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Effects of cold storage temperature on thermoreversible aging in heavily oxidized asphalt binders

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Abstract

In order to better understand the thermoreversible aging characteristics of asphalt after heavy oxidation, six selected heavily oxidized asphalt containing different proportions of $C_{20}H_{42}$ are subjected to extended bending beam rheometer (ExBBR) tests, while modulated differential scanning calorimetry (MDSC) and atomic force microscope (AFM) are also performed on the asphalt. The results indicate that the introduction of heavy oxidation will not only affect the low temperature properties of the asphalt but also its thermoreversible aging characteristics, but this effect will receive an influence of the storage temperature.

This is due to the increase in viscosity of the asphalt after heavy oxidation, which makes it difficult for $C_{20}H_{42}$ to migrate, while the increase in storage temperature can release the potential of the degree of thermoreversible aging of the asphalt by accelerating the movement of wax molecules. And like $C_{20}H_{42}$, the grade loss of asphalt blended with $C_{30}H_{62}$ is different at different storage temperatures and oxidation levels, which proves that for heavily oxidized asphalt, multiple storage temperatures should be selected for testing its grade loss. Combined with thermal and morphological analysis, the introduction of heavy oxidation not only increases the amount of wax precipitation, but also significantly increases the thermoreversible aging of the asphalt by lowering the T_g of the asphalt and forming more perfect crystals during low temperature storage. Finally, by comparing the grade loss due to cold storage and extended oxidation, it can be found that the incorporation of $C_{20}H_{42}$ will make the asphalt more influenced to physical hardening. Therefore, it is necessary to study the effect of asphalt wax content on the degree of thermoreversible aging of heavily oxidized asphalt.

Keywords: Asphalt binder; Thermoreversible aging; Crystalline wax; Oxidative aging; Physical hardening

1. Introduction

Petroleum asphalt is one of the most widely used materials in pavement construction owing to its good bonding and waterproofing properties. Because of its unique characteristics such as excellent waterproofing, high damping ratio, good integrity, asphalt has also been utilized in other applications such as water conservancy, railway engineering. Countries such as the United States, Germany, Japan, and China has carried out a large number of studies on asphalt concrete subgrade [1, 2]. Compared with other ordinary subgrade, the advantages of

asphalt concrete subgrade mainly include the following five aspects: (1) reducing the stress effect of upper loads on the subgrade and transferring them to the lower structure; (2) keeping the moisture content of the subgrade stable as an impermeable layer; (3) acting as a support layer for ballast and reducing ballast splash; (4) alleviating train vibration and reducing noise; (5) reducing the whole life cycle cost of the track structure due to the reduction of later maintenance [3, 4]. The authors' team has systematically studied the damage pattern of full-section railway asphalt concrete subgrade and found that when asphalt concrete is positioned at a lower level in the railway subgrade, the train loads tend to be smaller, thus determined that thermal shrinkage cracks become an important factor affecting the durability of asphalt concrete [5, 6]. Among the many influencing factors, thermoreversible aging could affect asphalt concrete subgrade performance in the long run [7-9]. One of the main reasons is that other forms of aging mainly occur on the pavement surface while thermoreversible aging occurs in the whole asphalt pavement structure, especially in cold areas.

In order to understand the mechanism of thermoreversible aging as an important basis for the production of high-quality asphalt and solving related pavement distresses, in recent years researchers have devoted themselves to explain the causes of physical hardening in asphalt. The term physical hardening was first introduced by Anderson from Shell Laboratories, Amsterdam, The Netherlands, who attributed the phenomenon to the precipitation of waxes and the aggregation of asphaltenes, noting that both processes are very slow at low temperature [10]. Some asphalt will form large microcrystalline wax crystals during long-term low-temperature storage and are affected by the change of pavement temperature. Therefore,

63 different asphalt could have different allowable wax contents, which is also the reason why
64 wax is considered to be the main factor in thermoreversible aging [11, 12]. In contrast, studies
65 have shown that waxes in asphalt are a mixture of alkanes, cycloalkanes and aromatic
66 compounds and dominated by n-alkanes with carbon numbers of C_{15}~C_{57} [13]. Kovinichi found
67 that although $C_{20}H_{42}$ can promote the physical hardening in asphalt, this tendency is affected
68 by the base asphalt binder and is not determined by the content of $C_{20}H_{42}$ solely [14].
69 Subsequent studies found that not all waxes will aggravate the thermoreversible aging of
70 asphalt. Compared with $C_{20}H_{42}$, $C_{30}H_{62}$ is significantly less effective in promoting the physical
71 hardening of asphalt, while squalanecan improve the low temperature performance of asphalt
72 without aggravating the degree of thermoreversible aging [15, 16].

73 Despite a number of studies have conducted a series of studies on the effects of waxes
74 with different numbers of carbon molecules on the thermoreversible aging behaviour of asphalt,
75 the low temperature performance and grade loss of asphalt in most of these studies are
76 measured after a PAV-20h test using the standard BBR procedure [15, 17, 18]. In recent years
77 some practitioners have questioned this conventional PAV aging method, as Simon's research
78 has shown that the PAV-20h test is only able to simulate the condition of asphalt pavements
79 after 5-6 years of use, which is clearly not suitable to reflect the field aging of the pavement
80 surface after a long term service [19]. In order to be able to simulate the long-term use of
81 asphalt pavement, Simon proposed to prolong the aging time of PAV test in another study to
82 prepare heavily oxidized asphalt, and found that the asphalt under a PAV-40h test can simulate
83 the condition of asphalt pavements after 10-15 years of use [20]. Other researchers have

84 explored the low-temperature performance distribution of different asphalts after extended
85 aging. Kilger's research shows that the effect of recycling oils, including bio-oil and REOB,
86 on the improvement of asphalt's low-temperature performance tends to be weakened after
87 heavy oxidation compared with asphalt without recycling oils [21]. Ma, on the other hand,
88 found that the degree of thermoreversible aging of recycled asphalt blended with three different
89 rejuvenators can only be distinguished after RTFO and PAV aging, and increases with the
90 extension of PAV aging time [22]. This indicates that like the low-temperature properties of
91 asphalt, the degrees of thermoreversible aging of different asphalts after severe oxidation will
92 also be different, and thus it is necessary to investigate the mechanism of the effect of waxes
93 on the physical hardening properties of heavily oxidized asphalt binders.

