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Impacts investigation of gas barrier on efective thermal conductivity and service life of vacuum insulation panel

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Vacuum Insulation Panels (VIPs) are highly efficient thermal insulation materials with extremely low **thermal conductivity based on the vacuum principle. With the sealing properties of the gas barrier envelopes, a long service life of the VIP is obtained. The mechanism and infuence factors of gas and water vapor permeability were mathematically analyzed to explore the infuence of gas barrier envelopes on the thermal performance of VIPs. Three typical gas barriers were studied, and the selection of the gas barrier and other aspects of optimization were involved. The relationships among** temperature, humidity, solubility coefficient, diffusion coefficient, and permeability were concluded, **which shows that temperature has a much greater efect on the permeability of the gas barrier relative to humidity. The numerical analysis and infuencing factors of VIPs' service life were also exemplifed with three diferent types of gas barrier envelopes. The experimental results show that depending on the environment, the temperature has a major impact on the efective thermal conductivity and service life of VIP. The research was signifcant in the selection of gas barriers, the optimization of the performance, and the development of vacuum insulation material.**

List of symbols

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Greek symbols

Abbreviations

In recent years, the issue of global warming has garnered signifcant attention and has emerged as a topic of considerable interest and concern within the global scientific community and wider society. The likelihood of experiencing extreme weather events has been steadily increasing, with summers becoming hotter and winters growing colder. In response, there has been a corresponding surge in energy consumption, particularly for cooling and heating systems, to maintain acomfortable living environment. This has resulted in a significant rise in carbon emissions, thereby exacerbating the greenhouse efect and posing formidable challenges for the future development of human society. Ever since China's successful implementation of its carbon peak and carbon neutrality strategy, the nation has witnessed a remarkable surge in the popularity and growth of its low-carbon economy. The development of green energy and energy-saving materials has been accorded the utmost priority, as the country continues its steadfast commitment towards achieving a more sustainable and eco-friendly future¹. Figure [1](#page-2-0) illustrates the extensive applications of Vacuum Insulated Panels (VIPs) in areas such as building energy conservation, cold chain logistics, fne chemicals, household appliances, etc. VIPs have emerged as a highly efficient and promising energy-saving material, with vast prospects for further development.

Figure [2](#page-2-1) shows the schematic of VIP, which is composed of three parts: gas barrier, porous core material, and getter. The core material is input into the gas barrier envelopes and then vacuum sealed to obtain a high vacuum internal structure, the thermal conductivity of VIP can be as low as 2mW/(m**·**K), which is the lowest of thermal insulation materials^{2-[5](#page-9-2)}. With energy conservation and environmental protection advantages, VIP is increasingly widely used.

The thermal conductivity of the core material determines the thermal insulation performance of the VIP to a certain extent. Porous materials such as fumed silica, aerogel, glass fber, and microporous polyurethane are generally selected as core materials. Taking the ultra-fne glass fber core material as an example, on the one hand, the fber staggered lap leads to a signifcant increase in the heat conduction path, so the heat conduction coefficient is effectively reduced. On the other hand, the pore size formed by the micron super-fine glass fiber lap is small, and the size of a nanometer can be formed afer vacuum suction, which can efectively eliminate the convective heat transfer, to obtain a low thermal conductivity^{[7,](#page-9-3) [8](#page-9-4)}.

The unique vacuum structure inside the VIP is the key to its low thermal conductivity. To ensure the thermal insulation performance of the VIP over a long time, it is very important to maintain the stability of the vacuum degree in the VIP. As shown in Fig. [3](#page-3-0), the sensitivity of diferent core materials to pressure is diferent, resulting in great differences in their thermal insulation performance under different vacuum degrees 10^{-12} 10^{-12} 10^{-12} .

