## [Regular Paper]

# Water Accumulation and Behavior of Surfactant Associated with Moisture Permeation in Bituminous Pavement on Concrete Deck Bridge

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Water-related damage is one of the major problems for the durability of the bituminous pavement and concrete slabs of concrete deck bridges. In particular, damage can be drastically accelerated by the intervention of water and some chemicals such as surfactants and salts. Therefore, the mass transfer mechanism of water and the mechanism of chemical absorption in pavement materials are important to study. The authors previously pointed out that water storage in bituminous pavement layers is caused by moisture vapor in the air, and developed a new moisture permeation test apparatus to analyze the mass transfer. Based on this test method, the present study showed experimentally that water and surfactant accumulate in the bridge deck pavement and concrete slabs. Water accumulation in the pavement and deck slabs increases according to the daily climatic fluctuation. The repetitive moisture permeation test showed that alkylphenol-ethoxylate type surfactant applied on the top surface permeates through water-impermeable pavement layers and accumulates in the concrete slab.

#### Keywords

Bituminous mixture, Concrete slab, Surfactant, Moisture permeation, Mass transfer

## 1. Introduction

Water-related damage is one of the major problems for the durability of the bituminous pavements and slabs of concrete deck bridges, because water-mediated damage usually progresses rapidly. Moisture storage mechanisms are important to study as well as the mass transfer mechanism of chemicals in pavement materials. Water accelerates the fatigue failure of concrete structures drastically<sup>1</sup>). In addition to mechanical issues, the intervention of some chemicals such as surfactant and salt also causes damage to concrete structures through chemical effects. For example, salt damage is a typical issue for concrete structures. Disaggregation is another type of irreversible ultimate damage that asphalt or cement binder resolves and eventually the mixture layer decomposes to aggregates. However, the mechanisms have not yet been clarified.

Water intrusion into pavement mixtures is considered to result from penetration of liquid water via surface cracks and/or comparatively large connected voids of the pavement<sup>2),3)</sup>. However, the authors previously found that the cause of water accumulation in pavement mixtures is water vapor permeation and condensation, and developed a new test method to simulate mass transfer in a transitional cyclic environment<sup>4)</sup>. The moisture permeation test suggested that moisture vapor in the air permeates easily into the bituminous pavement layers and accumulates, even though the surface course is practically impermeable to liquid water. Therefore, bituminous pavement mixtures respire (breath) significant amounts of water every day through vapor-state permeation via connected micropores despite the impermeability to liquid water.

As extrinsic chemicals accumulate in concrete structures, surfactant has been detected in several concrete bridge structures<sup>5)</sup>. The source of surfactant is supposed to be window washer fluid or suspended particulate matter (SPM)<sup>5),6)</sup>. Discussions on the circulation of environmental substances are necessary, but the mechanism of diffusion and accumulation in pavement structures is also important to clarify.

This study focuses on refining the mechanism of moisture permeation through pavements by the repetitive moisture permeation test, and also understanding

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the cause of chemical accumulation in bridge deck concrete slabs utilizing the effective moisture permeation test. Although the authors previously proposed the fundamental mechanism of water accumulation as vapor permeation and condensation under specific transitional moisture conditions, the approach was only oneday simulation and for fresh water vapor as a transfer fluid<sup>4</sup>).

This study investigated the following two issues involved in mass transfer in a bituminous pavement and concrete deck slab. The effect of repetition of cyclic climatic fluctuations on water accumulation in bridge pavement. The permeation and accumulation of chemicals in a concrete slab.

## 2. Experimental

This study was intended, firstly, to evaluate the effect of repetition of cyclic environment fluctuations to which actual structures are exposed under natural climate conditions. Secondly, the experiments attempted to demonstrate that chemicals such as asphalt and surfactant permeate into a bituminous mixture and accumulate in a concrete slab.

A bridge deck pavement specimen with surfactant applied on the top surface was used for repetitive moisture permeation testing under cyclic transitional environment conditions. Water absorption was monitored during the permeation test, and other chemicals such as surfactant and asphalt were analyzed by extraction and proton nuclear magnetic resonance (<sup>1</sup>H NMR) spectroscopy after the moisture permeation procedures.

## 2.1. Moisture Permeation Test

The moisture permeation test apparatus consisted of the temperature/humidity programmed-control chamber, cooling unit, electric balance, and data logging device as shown in **Fig. 1**. The apparatus was designed to investigate one-dimensional moisture permeation, and can accurately simulate the cycles of transitional environment condition patterns by controlling the chamber room temperature, relative humidity, and specimen bottom temperature. The system can also record actual environmental conditions and the weight of the specimen holding unit.