94 In view of this, the purpose of this paper is to investigate the interaction between
95 thermoreversible aging and irreversible aging based on extended bending beam rheometer
96 (ExBBR) tests, using six different asphalt samples blended with pure wax, and analyzing the
97 grade loss of heavily oxidized asphalt by introducing three different aging methods, RTFO,
98 PAV-20h, and PAV-40h test. Based on the results of ExBBR tests, modulated differential
99 scanning calorimetry (MDSC) and atomic force microscopy (AFM) were used to analyze the
100 mechanism of the effect of wax on the physical hardening properties of heavily oxidized asphalt.

2. Experimental program

2.1. Materials

2.1.1 Asphalt binder

According to research, the wax content of asphalt will affect the road performance of asphalt pavement. In this paper, six kinds of asphalt from different crude oils were selected, among which the base asphalt from Venezuela came from the Strategic Highway Research Program (SHRP) Material Reference Library (MRL), which is labeled as ABG, according to the research there is basically no wax in ABG [23]. While the other asphalt samples are used for comparison, which are labeled as SWE, CAL, MAO, UNK, and ZNH, whose source and performance grade (PG) are shown in Table 1. The first three kinds of asphalt binders are commonly used in other countries while the rest of the asphalt binders usually used in asphalt pavement construction in China. To compare the oxidative aging with thermoreversible aging, in this research, the asphalt binders are tested after going through the rolling thin film oven (RTFO) as well as 20h and 40h of pressure aging vessel (PAV) tests.

Table 1. Basic properties of asphalt binders.

Sample Code	Source	Limiting high temperature PG (°C)	Limiting low temperature PG (°C)
ABG	Venezuela	65.2	-29.7
SWE	Sweden	63.8	-31.3

CAL	Caltex, Korea	67.1	-26.5
MAO	Maoming, China	64.9	-28.6
UNK	Unknown, China	65.7	-29.5
ZHH	Zhonghai, China	66.3	-27.7

2.1.2 Additives

According to previous research on wax in asphalt [15], the waxes in asphalt are mainly composed of saturated straight-chain hydrocarbons. Since waxes are an important cause of thermoreversible ageing of asphalt. Therefore, two commercial n-alkanes produced by Sigma-Aldrich were chosen as pure wax additives in this study, namely n-eicosane ($C_{20}H_{42}$) and n-dodecane ($C_{32}H_{66}$), with a purity of 99%. Considering that most asphalts for paving have a wax content of less than 7% [24-26], the percent of wax additives were blended at 1%, 3%, 5% and 7% by mass of asphalt binder. To ensure that the wax can be uniformly distributed in the asphalt, the base asphalt binder is heated to 165°C during the mixing process and the prepared model asphalt is sheared for 1h using a shear mixer.

2.2. Test Methods

2.2.1 Extended bending beam rheometer (ExBBR)

Among different low-temperature evaluation methods developed for asphalt binder in recent years [27, 28], the most mature and widely used one is the bending beam rheometer (BBR) test to measure the low temperature performance grade (PG) of asphalt binders.

131 However, it has also been shown that asphalt pavement designed based on PG performance
132 suffered from severe low-temperature cracking from time to time [29], due to the increase in
133 stiffness of asphalt binders at low temperatures with increasing storage time [30, 31]. Based on
134 this, the ExBBR test was developed. Compared to the regular BBR test, the ExBBR test
135 requires testing the PG performance of the asphalt in six states (3 conditioning time×2 storage
136 temperature) separately. The grade loss obtained from the ExBBR test can be used to evaluate
137 the degree of thermoreversible aging of asphalt. According to the requirements of AASHTO
138 TP122-17, the grade loss of asphalt should be less than 6°C.

139 *2.2.2 Modulated differential scanning calorimetry (MDSC)*

140 To understand the thermoreversible aging process of asphalt binders throughout the
141 service temperature range, the MDSC test was chosen for the study to analyze the thermal
142 behavior of asphalt. In contrast to conventional thermal analysis tests, the MDSC test allows
143 the total heat flow signal to be divided into reversing and non-reversing heat flow signals,
144 which has been favored by many researchers [32, 33]. The procedure is as follows. First, the
145 sample is heated to 120°C and then left at this temperature for 10min so that to melt all the
146 crystal structures. Second, the specimen is cooled down to 80°C at a cooling rate of 20°C/min
147 to avoid overrate oxidation and saving time. Next, the sample is rapidly cooled to -90°C at
148 10°C/min and kept for 10min. Then, the asphalt sample is heated to 120°C at a heating rate of
149 600°C/h (10°C/min). Finally, the total heat flow, reversing heat flow, non-reversing heat flow
150 and the first derivative of reversing heat flow are employed as the four key parameters to
151 characterize the thermal behavior of asphalt binder.

2.2.3 Atomic force microscope (AFM)

As a visco-elastic-plastic material with significant temperature dependence, asphalt has been the subject of many studies on the formation and growth mechanisms of the 'bee-like' structure in the AFM microstructure parameters. It has been shown that the 'bee-like' structure can be explained based on the wax nucleation theory, leading to the conclusion that its morphological characteristics are related to the heating history, cooling rate and aging degree of the sample [34-36]. As this study is focused on the differences in the microscopic morphology of different asphalts after low-temperature storage, the asphalt is stored at low temperature for 72h, and the surface of the asphalt is tested in amplitude mode using an AFM probe. In order to ensure the repeatability of the observation results, three replicates were carried out for each asphalt sample.

