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Refrigeration lubricant Structure Effect on the Foaming Phenomenon in the POE-Refrigerant Mixture

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ABSTRACT

POE refrigeration lubricants are synthesized by different polyols and fatty acids, these various combinations can create different lubricant properties to meet requirements from different refrigeration systems. As global phase down schedule of HFCs, refrigeration lubricant makers focus on modifying structures of incumbent lubricants to apply for alternative refrigerant, such as expanding miscibility range in R32, or reducing viscosity-dilution in R1234ze. However, the modifications usually focus on the miscibility, solubility and working viscosity, there are no many attentions on the other properties, such as refrigerant absorption, desorption, heat transfer, and foaming. Our goal in this paper is to check the effect on foaming property from related structure modification.

Foaming is important phenomenon in refrigeration system, it affects not only oil return, but lead to start-up problem. To detailed describing foaming phenomenon, two stages of bubble life mechanisms must be taken into consideration: bubble generation stage and bubble stable/collapse stage. After investigating a lot of parameters, such as refrigerant solubility in lubricant, surface tension, contact angle, viscosity, density, saturated pressure, temperature and vapor pressure, we find each parameter can play a different role on bubble generation and collapse stage. We try to give a preliminary look on how lubricant chemical structure effect on these parameters, and further checking foaming phenomenon of refrigerant-lubricant mixture through conducting experiments.

In this study, fully branched and fully linear POE with ISO32 and ISO55 were chosen to mix with traditional HFCs, and alternative low GWP refrigerants by different ratios, and conducted foaming tests under different pressure. We also discuss viscosity effect by comparing different viscosity grades range from ISO32 to ISO170. Results show the path that lubricant type influencing foaming phenomenon is through altering solubility and working viscosity of refrigerant-lubricant mixture, then following resulting different foam characteristics. The investigation could provide a better understanding of foaming mechanism, and the direction to well-design lubricants to overcome foaming issue in refrigeration system.

1. INTRODUCTION

As the Kigali amendment was accepted and ratified, HFCs phase-down continue happening in this world. Although HFOs have PAFS issues, the low GWP characteristic is considered one of the HFC alternative refrigerants. Converting the HFCs to HFOs, the refrigeration lubricants also need to evaluate whether they are suitable HFOs or not (Chen et al., 2018, 2022). In these investigations, we synthesized the new structure of refrigeration lubricants by different linear or branched fatty acids and polyols to solve the issue of higher solubility and lower working viscosity in HFO or HFO blend systems. However, in the real HVAC systems, lubricant evaluation not only focuses on miscibility or solubility characteristics, foaming phenomenon is also very important due to lubricant would drain out carried by the refrigerant and cause system to shut down. Foam characteristics of refrigerant/lubricant mixture directly affect the desorption and absorption rate through different static surface tension and dynamic surface tension. (Goswami, D.Y. *et al.* 1998). The common method to solve the foaming issue is adding antifoaming reagent, but it may invalid by running out of additives. We consider the possibility of solving the foaming issue by structure design to generate different type of POE refrigeration lubricants and conducting experiments, try to investigate the foaming phenomenon of refrigerant-lubricant mixture.

2. EXPERIMENTAL

2.1 Lubricants

In the structure design, the linear fatty acids can easily have long chain length to adjust the miscibility or solubility with refrigerants, the branched fatty acids have short chain length but more steric effect to easily increase the viscosity. In this study, we want to realize the chemical structure effect on the foaming phenomenon of refrigerant-lubricant mixture. We selected different types of POE and measured the properties of these lubricants as shown at Table 1.

Table 1: Properties of lubricants

Item	POE-A	POE-B	POE-C	POE-D	POE-E
Type of POE	Full linear	Full branched	Full linear	Full branched	Full branched
40°C kinematic viscosity (cSt)	32	32	55	55	170
25°C kinematic viscosity (cSt)	58	68	112	116	469
VI	144	94	144	105	90
Density@25°C	0.9965	0.9421	1.0018	0.9488	0.9552
Surface tension@25°C	25.99	25.18	25.42	25.58	27.10
Miscibility 20% oil in R134a	-24	-35	-20	-26	-7
Miscibility 20% oil in R1234ze	<-60	<-60	<-60	<-60	<-60

2.2 Foaming instrument

Typically, lubricants followed ASTM D892 standard test method for foaming characteristics of lubricating oils and modify the bubble source from air to refrigerants. By the bubbling through the oil at different temperatures, letting stand for 5 and 10 mins, and recording the foaming tendency as shown at Figure 1. We also modified prof. J. R. Barbosa's equipment (Fortkamp and Barbosa Jr, 2015) to do the foaming test with refrigerants. (Figure 2) Charge lubricants into the chamber with magnetic stir. Removing the air with vacuum pump, and then charging refrigerant into the chamber. Put the autoclave into the water bath keeping it in 25°C and stir 150 rpm to well mix the refrigerant and lubricant 30 mins. Release the refrigerant by ball valve to observe the foaming phenomenon.

