Evaluation of organic coatings for the promotion of dropwise condensation of steam

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Abstract—Several new polymer coatings, as well as gold and silver, were evaluated for their ability to promote and sustain dropwise condensation of steam. Long-term behavior on 25-mm-square specimens was qualitatively observed in an endurance apparatus and heat transfer coefficients on single horizontal tubes were determined in a separate heat transfer apparatus. The organic coatings were successful in promoting good quality dropwise condensation for prolonged periods of times (> 12,000 h). Dropwise heat transfer coefficients as large as six times the film condensation value were obtained with these coatings and results were not dependent upon the thermal conductivity of the wall.

1. INTRODUCTION

ALTHOUGH dropwise condensation of steam has been studied for over 50 years, this unique area of heat transfer has remained as a laboratory curiosity primarily because permanent hydrophobic coatings have not been developed to the satisfaction of condenser designers. Noble metals as well as organic polymers have been used as permanent hydrophobic coatings [1] and, in recent years, various experimental investigations have been conducted to understand the variables which affect the long-term endurance of these coatings when exposed to steam [2-4]. In 1970, Bernett and Zisman [5] showed that pure water spontaneously wets noble metals which are completely free of organic or oxide contaminants. Erb and Thelen [6], however, obtained excellent dropwise condensation on electroplated gold, silver, rhodium, palladium and platinum surfaces. In 1979, Woodruff and Westwater [2] studied dropwise condensation of steam on electroplated gold surfaces, and they found that a minimum thickness of approximately 0.2 μ m of gold was required to obtain perfect dropwise conditions, otherwise mixed condensation would occur. A similar study for electroplated silver was conducted by O'Neill and Westwater in 1984 [3]. They concluded that the lifetime of the silver as a promoter of dropwise condensation depends upon the plating thickness and composition, as well as upon the base metal preparation. When perfect dropwise conditions were observed, they were attributed to relatively large amounts of carbon which were present in the plating, indicating that the carbon was probably the true promoter of dropwise condensation.

As an alternative to noble metals, organic materials with low surface energies can be applied to the condensing surface to yield excellent dropwise conditions [7–9]. However, two difficulties must be overcome before these organic coatings can be used commercially. The first is that there must be a good, long-term adhesion between the coating and the metal substrate. Generally, the thicker the coating, the better its resistance to oxidation and moisture. The second difficulty is that these organic materials have very low thermal conductivities, which requires that the maximum coating thickness must be kept below approximately 5 μ m. Otherwise, the benefits of dropwise condensation are overcome by the increased thermal resistance of the coating itself.

Recently, Holden et al. [4] evaluated 14 polymer coatings for their ability to promote and sustain dropwise condensation of steam at atmospheric pressure. Several of these coatings were also tested for their heat transfer performance by applying them to the outside of horizontal copper tubes. Results indicated that the steam-side heat transfer coefficient can be increased by a factor of 3-8 through the use of these polymer coatings. Furthermore, some of the coatings, after more than 2 years of direct exposure to steam, were still showing good to excellent dropwise condensation behavior. However, they observed that none of the ultra-thin (i.e. $< 1 \mu m$) polymer coatings was capable of completely protecting chemically reactive substrates (such as copper and its alloys) from a steam/ water environment. It was apparent that continued oxidation of the substrate led to failure of the coating and they therefore recommended that further tests be conducted with non-reactive substrates or substrates plated with a non-reactive sub-layer. Non-reactive metals such as stainless steel and titanium are well known for their poor thermal conductivities and a controversy still exists in the literature regarding the effect of substrate thermal conductivity on dropwise heat transfer performance [1].

The objective of the present work was therefore to

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explore the use of additional polymer materials and thin-film coating techniques on various substrates to discover successful ways to promote continued, longterm dropwise condensation. Furthermore, the influence of substrate thermal conductivity, substrate roughness and coating thickness on dropwise condensation heat transfer performance was to be investigated.