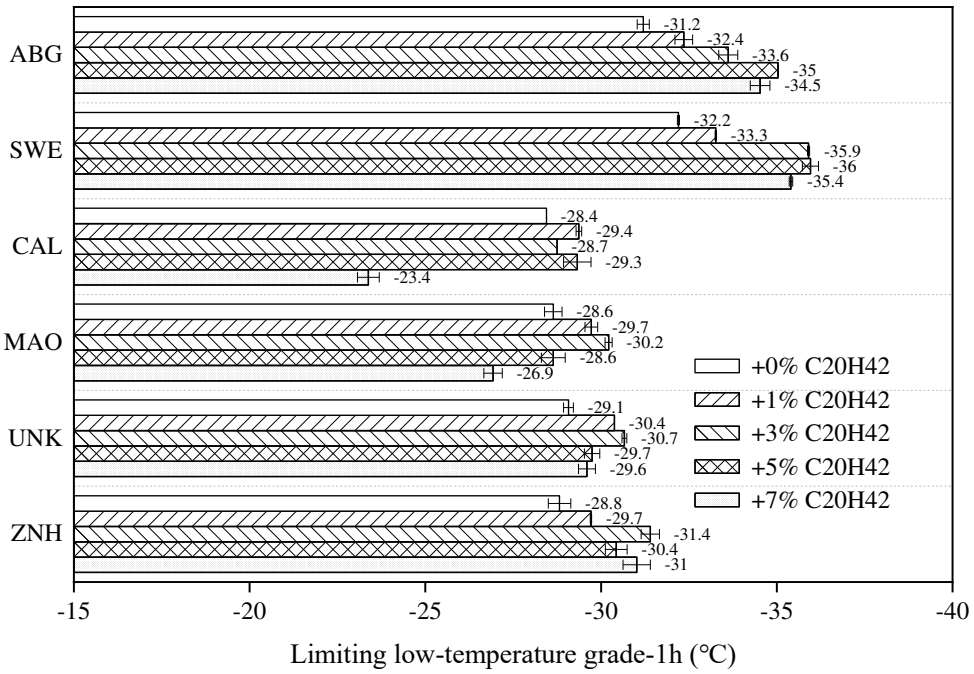
3. Results and discussion

3.1 Oxidation effects on limiting grade of asphalt binder

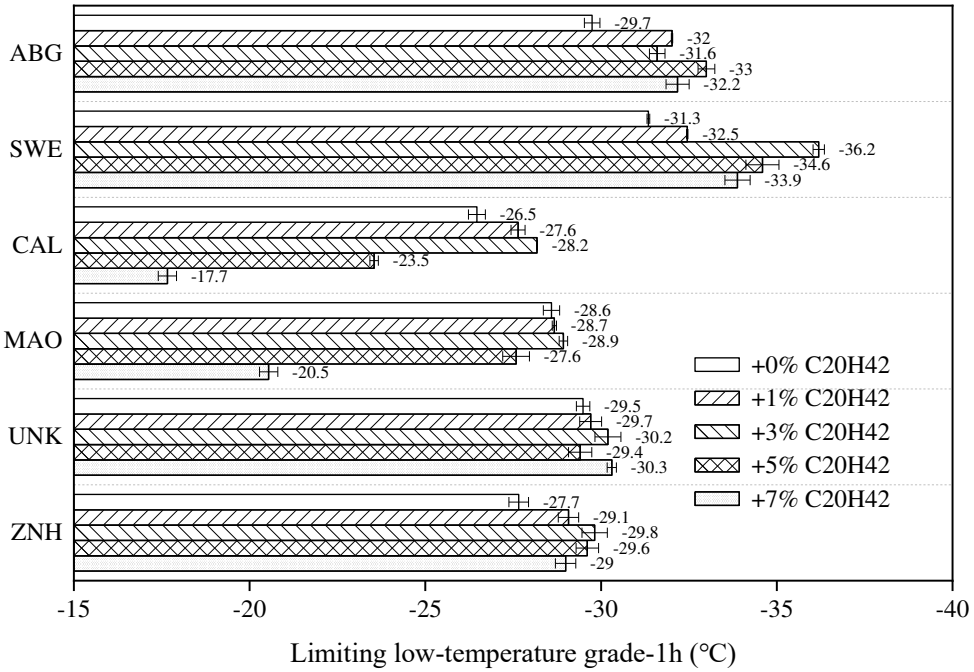
Fig. 1 shows the oxidation effects on limiting grade of asphalt binder based on ExBBR test. As can be seen, the addition of $C_{20}H_{42}$ to different RTFO aged asphalts effectively decreased the limiting grade of asphalt in the beginning. In fact, the addition of 5% of $C_{20}H_{42}$ to ABG and SWE exhibited the best improvement in terms of low-temperature performance (limiting grade was reduced by 4°C), followed by MAO, UNK and ZNH (limiting grade was reduced by 1°C~2°C) while CAL had the least improvement (limiting grade was reduced by less than 1°C). However, the limiting grade of most asphalts began to increase after adding 3%

of $C_{20}H_{42}$ while for ABG and SWE the turning point was at 5%. Combined with previous research results [37, 38], this may be due to the different wax contents of asphalts from different origins which asphalt with low wax content can accommodate more $C_{20}H_{42}$ before saturation.

By comparing the limiting grade of asphalt at different aging stages: 40h PAV>20h PAV>RTFO, for most asphalts compared to RTFO the limiting grade of asphalt after 40h PAV has increased by 1℃~2℃. Meanwhile, it is worth noting that the limiting grade of ABG and SWE after 40h PAV starts to increase with more than 3% of $C_{20}H_{42}$, which indicates that the introduction of heavy oxidation will not only lead to the deterioration of asphalt low temperature performance, but also lead to the decrease in the capacity for n-alkanes in asphalt.



(a) RTFO



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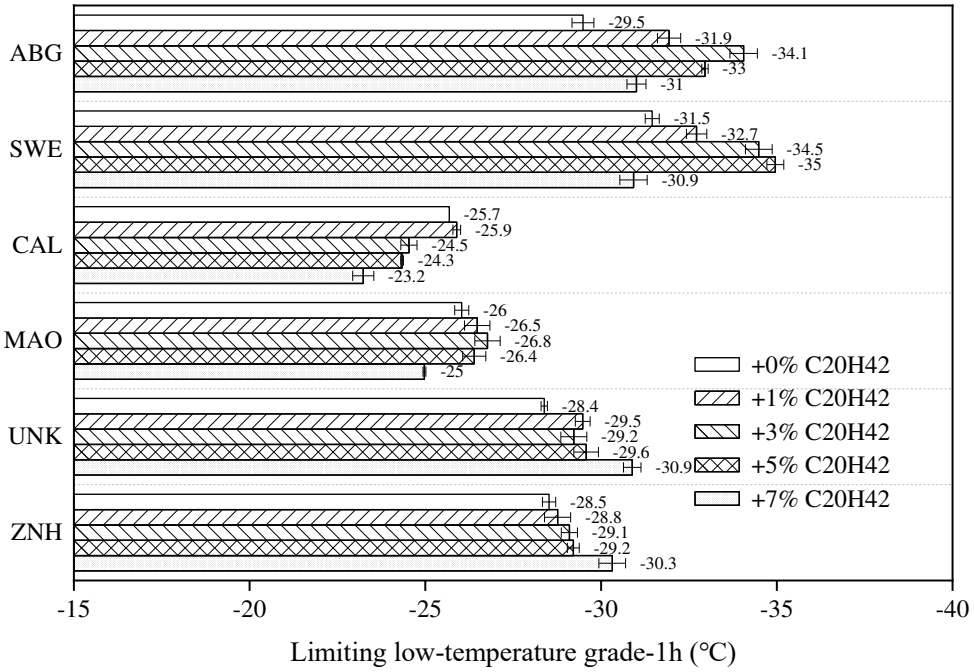
184 (b) 20h PAV



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186 (c) 40h PAV

187 **Fig. 1.** Oxidation effects on limiting grade of asphalt binder.

