

UDC 628.349.087.7

DOI 10.56525/HBLM2998

INVESTIGATION OF THE AGING PROCESS OF TAR AND DEEPLY OXIDIZED BITUMEN

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Abstract. This study investigates the patterns of thermo-oxidative aging of tar and deeply oxidized bituminous binders with different degrees of oxidation. The processes of technological and chemical aging occurring under the influence of high temperatures and atmospheric oxygen during the production, transportation, and operation of asphalt concrete pavements were considered. Tar IV and deeply oxidized bitumen samples with different softening temperatures were used as research objects. The aging kinetics were evaluated by changes in the softening point after aging according to GOST 33140-2014, as well as under technological aging conditions at 163 °C for 5 hours with continuous stirring. Based on the obtained experimental data, the rates of chemical and technological aging of the binders were calculated. It was established that the maximum rate of property changes is observed during the first hours of aging. The results showed that tar undergoes the most intensive property changes during technological aging, whereas deeply oxidized bitumen is more susceptible to chemical (thermo-oxidative) aging. It was revealed that an increase in the degree of bitumen oxidation leads to higher rigidity, viscosity, and aging susceptibility due to the increased content of resin-asphaltene structures. The obtained results make it possible to evaluate the influence of aging processes on the properties of bituminous binders and can be used in the development of compounding technologies and improvement of the durability of road bitumen.

Keywords: bituminous binder, tar, deeply oxidized bitumen, thermo-oxidative aging, aging kinetics.

Introduction

Authors [1-4] consider thermo-oxidative aging of binders as a two-stage process: technological aging or “short-term” aging [3, 4], during which the bituminous binder undergoes intensive changes in properties and their significant deterioration. This stage includes the preparation of asphalt concrete mixtures, when a thin film of binder comes into contact with mineral aggregates heated to high temperatures, as well as transportation and subsequent paving of the road surface; and operational or “long-term” aging, during which fatigue cracks appear on the surface of the pavement [3-5].

As previously noted, technological aging causes an intensive “metamorphosis” of the organic binder: the amount of maltene components decreases, oxidative destruction leads to the formation of radicals that polymerize into highly carbonized compounds, increasing the content of resin-asphaltene structures [6]. Based on this, the author of [2] suggests that bitumen produced by liquid-phase oxidation is more susceptible to aging processes.

In study [7], investigations were carried out to determine the aging index of bitumen. It was found that the aging index increased by 5 points during the technological aging stage, whereas during operational aging, after 8 years of pavement service, it increased by only 1 point.

It should be noted [5, 8-10] that the organic binder in asphalt concrete undergoes four stages during operational thermo-oxidative aging: strengthening of the internal structure followed by stabilization, onset of destruction, and final failure. Initially, light components evaporate, oils transform into high-molecular resins, and resins convert into asphaltenes. As a result, a coagulation network of asphaltenes is formed, structural-rheological properties improve, adhesion between the bituminous binder and mineral aggregate increases, and the pavement becomes sufficiently strong and plastic.

The second stage of aging is accompanied by an increase in asphaltene content and a growth in the rigidity of the spatial structural network formed by asphaltenes.

At the final stages of the pavement life cycle, destructive processes occur during operation: resins are washed out, the asphaltene content increases, asphaltenes begin to compete for the dispersion medium, separate contact zones between asphaltenes become exposed, cohesive interactions decrease, and carbonization processes occur, i.e., carbon enrichment. As a result, the binder in the asphalt concrete structure becomes viscous, rigid, brittle, less plastic and elastic, while moisture resistance sharply decreases, leading to shrinkage and surface cracking.

Materials and research methods

To determine the kinetics of the thermo-oxidative aging process, a standard method for determining the change in the mass of a bituminous binder sample and its softening point after aging according to GOST 33140-2014 was used. However, samples of the binder were additionally taken every hour during 5 hours to determine the required quality indicators.

To study the dependence of adhesion properties on aging time, mixtures of mineral filler – white marble (an alkaline stone material with low silicon content) – were prepared with: Tar II; deeply oxidized bitumen with softening temperatures of 48, 55, 63 (I), and 71 °C; compounded bitumen obtained by mixing deeply oxidized bitumen with softening temperatures of 48 and 55 °C with tar modified by additives D11 and D22; and compounded bitumen obtained by mixing deeply oxidized bitumen with softening temperatures of 55 and 63 °C with Tar II.