2. EXPERIMENTS

Endurance apparatus

Endurance tests were made by continuously exposing prepared samples, which were mounted on a heat sink, to steam at atmospheric pressure. The endurance apparatus consisted of a steam chamber, a heat sink and a de-superheater. Complete details of this apparatus and its operation are given in refs. [4, 10]. Three metal substrates were used for the specimens : oxygenfree, high-conductivity (OFHC) copper, 90-10 copper--nickel and titanium. All the test specimens were 25-mm-square; the copper and titanium specimens were 0.76 mm thick while the copper-nickel specimens were 1.52 mm thick. In addition to smooth substrates, three surface roughnesses were evaluated for their effect on coating adhesion. These included: number 40 glassgrit blast at a gage pressure of 138 kN m⁻², number 220 aluminum oxide grit blast at a gage pressure of 276 kN m⁻² and industrial-size glass-bead blast at a gage pressure of 689 kN m⁻². After surface preparation, all specimens were cleaned for 10 min in an ultrasonic bath of ethanol. The specimens were then sent to various suppliers for application of the polymer coatings.

Visual observations of dropwise condensation on the specimens were conducted periodically. Selected specimens were removed and photographs were taken of these specimens with a scanning electron microscope following the procedures described in ref. [10]. In addition to the visualization of the dropwise performance, tests were also performed to provide a relative indication of adhesion and hardness [10]. Table 1 provides a listing of the coatings which underwent endurance testing. These coatings included electroplated gold and silver and the following organic materials : fluoroacrylic,* parylene,† No-Stik‡ and Emralon.§ Complete details regarding these coatings are provided in ref. [10].

Heat transfer apparatus

In order to determine the quantitative effect of a coating on the heat transfer coefficient, a separate apparatus, as shown in Fig. 1, was used to study dropwise condensation on a horizontal tube. Steam was generated from distilled water in a glass boiler. The steam then passed through a reducer, flowed into a 180° bend and then down into the stainless-steel test section. The test tube was mounted horizontally behind a viewing port in the center of the test section. Steam that did not condense on the tube passed to an auxiliary condenser. All condensate was returned to the boiler by gravity through stainless-steel tubing. Cooling water for the tubes was pumped through a throttling valve to control the flow from zero to a maximum velocity of 4.3 m s⁻¹ through the tube.

A mercury-in-glass manometer, calibrated in millimeters, was used to measure the internal pressure of the system. The cooling water temperature rise through the test tube was measured very accurately by a dual-channel quartz thermometer as well as by a copper-constantan thermopile having 10 junctions on either end. Throughout all of the runs, the measured temperature rise of the quartz thermometer and the thermopile agreed to within ± 0.03 K. A calibrated rotameter was used to measure the coolant flow rate through the tube. With the exception of the manometer and rotameter readings, which were entered manually, all data were interfaced through a data acquisition/control unit.

Since the presence of non-condensing gases can result in significant errors in the dropwise heat transfer coefficient, considerable attention was given to avoid this problem. As documented earlier [10], the test apparatus was extremely leak-tight with a leak rate less than 2 mmHg in a 24-h period (at a pressure of 85 mmHg). In addition, the use of continuous purging using a cold trap-vacuum pump arrangement resulted in virtually no non-condensing gases being present. Additional details of this apparatus may be found in ref. [11].

Each test tube was machined out of OFHC copper, 90–10 copper–nickel, 6061 aluminum or 304 stainless steel. Each had an effective condensing length of 0.133 m, an inside diameter near 13 mm and an outside diameter of either 19.0 mm (thick wall) or 14.2 mm (thin wall). The thin-wall tubes were coated with a wash primer followed by fluoroacrylic on an 'as machined' surface. One copper thin-wall tube and one copper-nickel thin-wall tube were electroplated with silver. The thick-wall tubes were coated with either fluoroacrylic, parylene-D, No-Stik or Emralon following various surface roughness procedures as described in ref. [10].

The dropwise heat transfer coefficient on the outside of the test tubes was determined by subtracting the inside and wall resistances from the measured overall heat transfer resistance as described in ref. [11]. The overall heat transfer coefficient was determined from the measured values for the total heat transfer

^{*}A thermoplastic coating developed by the Naval Research Laboratory.