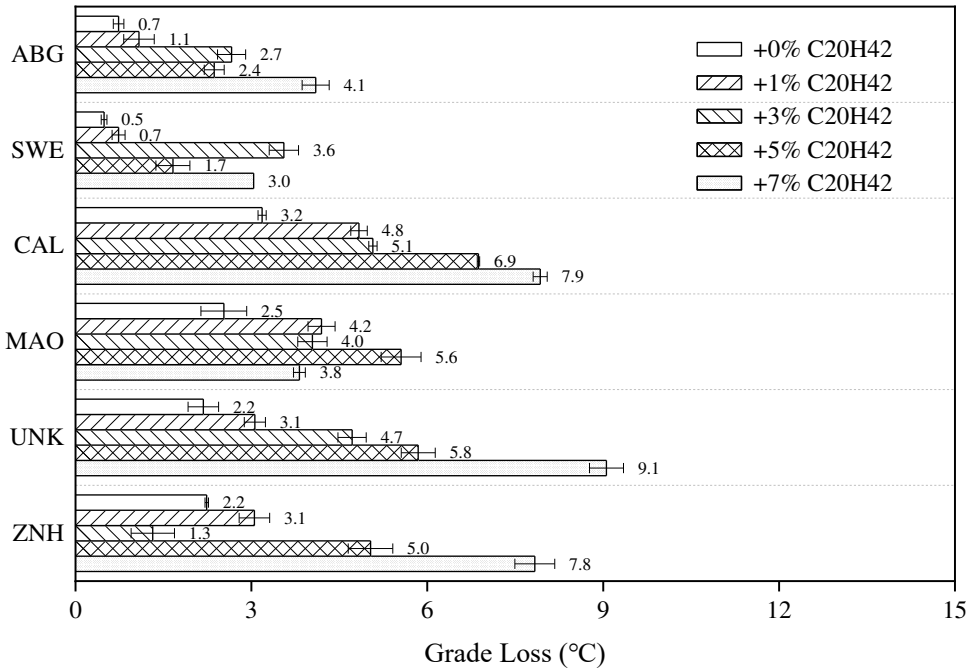
3.2 Oxidation effects on grade loss of asphalt binder

If only based on the limiting grade of the asphalt binder, it can be easily concluded that for most asphalt the addition of $C_{20}H_{42}$ is an effective solution to improve the low temperature performance of asphalt and that this improvement is almost independent of the effects of oxidative aging. However, some studies in recent years have shown that with the extension of the storage time of asphalt in low-temperature environment, the wax and asphaltene in the asphalt begin to slowly aggregate and exhibit physical hardening, which indirectly affects the low-temperature performance of the asphalt [25, 39].

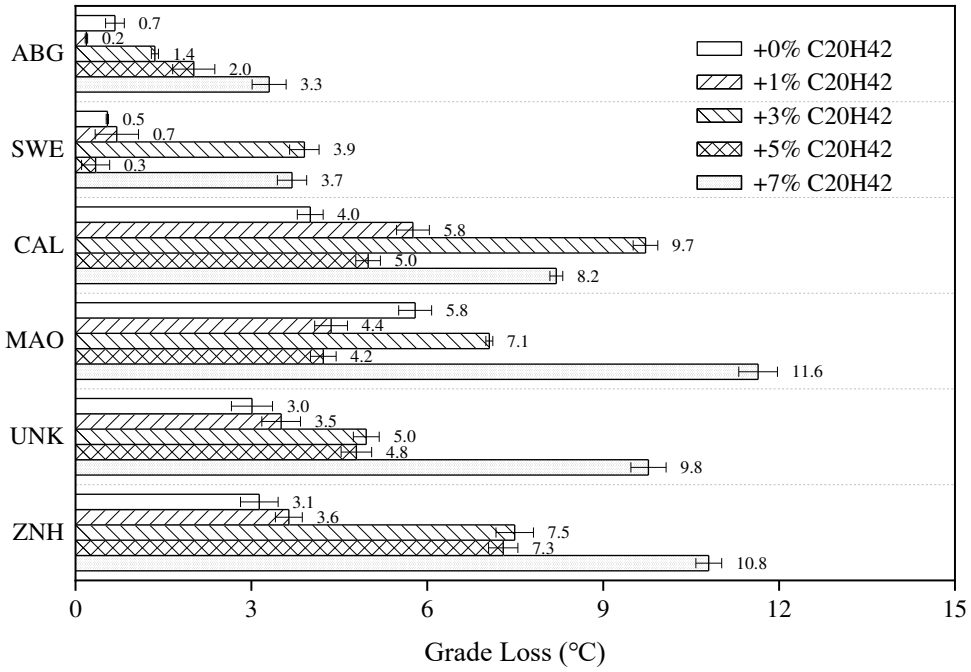
According to the result of grade loss of asphalt blended with $C_{20}H_{42}$ in low temperature environment given in Fig. 2, it can be observed that there is no significant difference in the grade loss of six asphalt without wax in RTFO aging stage, while the grade loss of six asphalt after blending with $C_{20}H_{42}$ can be easily distinguished. Among the six asphalt, CAL, MAO, UNK and ZNH blended with high content of $C_{20}H_{42}$ all showed significant hardening behavior with grade loss higher than 6°C. In contrast, the degree of thermoreversible aging of ABG and SWE, on the other hand, is not significantly affected by n-alkanes, and the maximum grade loss is only about 4°C. Referring to the research of Ding [23], this is due to the fact that ABG contains almost no wax and thus the asphalt can accommodate more wax before saturation.

Compared to the RTFO aging stage, the grade loss of most asphalt has increased after PAV. However, for ABG and SWE blended with C20H42, the grade loss of asphalt after 20h PAV remained almost unchanged. In contrast, after the introduction of 40h PAV, the grade loss

of the above two asphalt after heavy oxidation has increased to around 6°C. This can be explained using the findings of Tabatabaee [40, 41], that is, the presence of waxes will promote the phase separation of asphaltene components in asphalt. As the degree of oxidation increases, the increase of asphaltene content in asphalt will accelerate the rate of phase separation, resulting in a rapid increase in the hardening tendency of asphalt. However, this theory is not sufficient to explain the effect of the thermo-oxidative aging on the degree of thermoreversible aging of the other four types of asphalt. Taking MAO+3% $C_{20}H_{42}$ as an example, the grade loss of asphalt at the 20h PAV and 40h PAV stages is 7.1°C and 7.6°C, respectively. While for UNK+3% $C_{20}H_{42}$, the grade loss of heavily oxidized asphalt decreased from 5.0°C at the 20h PAV stage to 3.2°C at the 40h PAV stage.