As the packaging material of the VIP, the gas barrier has a certain degree of gas resistance and water vapor resistance but is not airtight. Gas can penetrate the VIP through the surface of the gas barrier, or dissolve into the gas barrier through adsorption-difusion, which will greatly afect the vacuum degree of the VIP, and then affect the long-term stability of its thermal insulation performance¹³. Zhou¹⁴ designed a vacuum insulation array (VIA) with independent vacuum units, which can achieve local damage without afecting the thermal insulation properties of the overall structure, so it can be pierced and cut or reassembled, which is a breakthrough in the feld of VIP research, but still cannot avoid fatigue damage of gas barriers under long-term use conditions. Therefore, to improve the thermal performance of VIPs, it is necessary to study and optimize the performance of the gas barrier.

There is much theoretical research on gas barriers. Hubert Schwab and Ulrich Heinemann^{[15](#page-10-3)} studied the permeability of oxygen and water vapor of AF, MF1, and MF2 gas barriers under diferent VIP specifcations and humidity. The results showed that if the boundary of the VIP is increased by 2 times, the pressure growth rate in the VIP would be reduced by half. Water vapor permeability is closely related to the core material's water absorption characteristics and the environment's relative humidity. S. Brunner¹⁶ adopted Fib-Etching technology to study the influence of different aluminum thicknesses on reducing gas and water vapor permeability. Wang¹ discussed a new type of low thermal conductivity and high-temperature vacuum insulation composite composed

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Figure 1. Application felds of VIPs.

Figure 2. Schematic of VIP⁶.

of a C*^f* /SiC sealing shell, which has great application potential as a thermal protection system material. Hubert and Similler^{18, [19](#page-10-7)} also did a lot of related research on the influence of gas penetration of barrier membranes on the service life of VIPs and put forward the gas permeability performance index of VIPs in diferent areas within the required service life range. Zhang²⁰ studied in detail the performance of gas barriers and the influence of boundary thermal efect on the performance of VIPs and successfully developed a nano gas barrier that replaced aluminum foil. Li^{[21](#page-10-9)} systematically analyzed the factors affecting the performance of gas barriers, discussed the infuence of the thermal bridge efect, gas penetration, and the size on the performance of VIPs discussed the ways and requirements to reduce the thermal bridge efect and gas penetration, and summarized the methods to improve the performance of the gas barrier. In their research, Oliver Miesbauer^{[22](#page-10-10)} proposed a new idea of opaque high-barrier films. The barrier and mechanical properties of these films and their applicability in construction are discussed. Li[23](#page-10-11) used three flms (AF, MF, and GF) and VIPs (thermal insulation core material-precipitated silica) using these three films for the degradation study. The effects of temperature, alkali, and local stress concentration on the properties of gas barriers were discussed. Glenn De Meersman²⁴ studied the experimental process of VIP degradation under diferent humid and hot conditions at room temperature and controlled climate chambers

and determined the Sd value of aluminum cladding based on these measurements. Martin^{[25](#page-10-13)}, explained the reasons for the diferent thermal resistance values of refective multi-foil insulations under diferent test methods and introduced the standard calculation procedure for calculating the thermal resistance of refective multi-foil insulations. The studies mentioned above are mostly the adjustment of different macro parameters and their impact on the service life of VIPs, such as VIP size, gas barrier thickness, gas barrier type, etc., but the research on the mechanism of the gas barrier permeation is minimal. To ensure the long-term stability of VIPs' thermal insulation performance, improving the performance of the gas barrier in principle is an important issue that needs to be studied urgently.

In this paper, the principle and infuencing factors of the permeability of the gas barrier are analyzed, and the mathematical model is established to study the infuence results in theory, which is of great signifcance to optimizing the gas barrier's performance and extending the service life of VIPs.

The permeability of the gas barrier

Penetration mechanism

The gas barrier is composed of polymer materials with gas and water vapor resistance but is not airtight. As shown in Fig. [4,](#page-3-1) gaseous molecules such as oxygen, nitrogen, and water vapor present in the service environment can permeate through the gas barrier and infltrate the interior of VIPs, leading to a disruption in the vacuum integrity of VIPs and consequently exerting a detrimental impact on thermal insulation performance. Most scholars believe that the penetration process of gas to the gas barrier is a single-molecule difusion process, which belongs to the mass transfer process²⁶. When the gas penetrates the gas barrier, it goes through three continuous processes: (1) the dissolution of the gas on the surface of the gas barrier, (2) the difusion of the gas from high concentration to low concentration in the gas barrier, (3) the solution of the gas from the surface on the other side of the gas barrier. There is no chemical reaction between the gas barrier and the gas, and the internal and external environment of the gas barrier will be in dynamic equilibrium when the permeation process is completed 2^7 .