[†] Generic name for members of a thermoplastic polymer series developed by Union Carbide Corp. Parylene-N, known chemically as poly-para-xylene, contains a central benzene ring attached to which are two CH_2 molecules. Parylene-D is parylene-N with the addition of two chlorine atoms attached to the central benzene ring.

[‡] Trade name of a thermally conducting plastic coating developed by Plasma Coatings, Inc.

[§]Trade name of a fluorocarbon lubricant developed by Acheson Colloids Co.

Coating	Substrate/ surface	Thickness performance Hour					Hours of
		(μm)	Hardness	Adhesion	Initial	Long-term	operation
Fluoroacrylic	Ti/D	2-3	F	3 B	v. good	fair/good	> 16,000*
	Cu/D	2-3	F	3 B	v. good	poor	>16,000*
	CuNi/D	2–3	F	3B	v. good	poor	6500
	Cu–Au/D	2-3	F	3B	good	good	>15,000*
	CuAu/A	2-3	F	3B	good	poor	12,000
	Ti–Au/D	2–3	F	3B	good	good	>15,000*
Parylene-N	Cu/A	0.5	в	1B	good	poor	< 20
	Cu/A	1.0	В	1B	good	poor	< 20
	CuNi/D	0.5	В	1 B	good	poor	< 20
	CuNi/D	1.0	В	1 B	good	poor	< 4000
Parylene-D	Cu/A	0.5	HB	4B	excel	good	>12,000*
	Cu/A	1.0	HB	4B	excel	poor	<100
	Cu/D	0.5, 1.0	HB	4B	excel	v. good	> 12,000*
	CuNi/A	0.5	HB	4B	excel	good	>12,000*
	CuNi/A	1.0	HB	4B	excel	poor	<100
	CuNi/D	0.5, 1.0	HB	4B	excel	v. good	> 12,000*
	Ti/A	0.5	HB	4B	excel	poor	> 12,000*
	Ti/A	1.0	HB	4B	excel	poor	<100
	Ti/D	0.5, 1.0	HB	4B	excel	excel	> 12,000*
	Br/A	0.5, 1.0	HB	2 B	excel	poor	< 20
	Br/D	0.5, 1.0	HB	4B	excel	good	>12,000*
No-Stik(Cu)	Cu, Ti/U	60	4H	5 B	excel	excel	> 16,000*
	CuNi, Ti/U	60	4H	5B	excel	excel	>18,000*
No-Stik(Al)	Cu, Ti, CuNi/U	50	5H	5B	excel	excel	>9000*
Emralon-333	Ti, Br/U	13	F	5B	good	good/v. good	> 18,000*
	Br/U	13	F	5B	good	poor	6500
	Ti/U	13	F	5B	good	good/v. good	>16,000*
	CuNi	13	F	5B	fair	poor	<6500
Gold	Ti/D	0.5	_		excel	excel	>15,000*
Silver	Cu/A	1–2	_		excel	good	> 2000*
	Br/A	1-2			excel	fair	> 2000*
	CuNi/A	1-2	_		excel	excel	> 2000*
Rou A600 grit ; C	ighness —40 grit	Hardn B—softe		hesion -least			

Table 1. Endurance specimens and test results

B-220 grit; D-glass-bead 6H-hardest 5B-most

* Indicates that these samples are still in operation as of 1 October 1985.

rate and log-mean-temperature difference. The conduction resistance of the polymer coating was included in the outside heat transfer resistance. The inside heat transfer coefficient was determined using the Sieder-Tate equation with the appropriate leading coefficient determined by a modified Wilson plot technique. Spiral inserts were used to enhance the inside heat transfer coefficient for the tubes tested. This was necessary because the inside heat transfer resistance can become the governing thermal resistance during dropwise condensation. Appropriate correlations were used to account for fluid property variations with temperature and the fin effect of the tube ends outside the condensing section was included in the analysis.