(a) RTFO



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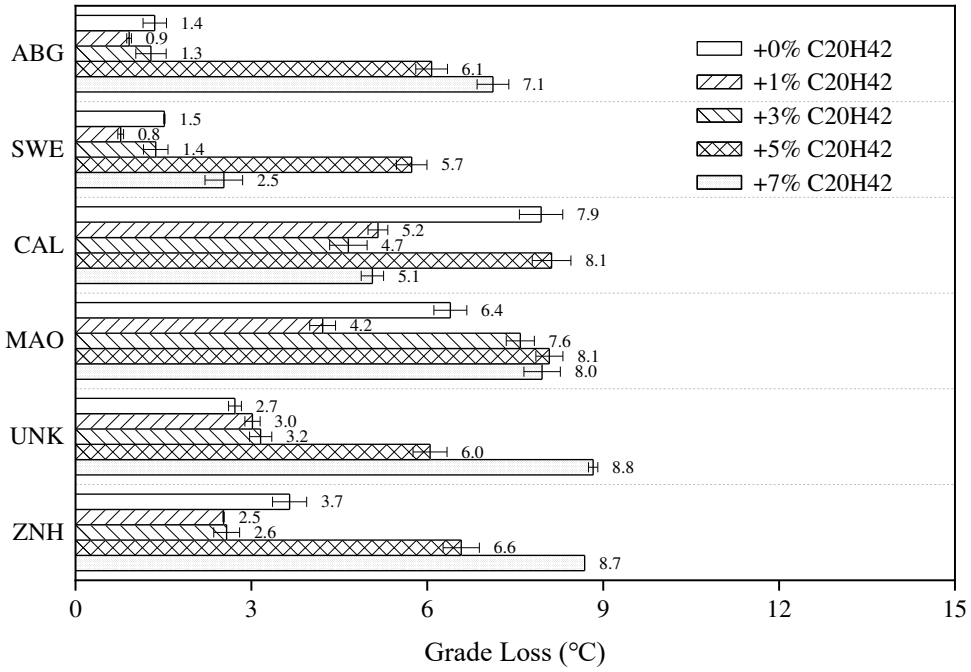
221 (b) 20h PAV



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223 (c) 40h PAV

224 **Fig. 2.** Oxidation effects on grade loss of asphalt binder.

3.3 Storage temperature effects on grade loss of asphalt binder

To further explain the effect of heavy oxidation on the grade loss of MAO+3% $C_{20}H_{42}$ and UNK+3% $C_{20}H_{42}$, this study chose -8°C as the control group based on the original storage temperature (-18°C) to investigate the effect of storage temperature on the grade loss of heavily oxidized asphalt, as shown in Fig. 3. It can be seen from the figure that when the storage temperature is increased from -18°C to -8°C, the grade loss of asphalt also shows an increasing tendency. But for the two different asphalt, the increase in grade loss differs. For MAO+3% $C_{20}H_{42}$, the degree of thermoreversible aging of the asphalt after 20h PAV and 40h PAV remains almost the same after increasing the storage temperature, whereas the difference in grade loss between each is only 0.4°C.As for UNK+3% $C_{20}H_{42}$, the increase in storage temperature unleashed the potential for the degree of thermoreversible aging of heavily oxidized asphalt, and at a storage temperature of -8°C, the grade loss of asphalt after 40h PAV increased by 3.5°C, surpassing the sample after 20h PAV.

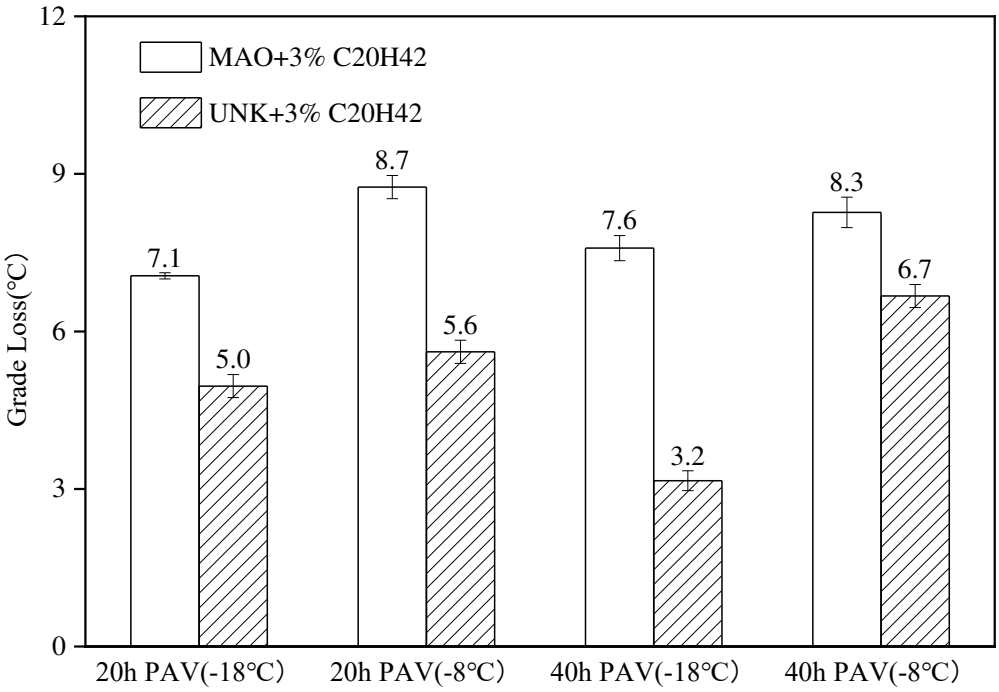


Fig. 3. Grade loss of asphalt binders under different cold storage temperature.

Such phenomenon can be explained according to the phase separation theory [42-45], according to which asphalt can be viewed as a binary mixture consisting of bitumen and wax, a typical binary mixture is shown in Fig. 4. As can be seen in the figure, the distribution states of the phases of the asphalt differ at different temperatures. When the temperature is high enough (T_1), the mixture is stable at any binary ratio. And as the temperature decreases, the mixture may go through a metastable state (T_2) and eventually reach an unstable state (T_3). After entering the metastable state or even the unstable state, the wax phase in the asphalt starts to separate from the asphalt phase, causing the aggregation of wax molecules and eventually leading to the crystalline precipitation of wax in the asphalt. Combined with the findings of Lei [46], it can be explained that the viscosity of asphalt increases after heavy oxidation, which makes it difficult for wax molecules to migrate and thus inhibits wax precipitation. Therefore,

even if heavily oxidized asphalt has reached an unstable state in a low temperature environment, there may not be significant thermoreversible aging inside the asphalt. In this case, the increase in storage temperature will effectively accelerate the movement rate of wax molecules, and the degree of thermoreversible aging of asphalt will be increased.