#### 2.2. Materials Tested

#### 2. 2. 1. Bridge Deck Pavement Specimen

A specimen of bituminous pavement on concrete bridge deck obtained from a drilled core (see **Fig. 2**) with a diameter of approx. 100 mm taken from an actual bridge deck structure<sup>7</sup> was used for the experiments in this study. **Table 1** shows the outlines of the specimen. The concrete of the deck slab was typical material with a nominal strength of 24 N/mm<sup>2</sup>. Pavement mixtures, porous mixture for surface courses and dense mixture for binder courses also had typical specifications for highway pavement.

The coefficient of gas permeability  $(k_g)$  for the



Fig. 1 Overview of Moisture Permeation Test Apparatus



Fig. 2 Specimen for Repetitive Moisture Permeation Test and Surfactant Application

Table	1	Specimen	of Bituminous	Pavement on	Concrete	Bridge Deck
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	Asphalt co	ncrete (course)	Cement concrete			
		Surface (Specimen-S)	Binder (Specimen-B)			Slab (Specimen-C)
Thickness	[mm]	40	35	Thickness	[mm]	33
Type of mixture		Porous	Dense	Slump	[cm]	$8 \pm 2.5$
Max size of aggreg	ate [mm]	13	13	W/C	[%]	55
Asphalt binder		Modified	StAs40/60	S/A	[%]	47
Asphalt content	[%]	5.6	5.0	AE water-reducing agent [kg/m <sup>3</sup> ] 2.89		2.89

StAs: Straight asphalt, W/C: Water cement ratio, S/A: Sand percentage.

asphalt concrete layers of the specimen was  $3.47 \times 10^{-8}$  cm/s, which was measured only for the binder course because the surface course consisted of porous asphalt mixture. The coefficient of liquid water permeability ( $k_w$ ) can be estimated from  $k_g^{80}$ , and the  $k_w$  of the binder course may be in the order of  $10^{-7}$  cm/s that is substantially impermeable to liquid water<sup>90</sup>. In contrast,  $k_g$  for the concrete slab was  $2.61 \times 10^{-7}$  cm/s. Therefore, it can be considered that a substantially waterimpermeable bituminous mixture covered a comparatively permeable concrete layer.

#### 2.2.2. Surfactant

Surfactants are used widely as detergent as well as emulsifier, foaming/defoaming agent, and antiseptic. Poly-oxyethylene nonylphenylether sodium sulfite is a typical surfactant produced in large quantities. Surfactant is recognized as one of the chemicals accumulated in social infrastructure widely such as pavements and bridges<sup>5)</sup>.

Anionic surfactant  $(n-C_9H_{19}-C_6H_4-O(CH_2CH_2O)_6-SO_3Na)$  with pH 9-10 was used in this study as an entraining chemical with moisture permeation. AE water-reducing agent for fresh concrete is a common surfactant agent and is also an anionic type surfactant, but has a different chemical structure from that of the alkyl-phenol-ethoxylate type surfactant.

## 2.3. Test Procedures

Experimental tests were performed as follows to observe the movement of moisture and surfactant coated on the top surface of the bituminous pavement.

#### 2.3.1. Specimen Preparation

The moisture permeation apparatus simulates one-dimensional mass transfer through a core specimen, so vapor leakage along the wall face of a specimen must be excluded to conduct a valid experiment. Therefore, the specimen was sealed except for the top face by wrapping circumferentially with self-welding type waterproofing seal tape. The proofing tape is made of unvulcanized butyl rubber and is autoadhesive, and is usually used as water sealing material for electric devices and wiring. Prior to the repetitive moisture permeation test, 1.6 g of surfactant was spread on top of the surface course as shown in **Fig. 2**. Consequently, the inner surface of voids of the porous asphalt specimen was coated with the sticky surfactant liquid.

## 2. 3. 2. Repetitive Moisture Permeation Test

**Figure 3** shows the controlled environmental conditions for the repetitive moisture permeation test. In order to perform many cycles, the cycle time was shortened from one day (24 h) to 8 h, which was determined by consideration of the thermal conductivity of the bituminous pavement, although the transitional profile of the environmental conditions was kept same.

#### 2.3.3. Solvent Extraction

After the repetitive moisture permeation test, the concrete slab was split off from the binder course.



Fig. 3 Controlled Environmental Conditions for Repetitive Moisture Permeation Test

Then, the concrete slab was crushed, coarse aggregates were removed, and the concrete was ground and sieved to a fine powder.