The obtained bitumen-mineral mixtures were heated in an oven at 163 ± 0.1 °C for 8 hours. The kinetics of changes in adhesion properties were determined quantitatively by measuring the mass loss of the sample in a thin film on the mineral material using the boiling method according to GOST 11508-74 (Method A).

Results and Discussion

Aging of bituminous binders causes irreversible changes in their chemical composition, properties, and structural-mechanical characteristics: the content of maltenes decreases, while the number of resin-asphaltene components increases, resulting in higher rigidity and viscosity and deterioration of low-temperature properties. Moreover, the greater the degree of bitumen oxidation, the more susceptible it is to aging.

Binder aging can be considered as a combination of two parallel processes: thermal aging caused by exposure to high temperatures and thermo-oxidative aging caused by the action of oxygen at elevated temperatures. In order to distinguish the contributions of thermal aging and thermo-oxidative aging to the physicochemical properties of binders, the following experiment was carried out.

Tar IV and deeply oxidized bitumen with a softening point of 63 °C were used as the research objects. For Tar IV and deeply oxidized bitumen 63, the dependences of the ring-and-ball softening point after aging according to GOST 33140-2014 and during technological aging were determined. Technological aging was carried out at a temperature of 163 °C for 5 hours under continuous stirring.

Based on the obtained dependences, the rates of chemical aging (in a thin film) (Figure 1) and technological aging (in bulk) (Figure 2) were calculated.

During transportation and storage, bitumen is subjected to various thermal effects, which leads to changes in the dispersion and variability of all its properties. The dependences presented in Figure 3.8 clearly demonstrate the nature of the aging process. For example, after 7 hours of technological aging, the softening point of deeply oxidized bitumen reaches 98.6 °C.

Analysis of the results showed that the maximum rate of change in the softening point of tar and bitumen occurs during the first hours of the aging process. Tar changes its properties most intensively during technological aging, whereas bitumen undergoes more intensive changes during chemical aging. As a result of vacuum distillation of fuel oil, tar apparently contains unsaturated compounds that are highly capable of polymerization and polycondensation reactions even at relatively low temperatures.

