstly, the distribution morphology of graphene in different mass fractions of graphene composite couplers were observed by a microscope. In order to better describe the distribution of graphene in the microscopic image, Pro Imaging version 1.1 (OPTON, Beijing, China) was used to identify the size of the graphene particles and obtain particle diameter information. Then, an experimental platform was established; the experimental schematic diagram is shown in Figure 2. The prepared graphene composite coupling agent was uniformly applied on the upper and lower surface of ASTM 1020 steel. The sensors were fixed at the place where the coupling agent was applied for testing. Four 0.2 mm plastic sheets were added between the sensor and the specimen to keep the thickness of the coupling agent constant. The dead weight of the sensor was used as pressure to keep the pressure of the sensor constant. The RITEC-RAM-5000 emitted a sinusoidal signal of 10 cycles used as an excitation signal and an oscilloscope was used to observe the waveform received by the receiving sensor at the excitation frequency of 5 MHz; 10 groups of experimental results were recorded. By changing the excitation frequencies, corresponding experiments were conducted on the graphene composite coupling agent with different graphene contents. At last, Origin Pro 2018 (OriginLab, Northampton, MA, USA) software was used to calculate the peak-to-peak value, attenuation coefficient, and energy value of each data. The calculation formula of the final result of each index can be expressed as:

$$X = \frac{1}{n} \sum_{i=1}^n x_i \quad (8)$$

where x_i is the calculated value of each test, n is the number of tests, and $n = 10$.

Figure 2. Schematic diagram of experimental platform.

4. Experimental Results

4.1. Microscopic Image of the Hydrogel–Graphene Composite Coupling Agent

As shown in Figure 3 and Table 1, Figure 3a is the micrograph with a mass fraction of graphene of 0.25%. There is less graphene in the picture and the agglomeration is not obvious. The graphene can be evenly dispersed into the hydrogel. When the mass fraction of graphene was 0.5%, the phenomenon of graphene agglomeration presented in the microscopic diagram became obvious, and larger agglomeration (maximum diameter: 7.4 μm) was formed. The quantity and density increased, and the dispersion was not relatively uniform. When the mass fraction of graphene is 0.75%, the diameter of the agglomerated particles (2.7 μm) formed in the dispersion are similar to that (2.5 μm) of 0.25%. The agglomeration is not obvious, and the dispersion uniformity is relatively good. When the mass fraction of graphene is 1%, the diameter of agglomeration particles increases significantly (average diameter: 5.1 μm). The agglomeration of large particles formed from graphene in the dispersion will increase the acoustic absorption of graphene, thus affecting the propagation efficiency of ultrasonic wave and reducing the energy propagated into specimens. However, when the graphene content is relatively low, the acoustic impedance of the composite coupling agent rarely changes, so the propagation efficiency of ultrasonic wave cannot be optimized well. In terms of graphene dispersion, when the mass fraction of graphene is 0.75%, the propagation of sound waves is most favorable.

Table 1. Graphene agglomerated particle diameter in micrograph of a hydrogel–graphene coupling agent.

Mass Fraction of Graphene	0.25%	0.5%	0.75%	1%
Maximum particle diameter (μm)	5.7	7.4	7.6	11.2
Minimum particle diameter (μm)	1.0	1.0	1.0	1.9
Average particle diameter (μm)	2.5	3.2	2.7	5.1

Figure 3. Micrographs of the hydrogel–graphene coupling agent. (a) graphene mass fraction is 0.25%, (b) graphene mass fraction is 0.5%, (c) graphene mass fraction is 0.75%, (d) graphene mass fraction is 1%.

4.2. Performance Evaluation Indexes of the Hydrogel–Graphene Composite Coupling Agent

The hydrogel–graphene composite coupling agent was used to perform a corresponding experiment; the results are shown in Figure 4a–c. Peak-to-peak values, attenuation coefficient, and energy value of the received signal were calculated. With the increase in mass fraction of graphene in the coupling agent, the variation trend of the three indexes was found to be similar. Figure 4d shows acoustic impedance and ultrasonic wave velocity with the increase of graphene mass fraction in the composite coupling agent.