The powder of the concrete slab was used for solvent extraction by the soxhlet method to identify the chemical components<sup>5)</sup>. The powder sample was preliminarily extracted with methanol to remove inorganic components. Subsequently, solvent extraction using chloroform was carried out for organic component extraction. A reference sample of concrete without the moisture permeation test was collected from the deck slab and prepared by the same procedures of extraction. **2. 3. 4.** <sup>1</sup>H NMR

<sup>1</sup>H NMR spectra was measured in a CDCl<sub>3</sub> solution to identify the chemical components of the extracts of concrete with/without repetitive permeation test. The spectra of the surfactant and the asphalt binder were also measured for comparative analyses of mass transfer.

# 3. Results and Discussion

## 3.1. Moisture Transfer

**Figure 4** shows the change in the weight  $(\Delta W)$  of the specimen unit (specimen + thermal insulator + cooling unit).  $\Delta W$  is basically equivalent to the amount of moisture absorption into the specimen.

The concrete bridge deck specimen was clearly respiring (breathing) considerable amounts of moisture every 8 h cycle, because  $\Delta W$  showed periodic changes in synchronization with the cycle of the permeation test condition. The amplitude of the cyclic change of  $\Delta W$ was about 2.5 g in the first twelve cycles, but that in the last twelve cycles was reduced to about 2.0 g.

The periodic change of  $\Delta W$  meant that moisture vapor condensed and evaporated in the specimen during each environmental cycle as shown in **Fig. 3**, and about 0.5 g of residual water accumulated every cycle as a result of the difference between the amounts absorbed and evaporated (evapotranspiration). The test system



Fig. 4 Weight Change of Specimen during Repetitive Moisture Permeation Test

demonstrates moisture mass transfer based on vaporstate permeation and dew-point simulation due to the temperature and humidity gap between the top and bottom of the specimen, so the chronological phase difference of temperature change between both sides of the bituminous mixture is probably the cause of the mass transfer imbalance in evapotranspiration.

Micro-pore blockage by accumulated water<sup>8)</sup> is the most significant factor in the declining moisture transfer rate through bituminous mixtures. Accumulated water blocks the internally connected pores because of the large surface tension of liquid water. This sealing effect is certainly one of the causes of permeability reduction. Moisture transfer rate decreases as water accumulation increased, particularly in the binder course. Furthermore, the water-retaining capacity of the concrete deck also gradually diminished due to saturation of water in the layers.

The most important result was that the average weight (envelope curve) gradually increased over the cycles of environmental change. Thus, even though the moisture accumulation in each cycle is small, at about 0.5 g/cycle, total water storage can become considerable through repetition for many cycles.

#### 3.2. Mass Transfer of Surfactant

Spectra were measured with/without the repetitive permeation test to investigate whether mass transfer of chemicals occurs through bituminous pavement. The difference between the spectra may also suggest the characteristics of accumulated chemicals.

Figure 5 shows the <sup>1</sup>H spectra of the surfactant (1), asphalt (2): before permeation test), and chloroform extracts from the concrete deck with (2): before permeation test) / without (4): after permeation test) the repetitive permeation test. The bottom two spectra (3), (4) clearly show the differences between the concrete extracts. The results indicate that many additional peaks are observed after the repetitive permeation test. None of the <sup>1</sup>H spectra of methanol extracts showed specific peaks associated with the surfactant, although



Fig. 5 <sup>1</sup>H NMR Spectrum of the Specimen and Components

some organics might be washed out with the inorganic component.

The peaks between 3.5-4.4 ppm were assigned to the oxy-ethylene moiety,  $-(O-CH_2CH_2)_n$ -(ethoxy group), in the surfactant (1). Note that these peaks are also observed in the spectrum (4). This result provides evidence that surfactant can permeate through the substantially water-impermeable bituminous mixture, and accumulate in the concrete slab. Incidentally, AE water-reducing agent for fresh concrete is also a surfactant, but the NMR spectrum is different from that of the surfactant in this experiment, and is difficult to differentiate from cured concrete. In fact, the spectrum of concrete (3): before permeation test) did not show specific peaks.

The peaks at 0.9, 1.2, 1.5 ppm are assigned to the alkyl protons of the CH<sub>3</sub>, CH<sub>2</sub>, and CH moieties, respectively, as shown in the spectrum of asphalt (2). Although these peaks were observed in various organic samples, the peaks in the spectrum of the concrete deck specimen (4): after permeation test) were definitely from asphalt binder in the layers of bituminous pavement, because there was no other source of such materials within the experimental system. Therefore, certain asphalt fractions may be dissolved and transferred to the bottom concrete slab entrained with moisture and surfactant.