The permeability coefficient, a measure of a gas barrier's ability to resist gas penetration, is determined by the dissolution-diffusion mechanism. According to this mechanism, the gas permeability coefficient can be expressed

Figure 4. Schematic diagram of the gas barrier permeation process.

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as the product of the gas solubility coefficient, which reflects the ability of a gas to dissolve in the barrier material, and the gas diffusion coefficient, which reflects the ability of a gas to diffuse through the barrier material²⁸:

$$
P = S \times D \tag{1}
$$

where *P* is the permeability coefficient, *S* is the solubility coefficient, and *D* is the diffusion coefficient. It can be seen from Eq. [\(1](#page-4-0)) that the permeability coefficient of the gas is related to the solubility coefficient *S* and diffusion coefficient *D*, so the permeability of the gas to the gas barrier depends on its diffusion ability and solubility ability. In a broad sense, the permeability coefficient of the gas barrier can be expressed by Eq. (2) (2) (2) :

$$
P = F_i \times G_i \times A_k \tag{2}
$$

where F_i is the influence coefficient of the gas barrier properties on permeability, G_i is the influence coefficient of gas characteristics on permeability, and A_k is the influence of environmental and other factors on permeability. It can be seen from Eq. ([2\)](#page-4-1) that the permeability coefficient of the gas barrier is affected by many factors, F_i and G_i are the essential parameters of the gas barrier and the permeable gas, and their values depend on the material of the gas barrier and the type of gas, which is not discussed here, the infuence of environmental factors on the permeability of the gas barrier A_k is only considered in this work. Based on the permeability mechanism, the relationship between temperature and humidity and the solubility coefficient S and diffusion coefficient D of the gas barrier is studied by numerical analysis, so that the infuence of temperature and humidity on the permeability of the gas barrier can be efectively obtained.

Efect of temperature on the permeability

The solubility *S* is a parameter indicating the solubility of the gas, which is related to the gas properties, the gas barrier structure, the ambient temperature and humidity, and the gas pressure. The diffusion coefficient *D* is a parameter indicating the difusion capacity of the gas, which is related to the type of gas and the ambient temperature and pressure. According to Arrhenius law²⁹, the solubility coefficient S and diffusion coefficient *D* of the barrier membrane as a function of temperature can be expressed by Eqs. [\(3\)](#page-4-2) and ([4\)](#page-4-3):

$$
S = S_0 exp(-\frac{E_s}{RT})
$$
\n(3)

$$
D = D_0 \exp(-\frac{E_D}{RT})
$$
\n⁽⁴⁾

where S_0 is the limit of the *S* of the gas barrier when the temperature goes to 0, D_0 is the limit of the *D* of the gas barrier when the temperature goes to $0, E_s$ and E_D are solution activation energy and diffusion activation energy respectively, *R* is the gas constant, and *T* is the absolute temperature.

The law is also followed for the permeability coefficient *P*:

$$
P = P_0 \exp(-\frac{E_P}{RT})
$$
\n⁽⁵⁾

where P_0 is the limit of the *P* of the gas barrier when *T* goes to 0, E_P is the permeation activation energy. According to Eq. [\(1](#page-4-0)):

$$
P_0 = S_0 \times D_0 \tag{6}
$$

$$
E_P = E_S + E_D \tag{7}
$$

$$
P = S_0 D_0 exp(-\frac{E_S + E_D}{RT})
$$
\n(8)

In general, the diffusion coefficient *D* increases with the increase in temperature, and the solution coefficient *S* of the gas varies with the characteristics of the permeate gas. The E_s is positive for invariant gases (H_2 , N_2 , O_2 , etc.), and the *S* increases with increasing temperature. For condensable gas (water vapor), the *E_S* is negative, and the *S* decreases with the increase in temperature.