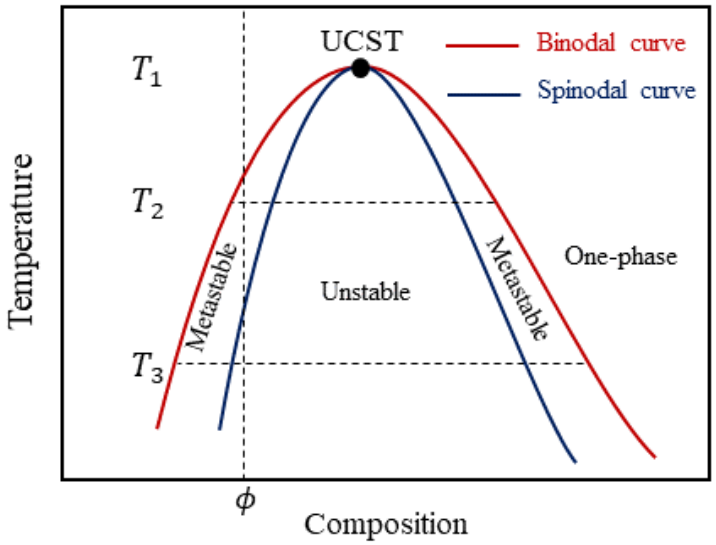


Fig. 4. Possible phase diagram of the asphalt at different temperature.

In order to be able to confirm whether the storage temperature will affect the precipitation and crystallization of n-alkanes other than $C_{20}H_{42}$ in asphalt. In this study, ABG with almost no wax is selected as the base asphalt, and $C_{30}H_{62}$ is used as the additive to study the effect of storage temperature on the grade loss of PAV aged ABG+5% $C_{30}H_{62}$, and the results are shown in Fig. 5. As shown in the figure, if only following the traditional test method, the grade loss of oxidized asphalt (-18°C, 20h PAV) is 2.8°C. By changing the storage temperature and oxidation degree of the tested asphalt, the grade loss is only -0.4°C under the optimal storage conditions (0°C, 20h PAV), while the grade loss of asphalt increased to 5.2°C under the worst storage conditions (-18°C, 40h PAV). This indicates that, like $C_{20}H_{42}$, the effect of $C_{30}H_{62}$ on

the degree of thermoreversible aging of asphalt is quite different at different storage temperatures and oxidation degrees. Therefore, in order to better characterize the thermoreversible aging of heavily oxidized asphalt, multiple storage temperatures should be selected to test the grade loss of the asphalt.

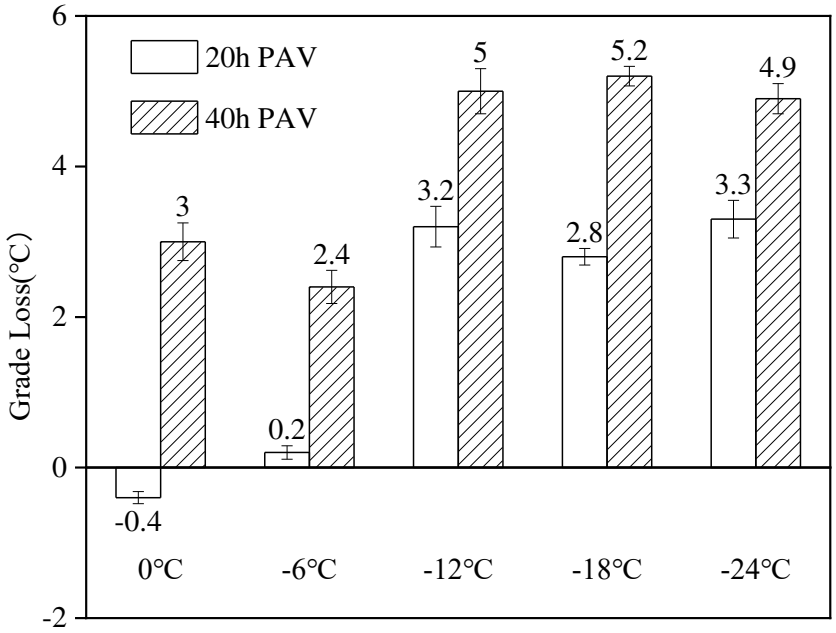
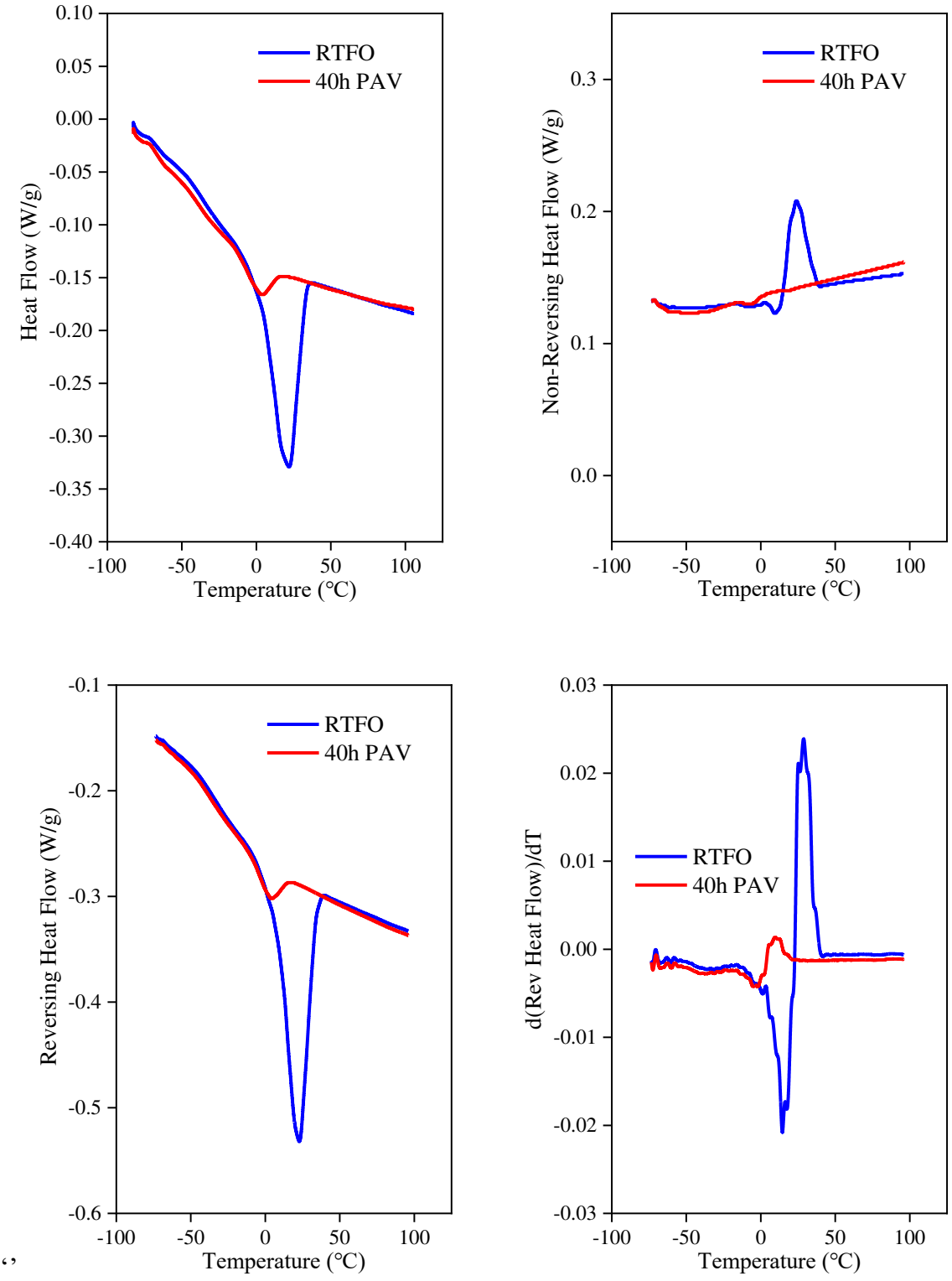