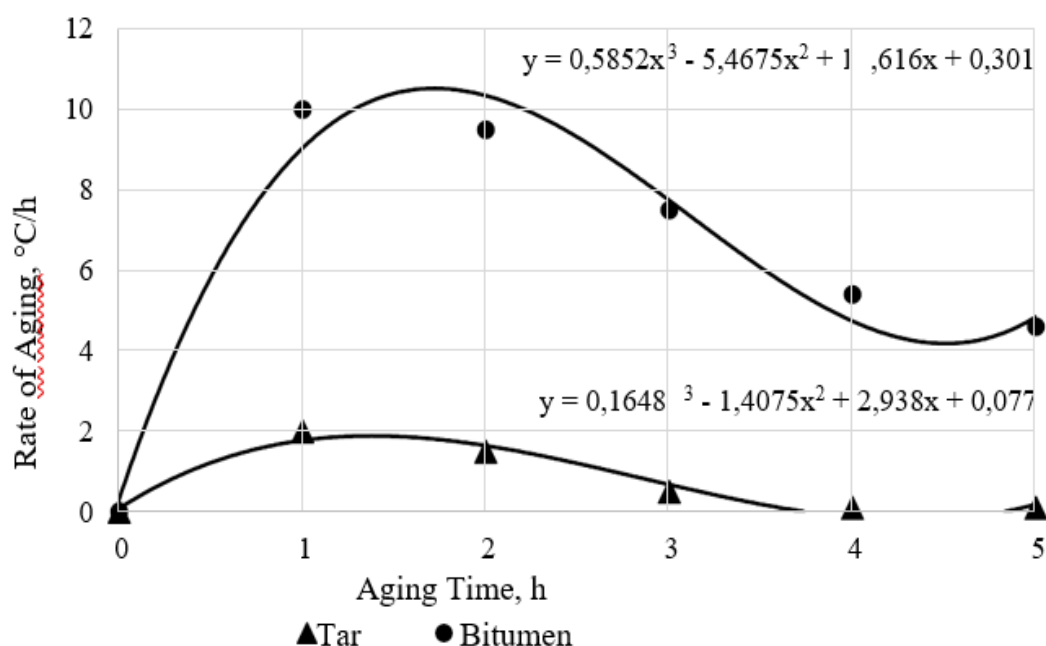


Figure 1 – Chemical Aging Rate of Tar IV and Deeply Oxidized Bitumen

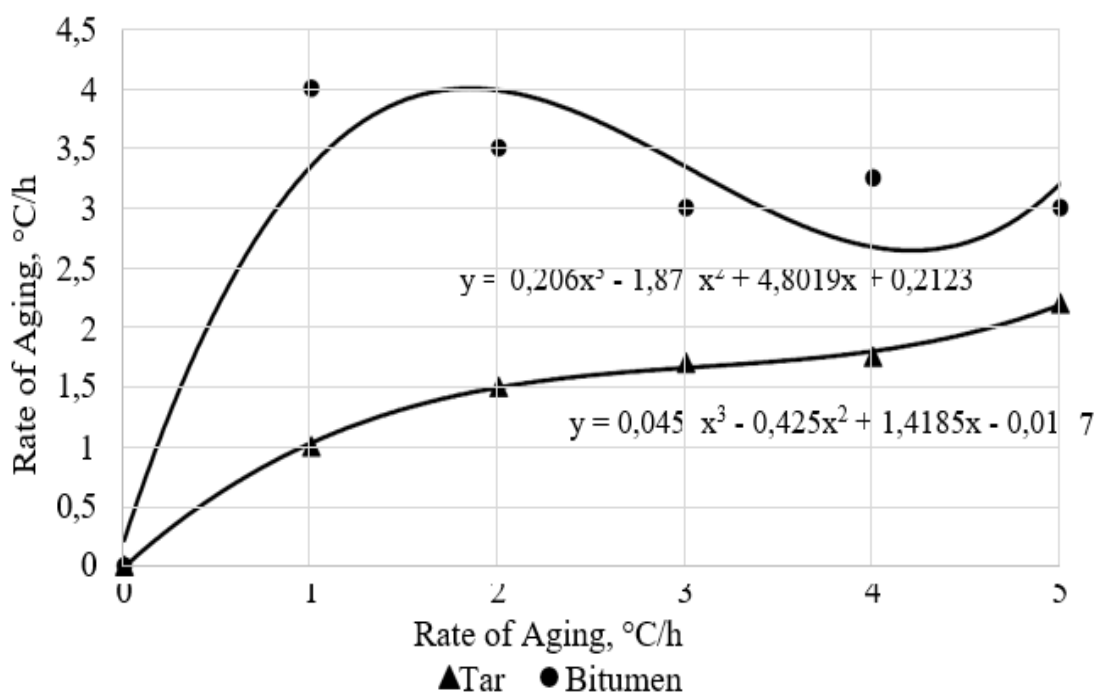


Figure 2 – Technological Aging Rate of Tar IV and Deeply Oxidized Bitumen 63

Therefore, the thermo-oxidative properties of the final product are mainly influenced by the rate of technological aging of tar during the compounding process of deeply oxidized bitumen with tar. However, the depth of oxidation of deeply oxidized bitumen also has a significant influence on the properties of the final bituminous product.

Conclusion

The conducted studies showed that the aging process of bituminous binders is accompanied by significant changes in their chemical composition and structural-mechanical properties. During aging, the content of the maltene fraction decreases while the number of resin-asphaltene components

increases, leading to higher viscosity, rigidity, and deterioration of the low-temperature characteristics of the binder. It was established that the degree of aging strongly depends on the depth of bitumen oxidation: deeply oxidized bitumen is more susceptible to thermo-oxidative changes.

Analysis of the process kinetics demonstrated that the most intensive change in the softening point of both Tar IV and deeply oxidized bitumen 63 occurs during the first hours of aging. Tar is characterized by a higher rate of technological aging, whereas deeply oxidized bitumen is more prone to chemical (thermo-oxidative) aging. The obtained results indicate the presence of a significant amount of reactive unsaturated compounds in tar that are prone to polymerization and polycondensation reactions even at relatively low temperatures.