Table [1](#page-5-0) lists the water vapor permeability of PET12μm, PET20μm, and aluminized PET18μm at diferent temperatures tested by the Gas Transmission Rate Tester, which verifes the correctness of the theoretical analysis. From another point of view, the difusion of gas molecules is actually with the help of the free volume or hole in the gas barrier density fuctuation appears as a channel, the gas barrier is generally composed of polymer, with the increase of temperature, the more the polymer expands, the more the polymer chain moves, resulting in the more opportunities for the appearance of such holes, and therefore the greater the permeability of the gas barrier.

Efect of humidity on the permeability

When the humidity of the environment rises, the moisture in the environment will difuse into the gas barrier, and the penetration of this moisture is equivalent to the addition of additives to the material, which plasticizes the polymer^{[30](#page-10-18), 31}. This plasticizing effect increases the number of holes in the film and makes the diffusion of the gas easier and faster. The effects of humidity and plasticization on gas barrier permeability can be expressed by Eq. ([9\)](#page-5-1):

Table 1. Water vapor permeability of three gas barrier materials at various temperatures. (Derived from^{[22](#page-10-10)}).

$$
e^{\eta \times X_w} = e^{\eta \times S \times P_{SAT} \times \overline{RH}} \tag{9}
$$

where η is the plasticity coefficient, X_w is the water vapor content, P_{SAT} is the saturation pressure, and \overline{RH} is the average relative humidity on both sides of the gas barrier. The effect of humidity on the permeability coefficient is different for different materials, when the humidity increases, the permeability coefficient of most materials will increase, and which of individual materials will decrease.

There are also some gas barriers whose permeability coefficients change slightly, as shown in Table [2](#page-5-2), mainly because the composition of these gas barriers is strong active molecules, the transmittance of the gas is almost not afected by the humidity, for the VIP, PE, and PET as the main component of its gas barrier is typical strong active molecules, so humidity has little effect on the permeability coefficient, which is far less than the influence of temperature. Therefore, the influence of temperature on the permeability of the gas barrier is mainly considered in the next work.

The service life ofVIP Theoretical model

Figure [5](#page-5-3) shows the mechanism of the permeation of water vapor and gas during the application of the VIP. The permeability of gas and water vapor through the gas barrier will lead to the increase of the VIP thermal conductivity, when the thermal conductivity exceeds a critical value, the VIP will be considered as failed. The American standard ASTM C 1484-01 stipulates that this value is 11.5 mW/(m·K), and the service life of the VIP is the corresponding use time in the event of failure.

Table 2. Water vapor permeability of three gas barrier materials at various humidity. (Derived from²²).

The thermal conductivity of the VIP is mainly affected by temperature, humidity, and pressure 32 , to predict the service life of the VIP, it is generally considered that the infuence of gas permeation and water vapor permeation on the thermal conductivity of the VIP is independent, which can be obtained by Eq. $(10)^{18}$ $(10)^{18}$ $(10)^{18}$ $(10)^{18}$ $(10)^{18}$

$$
\lambda_{ETC} = \lambda_{evac} + \lambda_{gas}(P_{gas}) + \lambda_w(X_w)
$$
\n(10)

where λ_{ETC} is the effective thermal conductivity of the VIP after use, λ_{evac} is the initial thermal conductivity of the VIP, $\lambda_{gas}(P_{gas})$ is the added value of the VIP thermal conductivity due to gas permeation, and $\lambda_w(X_w)$ is the added value of the VIP thermal conductivity due to water vapor permeation.

 $\lambda_{gas}(P_{gas})$ can be expressed by Eq. ([11](#page-6-1))³³:

$$
\lambda_{gas}(P_{gas}) = \frac{\lambda_{g0}}{(1 + P_{1/2}/P_{gas})}
$$
\n(11)

where λ_{g0} is the thermal conductivity of free static air, which is 26mW/(m·K), $P_{1/2}$ is the corresponding pressure when the thermal conductivity of the gas becomes $\lambda_{g0}/2$, which mainly depends on the pore size of the core material and the type of the gas, P_{gas} is the internal pressure of the VIP.