Presumably the surfactant can permeate into the substantially impermeable layer in a very fine mist or droplet of chemicals loaded into the micropores of the bituminous mixture by moisture vapor pressure. The effect of decreasing surface tension and viscosity of water caused by the surfactant's property of surface activity is another possibility.

These results indicated the following. Firstly, surfactant coated on the surface course can permeate through a bituminous mixture through which liquid water cannot permeate. The fact that surfactant accumulates in the concrete deck slab may be important for discussions on durability, because certain types of surfactant can have a negative impact (*e.g.* carbonation of concrete) on durability through chemical reactions such as ionic exchange. In fact, carbonation was observed in the concrete after the permeation test because part of the concrete slab section did not give red color reactions with phenolphthalein. Secondly, asphalt binder in bituminous pavement layers can also permeate the concrete slab entrained with moisture and surfactant. The fact that asphalt binder dissolves and washes out may be important in the mechanism of disaggregation together with the water immersed fatigue phenomenon caused by wheel loading.

## 4. Conclusions

The following conclusions were obtained in this study.

(1) Water storage in bituminous pavement changes in synchronization with the cycle of climate conditions, probably through respiration (breathing) moisture in the air every day.

(2) Cumulative water storage in a bituminous pavement on a concrete bridge deck transferred by moisture vapor increases gradually over the cycles (days) of environmental change.

(3) Laboratory simulation confirmed that surfactant (alkylphenol-ethoxylate type) coated on the surface course can be transferred through water-impermeable pavement layers and accumulates in a concrete slab.

(4) Asphalt binder in bituminous pavement layers can

also permeate the concrete slab entrained with moisture and surfactant.

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#### References

- Matsui, S., J. Jpn. Concrete Eng., 9, (2), 627 (1987).
  松井繁之、コンクリート工学年次論文報告集、9, (2), 627 (1987).
- Kandhal, P. S., Lubold, C. W., Roberts, F. L., The Journal of the Association of Asphalt Paving Technologists, 58, 40 (1989).
- Lottman, R. P., "Predicting Moisture-Induced Damage to Asphaltic Concrete," University of Idaho, Moscow, Idaho, National Cooperative Highway Research Program Report, 192 (1978).
- Sasaki, I., Moriyoshi, A., Hachiya, Y., Nagaoka, N., J. Jpn. Petrol. Inst., 49, (1), 33 (2006).
- Moriyoshi, A., Tabata, M., Kitagawa, H., Tokumitsu, K., Saeki, N., J. Jpn. Petrol. Inst., 45, (2), 84 (2002).
- Moriyoshi, A., Takano, S., Ono, M., Ogasawara, M., Tabata, M., Miyamoto, N., Ohta, S., *J. Automotive Eng.*, (1), 0653 (2000).
- Nomura, K., MD. Thesis, Univ. of Tokyo, Tokyo, Japan, 2003. 野村謙二, "道路橋のコンクリート床版防水工法に関す る研究,"東京大学学位論文, 2003.
- Sasaki, I., Moriyoshi, A., Hachiya, Y., J. Jpn. Petrol. Inst., 49, (2), 57 (2006).
- Japan Society for Civil Engineers, "Hoso-kogaku," Maruzen, Tokyo (1995). 土木学会編,"舗装工学,"丸善,東京 (1995).

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# 要 旨

#### 歴青系橋面舗装の透湿による水分蓄積とこれに随伴する界面活性剤の挙動

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水浸破壊は,橋面舗装およびコンクリート床版における耐久 性確保の大きな課題の一つである。特に,水に加えて塩分およ び界面活性剤のようないくつかの化学物質が介在すると,破壊 が著しく促進されることがある。したがって,舗装材料への水 の物質移動機構や化学物質の蓄積機構は重要な研究課題であ る。著者らは,歴青系舗装への水分の蓄積の主因は水蒸気によ るものであることを指摘し,物質移動の解明のための新しい透 湿試験装置を開発した。本報では,橋面舗装およびコンクリー ト床版に水および界面活性剤が蓄積することを,この試験法に より実験的に実証した結果を述べる。橋面舗装への水の蓄積 は,日周期の環境変化の繰り返しに従って増加する。また,舗 装表面にアルキルフェノールエトキシレート系の界面活性剤を 塗布して透湿試験を行うと,事実上不透水の舗装混合物中を透 過し,コンクリート床版中に蓄積されることを実験室で再現し た。

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