It was established that prolonged technological aging causes a significant increase in the softening point of deeply oxidized bitumen, indicating intensive structuring of the dispersed binder system. Therefore, during the compounding of deeply oxidized bitumen with tar, the key factor influencing the properties of the final product is the rate of technological aging of tar, while the depth of bitumen oxidation determines the material's resistance to further thermo-oxidative aging during transportation and service life.

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ИССЛЕДОВАНИЕ ПРОЦЕССА СТАРЕНИЯ ГУДРОНА И ГЛУБОКООКИСЛЕННОГО БИТУМА

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Аннотация. В работе исследованы закономерности термоокислительного старения

гудрона и глубооокисленных битумных вяжущих различной степени окисления. Рассмотрены процессы технологического и химического старения, протекающие при воздействии высоких температур и кислорода воздуха в процессе приготовления, транспортировки и эксплуатации асфальтобетонных покрытий. В качестве объектов исследования использовали гудрон IV и глубооокисленные битумы с различными температурами размягчения. Кинетику старения оценивали по изменению температуры размягчения после старения согласно ГОСТ 33140-2014, а также в условиях технологического старения при температуре 163 °С в течение 5 часов при непрерывном перемешивании. На основании полученных экспериментальных данных рассчитаны скорости химического и технологического старения вяжущих. Установлено, что максимальная скорость изменения свойств наблюдается в первые часы старения. Показано, что гудрон наиболее интенсивно изменяет свои свойства при технологическом старении, тогда как глубооокисленный битум более подвержен химическому (термоокислительному) старению. Выявлено, что увеличение степени окисления битума приводит к повышению его жесткости, вязкости и склонности к старению вследствие роста содержания смолисто-асфальтовых структур. Полученные результаты позволяют оценить влияние процессов старения на свойства битумных вяжущих и могут быть использованы при разработке технологий компаундирования и повышения долговечности дорожных битумов.

Ключевые слова: битумное вяжущее, гудрон, глубооокисленный битум, термоокислительное старение, кинетика старения.

ГУДРОН МЕН ТЕРЕҢ ТОТЫҚТЫРЫЛҒАН БИТУМНЫҢ ТОЗУ ҮРДІСІН ЗЕРТТЕУ

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Аңдатпа. Жұмыста әртүрлі тотығу дәрежесіне ие гудрон мен терең тотықтырылған битумды байланыстырғыштардың термототықтырғыш тозу заңдылықтары зерттелді. Асфальтбетон жабындарын дайындау, тасымалдау және пайдалану кезінде жоғары температура мен ауа оттегінің әсерінен жүретін технологиялық және химиялық тозу процестері қарастырылды. Зерттеу нысаны ретінде IV гудрон және жұмсару температуралары әртүрлі терең тотықтырылған битумдар қолданылды. Тозу кинетикасы ГОСТ 33140-2014 стандартына сәйкес қартаюдан кейінгі жұмсару температурасының өзгеруі арқылы, сондай-ақ 163 °С температурада 5 сағат бойы үздіксіз араластыру жағдайындағы технологиялық тозу кезінде бағаланды. Алынған тәжірибелік мәліметтер негізінде байланыстырғыштардың химиялық және технологиялық қартаю жылдамдықтары есептелді. Қасиеттердің өзгеруінің ең жоғары жылдамдығы тозудың алғашқы сағаттарында байқалатыны анықталды. Гудронның технологиялық тозу кезінде қасиеттері қарқынды өзгеретіні, ал терең тотықтырылған битумның химиялық (термототықтырғыш) тозуға көбірек бейім екені көрсетілді. Битумның тотығу дәрежесінің артуы шайырлы-асфальтенді құрылымдардың көбеюіне байланысты оның қаттылығының, тұтқырлығының және тозуға бейімділігінің жоғарылауына әкелетіні анықталды. Алынған нәтижелер битумды байланыстырғыштардың қасиеттеріне тозу процестерінің әсерін бағалауға мүмкіндік береді және жол битумдарының ұзақмерзімділігін арттыру мен компаундтау технологияларын әзірлеуде қолданылуы мүмкін.

Түйін сөздер: битумды байланыстырғыш, гудрон, терең тотықтырылған битум, термототықтырғыш тозу, тозу кинетикасы.