Fig. 5. Grade loss of ABG+5%C30 caused by isothermal conditioning.

3.4 Influence mechanism of oxidative aging on physical hardening of asphalt binder

In order to further explain the influence mechanism of oxidative aging on the physical hardening of asphalt, two aging stages of ABG+5% $C_{20}H_{42}$ are selected for the study, RTFO and 40h PAV, with a grade loss of 2.4°C and 6.1°C, respectively, and MDSC is used to investigate the thermal behavior of ABG+5% $C_{20}H_{42}$ at different oxidative aging stages, as shown in Fig. 6. Based on the non-reversing heat flow curves of the asphalt, it can be seen that there is an endothermic peak in the asphalt at both aging stages, while the area of the

279 endothermic peak reflects the amount of crystal fraction (CF) in the asphalt, which is usually
280 caused by the crystallization or precipitation of long-chain alkane compounds [47, 48].
281 Compared with RTFO, the temperature corresponding to the endothermic peak of asphalt after
282 40h PAV decreased from 9.6°C to -6.0°C, with an area increase of 12.3%. As for the reversing
283 heat flow of asphalt, significant difference in the shape of the DSC curves of asphalt at different
284 aging stages can be observed, indicating that phase separation occurs in asphalt after heavy
285 oxidation. The first derivative of reversing heat flow curves of the asphalt at the RTFO and 40h
286 PAV aging stages have an maximum value at 28.3°C and 9.5°C, respectively, which can be
287 regarded as the glass transition temperature (T_g) of the asphalt [9]. The decrease of T_g of asphalt
288 after heavy oxidation will lead to a larger proportion of $C_{20}H_{42}$ in high-elastic state in the early
289 cooling stage, and this part of $C_{20}H_{42}$ will be changed from high-elastic state to glassy state
290 with higher molecular chain stiffness after long-term low-temperature storage. This means that
291 the heavy oxidation will not only cause more waxes to crystallize out of the asphalt, but will
292 also significantly aggravate the degree of thermoreversible aging of the asphalt by lowering the
293 T_g of the asphalt to a more perfect crystal during low temperature storage.



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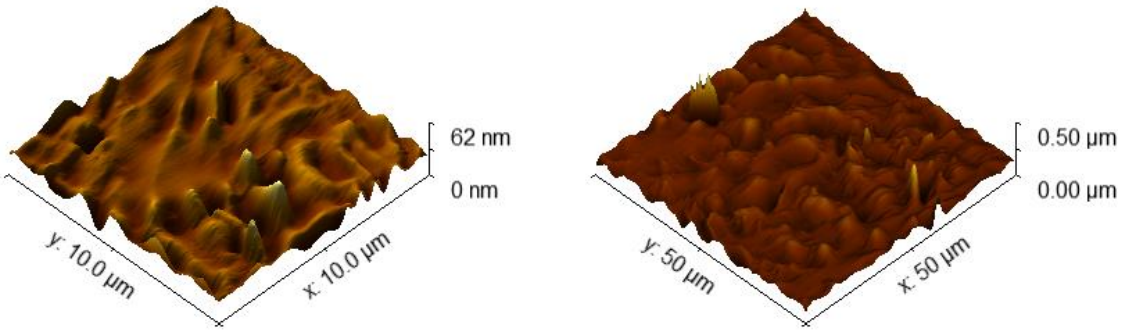
294 **Fig. 6.** Thermal behavior of ABG+5% $C_{20}H_{42}$ under different oxidation degree.

295 And the surface micromorphology images of ABG+5% $C_{20}H_{42}$ under two different aging
296 states in AFM amplitude mode are shown in Fig. 7. From Fig. 7, it can be seen that there are a
297 large number of peaks like protrusions on the asphalt surface after RTFO, while the number of
298 peaks on the surface increases after 40h PAV and its peak value also grows to 0.5μm. Such
299 peak like protrusion is related to the crystallization of microcrystalline wax and waxy
300 molecules when cooled to the test temperature [49]. To quantitatively evaluate the effect of
301 peak like protrusions on the surface morphology of the asphalt, the average roughness (R_a) and
302 root mean square roughness (R_q) are calculated by Gwyddion software to describe the
303 roughness of asphalt, the values of which are shown in Table 2. The results show that after
304 heavy oxidation, the R_a and R_q of the asphalt increased by 5.58 mm and 8.28 nm, respectively,
305 indicating that the increase in oxidation degree will lead to an increase in the surface roughness
306 of asphalt, that is, more wax will precipitate from asphalt crystals, leading to an increase in the
307 degree of thermoreversible aging of asphalt.