Figure 4. (a) Peak-to-peak values, (b) Attenuation coefficient, (c) Energy value of the received signal with hydrogel–graphene composite coupling agent. (d) Acoustic impedance values and ultrasonic wave velocity of the hydrogel–graphene composite coupling agent with the mass fraction of graphene.

In particular, Figure 4a shows that when the hydrogel–graphene composite coupling agent has mass fraction of 0.75%, the peak-to-peak values of the received signal is the highest. In this situation, a three-dimensional graphene structured powder disperses well. As can be seen from Figure 4b, within a certain range, with the mass fraction of

graphene increasing, the attenuation coefficient gradually decreases and the coupling effect of composite coupling agent on ultrasonic wave is positive. When the mass fraction of graphene is 0.75%, the attenuation coefficient is the lowest. However, the attenuation is greatest when the mass fraction of graphene is 1%. Figure 4c describes that when the mass fraction of graphene is 0.75%, the energy value of the gel-graphene composite coupling agent is the highest. As can be seen from the figure, of all the test frequencies, the compound coupling agent has the best effect when the mass fraction is 0.75%. As can be seen from Figure 4d, with the increase in mass fraction of graphene, the ultrasonic wave velocity of the composite coupling agent is basically unchanged, but its ultrasonic impedance value is gradually increased.

In other words, a small amount of graphene can significantly optimize the effect of the coupling agent, while excessive addition of three-dimensional graphene will increase the absorption of three-dimensional graphene to ultrasonic energy. As a dopant, graphene has special properties. When the composite material is prepared by combining with a polymer, it will exhibit a certain degree of absorption. Ying Wu et al. [22] reported the sound absorption properties of three-dimensional graphene-based composites. Due to sound absorption properties of graphene, the energy propagated into specimens will decrease. Furthermore, when the mass fraction of graphene is 0.75%, the energy value is much larger than that of other graphene contents' counterpart. In this situation, the acoustic impedance of the hydrogel-graphene composite coupling agent is more compatible with the specimen, thus the energy value of ultrasonic echo is effectively improved.

4.3. Microscopic Image of the Glycerol-Graphene Composite Coupling Agent

As is shown in Figure 5 and Table 2, a few large agglomeration particles appear when the mass fraction is 0.25%. In Figure 5b, the maximum size (6.9 μm) of the aggregate particles decreases but the number of particles is very large. During the propagation of ultrasonic waves, the interface of different media causes their attenuation. In Figure 5c, graphene is uniformly dispersed, and the average size (2.7 μm) of the agglomerated particles is moderate. Figure 5d shows that the aggregate particles (maximum: 11.2 μm) of graphene in glycerol are so large that a stable dispersion is hardly formed. This results in a great deal of ultrasonic attenuation. Excessive graphene cannot form uniform dispersion solution in glycerol, which results in inability of the composite graphene coupling agent prepared with a mass fraction of 1% to effectively promote ultrasonic propagation.

Figure 5. Micrograph of the glycerol-graphene coupling agent. (a) graphene mass fraction is 0.25%, (b) graphene mass fraction is 0.5%, (c) graphene mass fraction is 0.75%, (d) graphene mass fraction is 1%.

Table 2. Graphene-agglomerated particle diameter in micrograph of a glycerol–graphene coupling agent.

Mass Fraction of Graphene	0.25%	0.5%	0.75%	1%
Maximum particle diameter (μm)	6.9	6.5	6.4	11.2
Minimum particle diameter (μm)	1.0	1.0	1.0	1.0
Average particle diameter (μm)	3.1	3.2	2.7	5.1

4.4. Performance Evaluation Indexes of the Glycerol–Graphene Composite Coupling Agent

The glycerol–graphene composite coupling agent was used to perform a corresponding experiment; the results are shown in Figure 6a–c. Peak-to-peak values, attenuation coefficient, and energy value of the received signal was calculated. It can be seen that the change of the three indexes of the glycerol–graphene composite coupling agent is similar that of the hydrogel–graphene composite coupling agent. Figure 6d shows acoustic impedance values and ultrasonic wave velocity with the increase of graphene mass fraction in the glycerol–graphene composite coupling agent.

Figure 6. (a) Peak-to-peak values, (b) Attenuation coefficient, (c) Energy value of the received signal with the glycerol–graphene composite coupling agent. (d) Acoustic impedance values and ultrasonic wave velocity of the glycerol–graphene composite coupling agent with the mass fraction of graphene.