3. RESULTS AND DISCUSSION

Endurance of the coatings

As shown in Table 1, the long-term endurance of organic coatings is dependent upon the organic structure and thickness of the coating, and the surface

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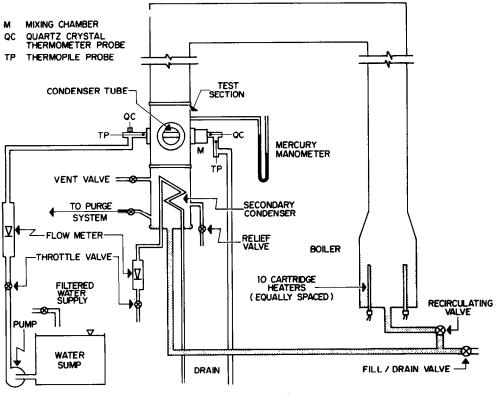


FIG. 1. Schematic of heat-transfer apparatus.

roughness and chemical activity of the substrate metal. All organic materials will absorb water to varying degrees which will cause swelling to occur. In addition, the diffusion of this moisture (and in particular oxygen) through very thin coatings will eventually expose the substrate to a corrosive environment. Therefore, the more impervious the coating is and the less reactive the substrate, the better the endurance will be. Also, if the substrate surface is roughened prior to coating it with an organic material, a more tenacious mechanical bond will exist. This is very important because of thermal expansion differences between the organic coating and the metal substrate.

In general, the fluoroacrylic coating was not as good as the parylene-D coating. As shown in Figs. 2(a) and (b), the fluoroacrylic coating on roughened copper and titanium samples appeared initially to give very good dropwise condensation. However, after 15,000 h of exposure, the copper substrate showed signs of corrosion and exhibited mixed condensation [Fig. 2(c)], whereas the less reactive titanium showed a less severe deterioration of dropwise quality [Fig. 2(d)]. Also, the two fluoroacrylic-coated specimens which had a vacuum-deposited gold sub-layer on a roughened substrate continued to produce good dropwise condensation in excess of 16,000 h. The gold on copper successfully eliminated substrate corrosion, and also improved upon the quality of the dropwise condensation. The smooth copper specimen, however, showed signs of corrosion, indicating a poor bond between the gold and the copper.

All but one of the parylene-N specimens failed within the first 24 h of testing. The roughened coppernickel specimen with the $1.0-\mu$ m-thick coating gave fair to good dropwise conditions for almost 4000 h in marked contrast to the smooth specimen. The parylene-N coating was the softest and least adhering of the organic coatings. In contrast to parylene-N, the parylene-D gave significantly better results. Once again, the smooth specimens failed earlier than the roughened specimens. Figures 2(e) and (f) show the excellent quality of dropwise condensation on the parylene-D coated, rough titanium specimens after 12,000 h of exposure. Clearly, the dropwise quality was better for the parylene-D than for the fluoroacrylic coatings. Apparently, the parylene-D provides a more close-packed, hydrophobic molecule which resists the inclusion of non-hydrophobic groups better than the fluoroacrylic molecule [12].

Figures 2(g) and (h) show the No-Stik specimens which exhibited excellent dropwise condensation conditions even after an exposure of 2 years. These coatings, however, are applied with an approximate thickness of 50 μ m and while they exhibit excellent hydrophobicity, their heat transfer performance was poor, as noted below. The Emralon coatings exhibited an interesting characteristic in that the resin base appeared to be eroded away with time, exposing more