(a) RTFO (b) 40h PAV

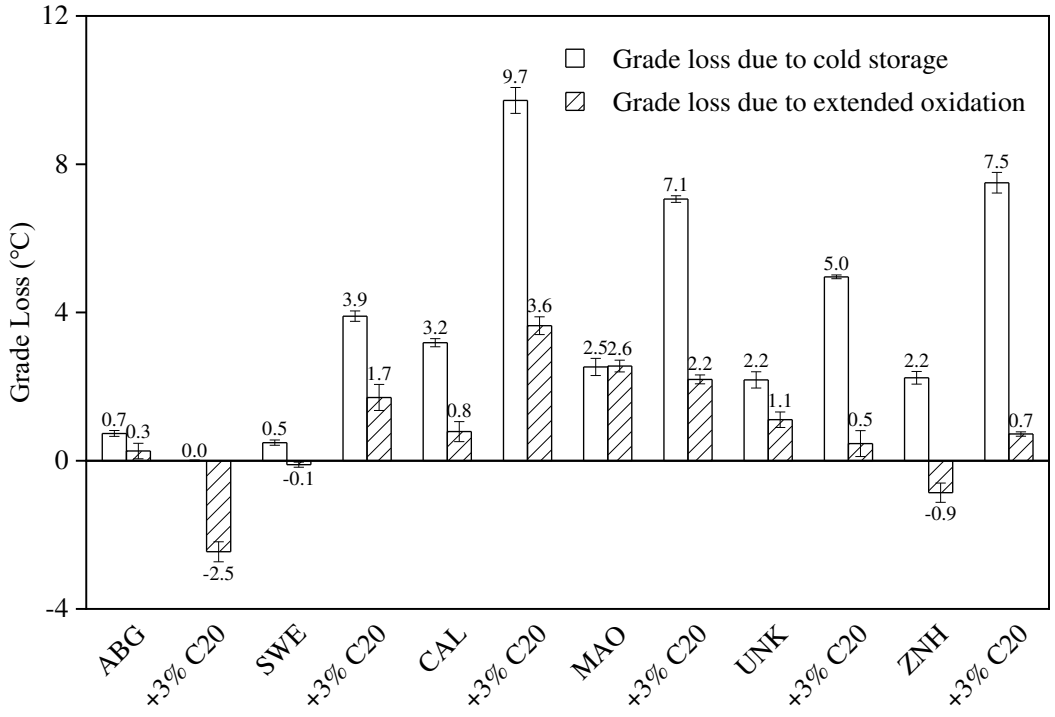
308 **Fig. 7.** AFM images of ABG+5% $C_{20}H_{42}$ under different oxidation degree.

309 **Table 2.** Surface roughness statistics of ABG+5% $C_{20}H_{42}$.

Oxidation degree	R_a (nm)	R_q (nm)
RTFO	10.68	15.17
40h PAV	16.26	23.38

3.5 Comparison between thermoreversible aging and oxidation

Fig. 8 compares the grade loss due to cold storage and extended oxidation. As shown, for most of the asphalts, physical hardening has a stronger impact on grade loss than oxidation. For the asphalt samples not blended with n-alkanes, there is no significant dominance of physical hardening on grade loss, and the difference between the two is less than half a 3°C. In contrast, with asphalt blended with $C_{20}H_{42}$, the grade loss due to cold storage increased significantly, and the grade loss gap caused by cold storage and extended aging grew to 6°C, which indicates that the grade loss of wax-blended asphalt is determined by physical hardening. Therefore, it is necessary to study the effect of asphalt wax content on the degree of thermoreversible aging of heavily oxidized asphalt.



320

321 **Fig. 8.** Grade loss of asphalt binder due to cold storage and extended oxidation.

322 **4. Conclusions**

323 In this paper, the effects of cold storage temperature on the thermoreversible aging
324 characteristics of asphalt binder samples doped with wax additives, especially heavily oxidized
325 asphalts, were studied through a series of laboratory experiments. The following conclusions
326 can be drawn from the results:

327 (1) With the appropriate addition of $C_{20}H_{42}$, the limiting grade of asphalt can be reduced,
328 while due to the different sources of asphalt crude oil, some asphalt with lower wax content
329 can accommodate more n-alkanes. While the introduction of heavy oxidation will not only lead
330 to the deterioration of asphalt low temperature performance, but also lead to the decrease in the
331 capacity for n-alkanes in asphalt.

(2) Among the six asphalt binders, for ABG and SWE blended with $C_{20}H_{42}$, the maximum grade loss of both asphalt binders was only about 4°C during the RTFO stage due to the extremely low wax content, while the grade loss of asphalt after 20h PAV was almost unchanged. However, after 40h PAV, the grade loss of asphalt grows to 6°C, which indicates that asphalt with good grade loss under low aging conditions may also show significant thermoreversible aging behavior after heavy oxidation.

(3) For the $C_{20}H_{42}$-doped asphalt with high viscosity, there is a tendency to decrease the grade loss of asphalt in low temperature storage after heavy oxidation instead. This is due to the viscosity of asphalt increases after heavy oxidation, which makes it difficult for wax molecules to migrate, thus inhibiting the precipitation of wax. In this case, the increase in storage temperature will effectively accelerate the movement of wax molecules, thus releasing the potential of the degree of thermoreversible aging of heavily oxidized asphalt.

(4) Similar to $C_{20}H_{42}$, for heavy oxidized asphalt blended with $C_{30}H_{62}$, the effect of $C_{30}H_{62}$ on the degree of thermoreversible aging of asphalt is quite different at different storage temperatures and oxidation levels, and the grade loss distribution of asphalt varies from -0.4°C to 5.2°C. Therefore, in order to better characterize the thermoreversible aging of heavily oxidized asphalt, multiple storage temperatures should be selected to test the grade loss of asphalt.

(5) The results of MDSC and AFM show that heavy oxidation will not only cause more wax to crystallize out of the asphalt, which leads to an increase in surface roughness, but also

significantly increases the thermoreversible aging of the asphalt by lowering the T_g of the asphalt and forming more perfect crystals during low temperature storage.

(6) By comparing the grade loss due to cold storage and extended oxidation, it can be observed that for most asphalt, physical hardening has a stronger impact on grade loss than oxidation. Especially for asphalt blended with $C_{20}H_{42}$, the grade loss gap caused by cold storage and extended oxidation grows from 3°C to 6°C. Therefore, it is necessary to study the effect of asphalt wax content on the degree of thermoreversible aging of heavily oxidized asphalt.

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