 $\lambda_w(X_w)$ can be expressed by Eq. $(12)^{34}$ $(12)^{34}$ $(12)^{34}$ $(12)^{34}$ $(12)^{34}$:

$$
\lambda_w(X_w) = b \times X_w \tag{12}
$$

where *b* is the tested coefficient related to the properties of the core material, X_w is the humidity of the VIP. Equation [\(13\)](#page-6-3) and Eq. ([14](#page-6-4))^{[35](#page-10-23)} conclude the relationship between P_{gas} and X_w over time:

$$
P_{gas}(t) = \frac{GTR_{\text{tot}}}{V_{\text{eff}}}(\frac{T_m P_0}{T_0})t
$$
\n(13)

$$
X_w(t) = f \times k_{out} \times \left(1 - exp\left(\frac{WVTR_{tot}}{m_{dry} \times f} \times t\right)\right)
$$
 (14)

where GTR_{tot} is the total gas permeability, V_{eff} is the effective pore volume of the VIP core material, T_m is the test temperature, T_0 is the standard temperature, P_0 is the standard atmospheric pressure, f is a constant related to the characteristics of the core material, k_{out} is the humidity of the environment, $WVTR_{tot}$ is the total water vapor permeability, t is the time.

Equation [\(10\)](#page-6-0) can be further deduced as:

$$
\lambda_{ETC} = \lambda_{evac} + \frac{\lambda_{g0}}{(1 + P_{1/2}/(\frac{GTR_{tot}}{V_{eff}}(\frac{T_m P_0}{T_0})t)} + b \times f \times k_{out} \times \left(1 - exp\left(\frac{WVTR_{tot}}{m_{dry} \times f} \times t\right)\right)
$$
(15)

The service life of the VIP can be predicted by Eq. (15) . It also shows that in addition to core material characteristics and other factors, the permeability of the gas barrier also affects the service life of the VIP. Therefore, the infuence of temperature on the service life of the VIP will be studied in the following work to verify the accuracy of the numerical analysis of the permeability of the gas barrier, which will have important guiding signifcance for the choice of the gas barrier and the application of the VIP.

Experimental methods

Tree kinds of VIPs with diferent gas barrier envelopes were prepared in this work, with glass fber as the core material, the size of each VIP is 300 mm \times 300 mm \times 10 mm. The structure of the three gas barriers is shown in Fig. [6,](#page-6-6) and the permeation parameters are shown in Table [3.](#page-7-0)

Figure [7](#page-7-1) is the schematic diagram of the test device for the relationship between VIP internal pressure and efective thermal conductivity. First, a vacuum pump is used to connect the gas inlet and outlet, afer holding the pressure at 0.1 Pa for 10 min, the vacuum valve is closed immediately, and the initial thermal conductivity of VIPs can be measured when the vacuum gauge is stabilized. Aferward, by controlling the opening and closing of the vacuum valve, the efective thermal conductivity of VIPs under diferent internal pressures can be measured, providing a reference for predicting the service life of VIPs.

Figure 6. Composition of AF(Al-foil), MF(Al-Metallized flm), and CF(Composite flm) gas barriers.

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Table 3. The permeation parameters of the gas barriers.

Figure 7. Diagrammatic representation of the experimental setup for exploring the correlation between the internal pressure of vacuum insulation panels (VIPs) and their efective thermal conductivity.

As shown in Fig. [8](#page-7-2), VIP samples named AF-VIP, MF-VIP, and CF-VIP, were placed in high-low pressure/ high-low temperature test chamber (20 °C, 70 °C, 120 °C, and 140 °C, 90% RH), taken out, and measured the effective thermal conductivity of VIPs every 7 days, with a period of 4 weeks. The effect of temperature on VIP performance can be derived by observing the change in the efective thermal conductivity of VIPs.