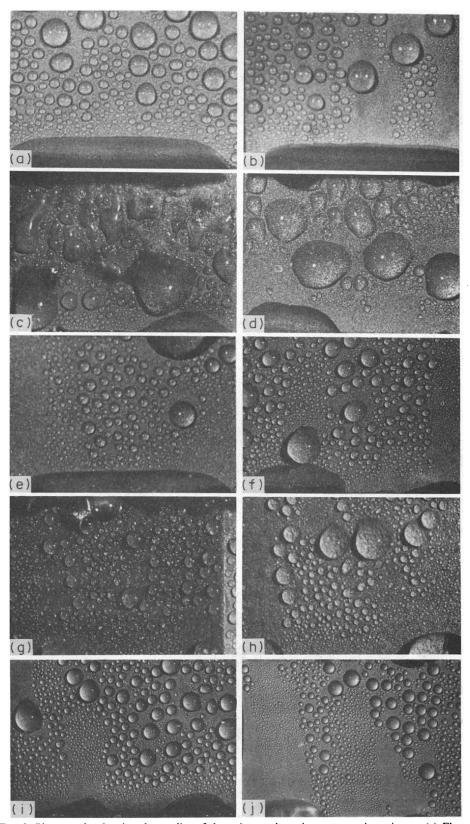


FIG. 2. Photographs showing the quality of dropwise condensation on coated specimens. (a) Fluoroacrylic, copper, rough (700 h). (b) Fluoroacrylic, titanium, rough (700 h). (c) Fluoroacrylic, copper, rough (16,000 h). (d) Fluoroacrylic, titanium, rough (16,000 h). (e) Parylene-D, titanium, rough, 0.5 μ m (12,000 h). (f) Parylene-D, titanium, rough, 1.0 μ m (12,000 h). (g) No-Stik (copper), titanium (18,000 h). (h) No-Stik (aluminum), copper (9000 h). (i) Gold, titanium (18,000 h). (j) Silver, copper-nickel (2000 h).

of the substrate. The quality of the dropwise condensation remained between good and very good even after an exposure of 18,000 h. Figures 2(i) and (j) show the excellent quality of dropwise condensation on the gold- and silver-plated specimens, respectively.

Heat transfer enhancement

Prior to processing filmwise or dropwise condensation heat transfer data using the Wilson plot method [11], appropriate values for the substrate thermal conductivity and the inside heat transfer coefficient must be determined [11]. Proper selection of substrate thermal conductivity was essential to obtain accurate values for the outside heat transfer coefficient, especially for the thick-wall tubes. A sensitivity analysis of the data reduction method showed that a 10% difference in wall thermal conductivity made little difference to the filmwise heat transfer coefficient. However, for the thick-wall stainless-steel tubes, a 10% difference in wall thermal conductivity could cause a 50% change in the dropwise heat transfer coefficient. Appropriate thermal conductivities were chosen from ref. [13].

The inside heat transfer coefficient, using a Sieder-Tate type of equation, was determined using a modified Wilson plot technique [11]. The Sieder-Tate leading coefficients were determined for each tube using spiral inserts to boost the inside coefficient substantially beyond the standard turbulent-flow, smooth-tube value of 0.027. The measured values of the leading coefficients ranged from a low of 0.068 to a high of 0.083 with an average value of 0.075 ± 0.008 . Differences in the measured values can be attributed to slight differences in tube inside diameters, spiral insert pitch and the effect of heat flux on the swirling flow [14]. The value of 0.075 (which indicates a tubeside enhancement near 3) was utilized to reduce all of the condensation data.

Figure 3 shows a summary plot of the condensation heat transfer coefficient vs heat flux. These data were taken for a thick-wall copper tube under vacuum conditions (P = 85 mmHg absolute) and with a steam velocity near the tube of 2 m s^{-1} . The filmwise data and the Nusselt prediction for a horizontal tube are included for comparison. The solid lines through the data represent the best fit assuming a constant heat transfer coefficient over the measured heat flux range. Typical estimated uncertainty limits for the data are also shown. Clearly, the Emralon and No-Stik coatings, because of their large thicknesses (near 13 and 50 μ m, respectively), showed no improvement over the filmwise results. The No-Stik coating performed better than the Emralon, even though it had a coating approximately four times as thick, because the No-Stik coating contained aluminum particles dispersed throughout the plastic. The parylene-D coatings showed enhancement factors of approximately 2 and 3 for the 1.0- and 0.5- μ m-thick coatings, respectively. The fluoroacrylic coating performed the best of the organic materials, showing an enhancement factor of approximately 5.5. Notice that the inclusion of the wash primer, which was utilized to give better adhesion, created an added thermal resistance, and lowered the effective enhancement ratio to approximately 3. It is interesting to note that the thermal conductivity of the parylene-D is estimated to be only one-third that of the fluoroacrylic, which is near that