Figure 6a shows that when the mass fraction of graphene in the glycerol–graphene coupling agent is 0.75%, the peak-to-peak value is the largest. When the mass fraction of graphene in the glycerol–graphene coupling agent is 1%, the peak-to-peak value is worse than that without graphene. As is shown in Figure 6b, when the mass fraction of graphene is 0.75%, the attenuation coefficient of the glycerol–graphene composite coupling agent is minimum. As is shown in Figure 6c, when the mass fraction of graphene is 0.75%, energy value of the glycerol–graphene composite coupling agent is the best. The glycerol–graphene composite coupling agent performed best at a mass fraction of 0.75% of graphene at all test frequencies. It can be seen from Figure 6d that the ultrasonic velocity values changed little with the increase of graphene mass fraction while the impedance value gradually increased after the addition of graphene.

The experimental results indicated that the appropriate amount of graphene added to glycerin has a positive coupling effect on the glycerin coupling agent, and the best effect is when the mass fraction of graphene is 0.75%. Firstly, the viscosity of pure glycerol coupling agent is not high, thus flowing freely. In the coupling process, it is easy to cause excessive loss due to the coupling force, and the coupling effect cannot be achieved, and even excessive friction between the sensor and the tested specimens may occur. Therefore,

appropriately increasing the viscosity of glycerol coupling agent can optimize ultrasonic detection and protect the tested specimens. It has been pointed out that as the mass fraction of graphene increases, the viscosity of graphene–glycerol dispersion increases [7]. With the increase in viscosity, the mobility of the graphene–glycerol composite coupling agent decreases accordingly, which reduces the micro-gap between the filling of joint surfaces of the coupling agent. It is not conducive to the propagation of ultrasonic waves. Therefore, when the mass fraction is 0.75%, three indexes are the maximum. Secondly, the addition of graphene changes the acoustic impedance of the composite coupling agent. The matching of acoustic impedance is enhanced, which reduces the energy attenuation of ultrasonic wave in the workpiece.

5. Discussion

In conclusion, when the mass fraction of graphene was 0.75%, the peak-to-peak value, attenuation coefficient, and energy value of the glycerin–graphene composite coupling agent and gel–graphene composite coupling agent were the best. Here are some explanations for the aforementioned plots. Firstly, in the experiment, the exciting frequency is 5 MHz, but the peak of the curve appears at 5.1 MHz. According to the relevant data, the manufacturing and packaging technology of the sensor will lead to deviation of the exciting frequency. Secondly, the three index parameters in this study showed a trend of increasing firstly and then decreasing with the increase in mass fraction of graphene. Secondly, when the mass fraction is constant, the attenuation coefficients of the two couplings do not change much with the increase in frequency. The reason is that the ratio of primary and secondary echo ultrasonic energy is small as the frequency changes. When calculated according to Equation (5), there is little difference in the attenuation coefficient.

6. Conclusions

1. Graphene was added to a hydrogel coupling agent and a glycerin coupling agent to prepare a hydrogel–graphene composite coupling agent and a glycerin–graphene composite coupling agent, which can change the acoustic impedance value, so that the coupling agent can better realize the acoustic impedance match between the sensor and the tested specimens, and improve the propagation performance of the ultrasonic wave.
2. Adding an appropriate amount of graphene to the coupling agent has a positive effect on the coupling effect and the acoustic propagation performance of the composite coupling agent will be improved. The properties of the composite coupling agents are arranged according to the following mass fraction: 0.75%, 0.5%, and 0.25%, graphene free and 1%. Graphene can effectively change the acoustic impedance of polymer hydrogels and glycerol, while the viscosity of glycerol effectively increases, so that the parameters of both composite coupling agents have good improvement. However, when the mass fraction of graphene is greater than 0.75%, the sound absorption effect of graphene itself will gradually become obvious, leading to the decline of three parameters.
3. At different test frequencies, when the mass fraction of graphene added in hydrogels coupling agent and glycerin ultrasonic coupling agent was 0.75%, the peak-to-peak value, attenuation coefficient, and energy value of ultrasonic echo showed the best effect, and the effect of composite coupling agent would not change with the change in signal frequency.

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