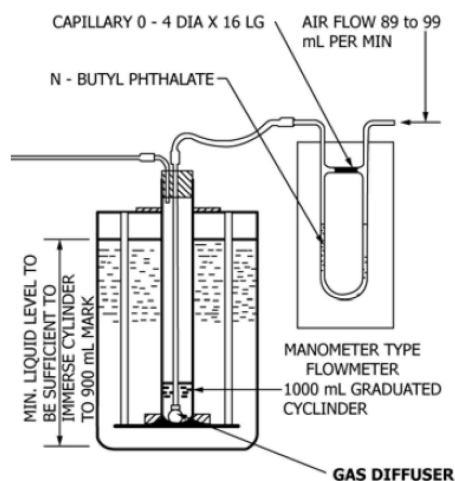


Figure 1: Foaming instrument- ASTM D892 standard test

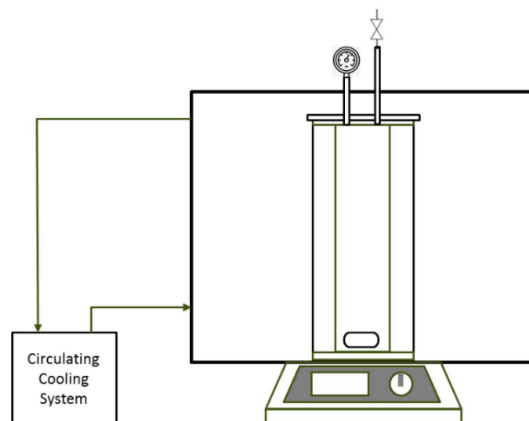


Figure 2: Foaming equipment-modified Prof. J. R. Barbosa's method

3. FOAMING TEST OF REFRIGERATION LUBRICANTS

3.1 Foaming test- ASTM D892 method

We utilized the ASTM D892 method recording the volume of foam at Table 2. When stopping air supply, lower viscosity oil like ISO-32 or 55 will be quickly foam collapses to 0 mL, the higher viscosity like ISO-170 foam will keep at high volume than other ISO grade.

Table 2: Foaming test- ASTM D892 method

Item	POE-A	POE-B	POE-C	POE-D	POE-E
ISO grade	32	32	55	55	170
sequence I (24°C)	0/0	0/0	0/0	nil/0	100/0
sequence II (93.5°C)	0/0	0/0	0/0	0/0	0/0
sequence III (24°C)	0/0	0/0	0/0	nil/0	60/0

We modified ASTM D892 method, changing the air supply to R134a and R1234ze, the lubricant selected POE-E and other procedure was same as ASTM D892. The test results are extremely different than air bubbling at Table 3.

Table 3: Foaming test- ASTM D892 with refrigerants

Item	POE-E		
	Air	R134a	R1234ze
Gas Flow rate 90~100 mL/min			
sequence I (24°C)	100/0	0/0	0/0
sequence II (93.5°C)	0/0	0/0	0/0
sequence III (24°C)	60/0	0/0	0/0

3.2 Foaming equipment-modified Prof. Barbosa's method

In the foaming test of prof. Babosa's method, we modified method by 150 rpm stirring rate, in order to let it more easily observation and repeatable. When releasing the pressure of autoclave, the refrigerants will desorb from the refrigerant and lubricant mixture. Foam will increase quickly to the max foaming height and then collapses to less than 0.5cm. Recording the max height and decay time. The time sequence of the foaming image was shown at Table 4. We conducted the foaming test of different oils with R134a and R1234ze, the results were shown at Table 5 & 6, and verified different amount of refrigerant effect on foaming at Table 7.

Table 4: Sequence of image for POE-E with R134a


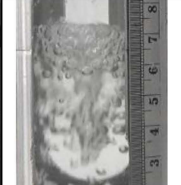
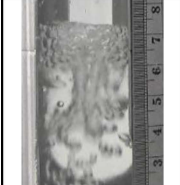

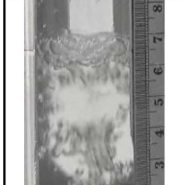

R134a@ 25°C	0 sec	1 sec	2 sec	5 sec	10 sec	20 sec
Initial pressure (MPa)	0.42	0.01	0.01	0.01	0.01	0.01
Foaming height pic						

Table 5: Foaming test with R134a

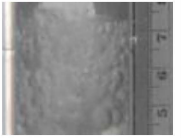
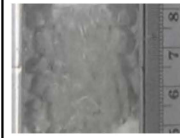