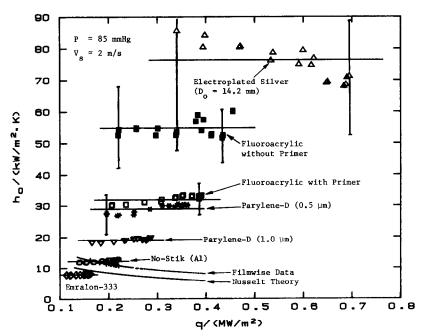


FIG. 3. Heat transfer results for dropwise condensation on coated horizontal tubes.

of Teflon [15]. Thus, even though the parylene-D coatings were thinner than the fluoroacrylic coating, their thermal performance was inferior. These results all point to the significant importance of having the thinnest possible coating. Finally, as expected, the silver electroplated tube gave the best thermal performance, exhibiting an enhancement ratio of approximately 8. The significant spread of the silver data and the trend of increasing heat transfer coefficient with decreasing heat flux were caused by the large uncertainty that results in using the Wilson plot method to infer an outside thermal resistance which is so small in relation to the inside and wall resistances.

Effect of substrate thermal conductivity

Figure 4 shows the dropwise heat transfer coefficient for fluoroacrylic-coated, thin-wall tubes which have been coated first with a primer. Data for four different tube materials are compared and it is evident that, within the uncertainty of the measurements, there appears to be no evidence that thermal conductivity of the wall influences the dropwise data. Unfortunately, since the primer and fluoroacrylic coating were applied by hand, it was impossible to accurately control the coating thickness from one tube to the next and variations in coating thickness could certainly have influenced the results. Therefore, two additional thin-wall tubes of copper and coppernickel were electroplated with silver and these data are shown in Fig. 5. Once again, within the estimated uncertainty of the measurements, there is no evidence of substrate thermal conductivity having an influence

upon the dropwise heat transfer coefficient. These results support the view of Rose [16-18] that rapid coalescence between drops (especially at high heat fluxes) might lead to an essentially uniform surface temperature and to a small effect of substrate thermal conductivity. The opposing view of Mikić [19] where the constriction resistance in the substrate, due to the non-uniform surface heat flux distribution, can influence the heat transfer, predicts that surface thermal properties can readily influence the data [20, 21]. Since the data taken during this investgation were obtained at high heat fluxes, where droplet coalescences and the sweeping effects of large drops are numerous, it is plausible that the constriction resistance effect may not be as important as at low heat fluxes. As a consequence, in operating condensers, the use of low-conductivity-but chemically inactivematerials, such as stainless steel and titanium, might be very desirable on which to apply organic coatings which would give long-term endurance and substantial heat transfer enhancement.

4. CONCLUSIONS

The long-term endurance of organic coatings to promote dropwise condensation of steam depends on the molecular structure and thickness of the organic material, and the surface roughness and chemical activity of the substrate metal. Adhesion of the coating improves when using a roughened substrate which is either chemically inactive or which has been flashed with a chemically inactive sub-layer. Organic coatings

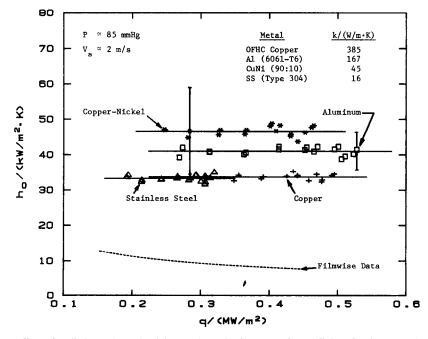


FIG. 4. Effect of wall thermal conductivity on dropwise heat transfer coefficient for fluoroacrylic-coated tubes.