Experimental results

According to the VIP service life theory model, combined with the VIP internal pressure and efective thermal conductivity test device, the service life of the three VIPs is predicted, VIPs' service environment is assumed at standard atmospheric pressure, 20 °C, and 50% RH. As shown in Fig. [9](#page-8-0), the green line is the thermal conductivity when the VIP fails. It can be observed that AF-VIP has the longest service life of more than 100 years, MF-VIP is second, and CF-VIP is the shortest, only about 70 years.

The difference between them has a crucial relationship with the composition of the gas barrier. Figure [6](#page-6-6) has given the composition structure of three types of gas barriers, of which the aluminum foil layer in the AF gas barrier is the thickest, and has the best barrier performance, but the over-thick aluminum foil layer will lead to the thermal bridge efect, which is also the reason why the initial thermal conductivity of AF-VIP is higher than that of the other two VIPs. The MF gas barrier with a multi-layer aluminum coated film can significantly reduce the efect of the thermal bridge efect but leads to a decrease in barrier performance. As a non-metalized flm, CF barrier flm has the lowest thermal bridge efect, and CF-VIP has the lowest thermal conductivity, but its service life is poor. Tis is because the core barrier layer in the CF gas barrier is the MEVOH layer. MEVOH has a high barrier to non-polar molecules such as oxygen, but is particularly sensitive to water molecules and thus exhibits high permeability, increasing VIPs' thermal conductivity and decreasing service life.

To compare the accuracy of theoretical models, the efective thermal conductivity of three VIPs afer aging at a temperature of 70 °C and 90% RH was obtained through experimental tests and theoretical predictions.

Figure 8. High-low pressure/high-low temperature test chamber.

Figure [10](#page-8-1) illustrates a minor deviation between the theoretically calculated thermal conductivity and the actual efective thermal conductivity. However, this deviation falls well within the acceptable range, afrming the reliability and accuracy of the experimental results. The observed discrepancy between the theoretical model and experimental results may be attributed to the volatilization of water within the core material, which had not been accounted for in the theoretical calculations. The core material used in this work is glass fiber, which is prepared by a wet process and will inevitably have moisture residue. The rise in temperature leads to the volatilization of moisture, which emerges as the primary factor contributing to the observed deviation between the theoretical and experimental values.

It can be seen from Fig. [10](#page-8-1) that the efective thermal conductivity of the three VIPs shows an increasing trend with the increase in temperature, and with further increase in temperature, the efective thermal conductivity of VIPs varies more signifcantly over time, as shown in Fig. [11,](#page-9-8) especially for CF-VIP, which fts well with the theoretical model. The permeability of the gas barrier is the main reason for the change in the thermal conductivity of VIPs, so the effective thermal conductivity of CF-VIP changes the most. The high temperature will exacerbate this trend of change, because the pinhole defect in the gas barrier will expand outward under the infuence of high temperature, forming a difusion channel, accelerating the rapid difusion of gas and water vapor, resulting in increased permeability of the gas barrier and a decrease in air tightness³⁶, thereby increasing the effective thermal conductivity of VIPs, which will accelerate the failure rate of VIPs, thus afecting the service life of VIPs. Therefore, it's necessary to choose gas barriers with better performance to cope with complex environments in practical applications, which has important guiding signifcance for the application of VIPs.

Figure 10. Comparison of effective and theoretical thermal conductivity at 70 °C and 90% RH.

Figure 11. The effective thermal conductivity of three VIPs at 120 °C and 140 °C, 90% RH.

Conclusions

The relevant factors affecting VIP were systematically studied through numerical analysis and experimental methods, combined with molecular motion dynamics and the Arrhenius Law, leading to the following conclusions being reached:

- (1) Temperature has a greater impact on the permeability of the gas barrier, while humidity has a lesser impact on the permeability of the gas barrier.
- (2) With the increase in temperature, the efective thermal conductivity of the VIP shows a continuously rising trend. In addition, prolonged exposure to high temperatures accelerates VIP failure, which signifcantly reduces its service life.
- (3) The permeability of the gas barrier directly affects the stability of the thermal insulation performance of the VIP, and the selection of a good gas barrier helps to deal with the infuence mechanism of the complex environment on the permeability and extend the service life of the VIP.

Data availability

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Competing interests

The authors declare no competing interests.

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