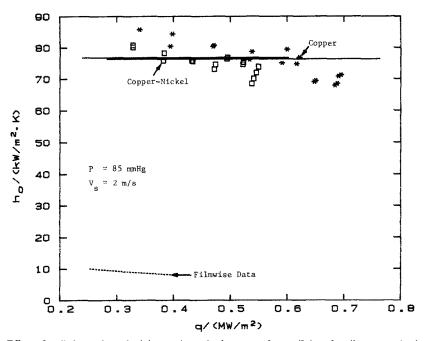


FIG. 5. Effect of wall thermal conducivity on dropwise heat transfer coefficient for silver-coated tubes.

with thicknesses near 1 μ m appears to be successful in promoting good to very good dropwise conditions for 2 years or perhaps more. If thick coatings are utilized, dispersion of metal particles within the organic material may help to reduce the added thermal resistance of the coating itself. Further research is needed to investigate new methods to apply organic coatings in ultra-thin films which can maintain a tenacious mechanical bond with the substrate.

On single horizontal tubes, organic coatings can provide dropwise condensation heat transfer coefficients of steam which are 3-6 times as large as the film condensation values. Although the uncertainty in the measured dropwise heat transfer coefficients was large, due to the use of the Wilson plot technique, the experimental evidence indicates that the effect of wall thermal conductivity did not affect the data. This result may be true at high heat fluxes where droplet coalescences and sweeping effects are numerous. The possible deterioration of the heat transfer results with exposure time to steam remains to be determined.

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EVALUATION DES REVETEMENTS ORGANIQUES POUR FAVORISER LA CONDENSATION EN GOUTTES DE LA VAPEUR D'EAU

Résumé —Plusieurs revêtements de nouveaux polymères, aussi bien que l'or ou l'argent, sont évalués en capacité de favoriser et maintenir la condensation de vapeur d'eau en gouttes. Le comportement à long terme sur des spécimens carrés de 25 mm de côté est qualitativement observé dans un appareil d'endurance, et des coefficients de transfert thermique sur des tubes uniques horizontaux sont déterminés dans des appareils différents. Les revêtements organiques sont efficaces pour donner une condensation en gouttes de bonne qualité, pendant des périodes de temps prolongées (>12 000 heures). Des coefficients de transfert thermique pour la condensation en gouttes jusqu'à six fois ceux de la condensation en film sont obtenus avec ces revêtements et les résultats ne dépendent pas de la conductivité thermique de la paroi.

BEWERTUNG ORGANISCHER BESCHICHTUNGEN ZUR FÖRDERUNG VON TROPFENKONDENSATION VON WASSERDAMPF

Zusammenfassung—Einige Beschichtungen aus neuen Polymeren, auch solche aus Gold und Silber, werden bezüglich ihrer Fähigkeit, die Tropfenkondensation von Dampf zu fördern und aufrechtzuerhalten, bewertet. Das Langzeitverhalten von 25 mm² Flächen wurde qualitativ im Dauerversuch beobachtet, die Wärmeübergangskoeffizienten werden an horizontalen Einzelrohren in einem separaten Wärmeübertragerapparat bestimmt. Die organischen Beschichtungen förderten die Tropfenkondensation gut über längere Zeiten (>12000 Stunden). Mit dieser Beschichtung werden Wärmeübergangskoeffizienten gemessen, die den sechsfachen Wert im Vergleich zur Filmkondensation erreichten, wobei die Ergebnisse unabhängig von der Wärmeleitfähigkeit der Wand waren.

ОЦЕНКА ОРГАНИЧЕСКИХ ПОКРЫТИЙ ДЛЯ СТИМУЛИРОВАНИЯ КАПЕЛЬНОЙ КОНДЕНСАЦИИ ПАРА

Аннотация — Оценена способность нескольких новых полимерных покрытий (а также золотых и серебряных) стимулировать капельную конденсацию пара. Проведено длительное качественное наблюдение над 25-миллиметровыми квадратными образцами в установке для исследования усталостной прочности, а также получены коэффициенты теплопереноса на одиночной горизонтальной трубе в отдельном теплообменном устройстве. Показано, что органические покрытия действительно стимулируют капельную конденсацию в течение длительных периодов времени (> 12000 часов). Для этих покрытий получены значения коэффициентов теплопереноса при капельной конденсации, в 6 раз превышающие их значения при пленочной конденсации, причем результаты не зависят от теплопроводности стенки.