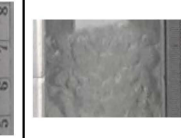






R134a@ 25°C	POE-A	POE-B	POE-C	POE-D	POE-E
Ratio (RF/ (RF+oil) wt%)	33%	33%	33%	33%	33%
Mixture Viscosity (cSt)	6.0	5.1	6.8	6.1	11.2
Initial pressure (MPa)	0.37	0.37	0.37	0.37	0.42
Max foaming height (cm)	2.8	3.0	2.9	2.4	1.5
Max foaming height pic					
Foaming height decay to <0.5 cm (sec)	3	3	3	3	10
Bubble size without foaming pic					

Table 6: Foaming test with R1234ze











R1234ze@ 25°C	POE-A	POE-B	POE-C	POE-D	POE-E
Ratio (RF/ (RF+oil) wt%)	33%	33%	33%	33%	33%
Mixture Viscosity (cSt)	5.1	4.2	7.6	5.0	6.2
Initial pressure (MPa)	0.24	0.23	0.25	0.24	0.25
Max foaming height (cm)	1.5	2	1.5	1.8	3.5
Max foaming height pic					
Foaming height decay to <0.5 cm (sec)	5	5	5	5	15
Bubble size without foaming pic					

Table 7: Foaming test with different ratio with R134a & R1234ze

	POE-E			
Ratio (RF/ (RF+oil) wt%)	33%	30%	33%	30%
Refrigerant	R134a		R1234ze	
Max foaming height (cm)	1.5	<0.5	3.5	<0.5

4. FOAMING MECHANISM

To investigate foaming mechanism, three stages of bubble life should be considered - bubble generation, bubble stable, and bubble collapse.

4.1 Bubble generation period:

It includes homogeneous and heterogeneous bubble formation (Blander, M. & Katz, J, L., 1975). For homogeneous bubble formation, pressure gap between inside and outside bubble must overcome interfacial tension of liquid to generate bubble. It obeys Young-Laplace equation as following equations

$$n = N * \exp\left(\frac{-W}{kT}\right) \quad (1)$$

$$\Delta P = P_G - P_L = \frac{2\sigma}{r} \quad (2)$$

That means interfacial tension and pressure difference between bubble surface will influence the critical bubble size, which represents minimum radius that a bubble could inception. Regarding bubble amount of generation (J_n), interfacial tension and liquid and vapor pressure difference are key factors, and following is viscosity.

$$J_{\eta,homo} = N \frac{\sigma}{\eta} \left(\frac{\sigma}{kT}\right)^{\frac{1}{2}} \left(1 - \frac{P_L}{P_V}\right) \exp\left(\frac{-16\pi\sigma^3}{3kT(P_V - P_L)^2}\right) \quad (3)$$

Small interfacial tension or large pressure gap leads to more bubble numbers generation. In real case, homogeneous bubble formation, bubble nucleation happens in bulk liquid, and it is not the main process because of lacking active sites to generate bubbles, therefore higher temperature or higher pressure gap are needed. Most bubbles are foaming through heterogeneous bubble formation. The bubble nucleation happens in defects on solid surface (or roughness), it will dramatically decrease bubble formation temperature due to different contact angle. But the key parameters and tendency are same as homogeneous bubble formation.

$$J_{\eta,heter} = N^{\frac{2}{3}} S \frac{\sigma}{\eta} \left(\frac{\sigma}{kTF}\right)^{\frac{1}{2}} \left(1 - \frac{P_L}{P_V}\right) \exp\left(\frac{-16\pi\sigma^3 F}{3kT(P_V - P_L)^2}\right) \quad (4)$$

$$m' = \cos \theta \quad (5)$$

$$S = \frac{(1-m')}{2} \quad F = \frac{2-3*m'+m'^3}{4} \quad (6)$$

After inception stage, a bubble would grow up quickly to its maximum bubble size. The driving force of this expansion comes from pressure, and retarding by viscosity (Barlow, E. J. & Langlois, W. E., 1962).

$$R_d = R_{cr} + (R_0 - R_{cr}) \times \exp\left(\frac{(P_V - P_L)t}{4\eta}\right) \quad (7)$$

Different refrigerants will provide different saturate vapor pressures, and generate different interfacial tensions with various lubricants. The more miscible refrigerant-lubricant pair means owning smaller interfacial tension, and results

in small bubbles due to small contact angle. In addition, viscosity and lubricant weight percentage of refrigerant-lubricant mixture will also affect bubble radius, i.e. higher lubricant concentration (or more viscous lubricant) will lead to small bubble (Hung et al, 2020, 2021).

$$R_{d,mix} \propto \theta \left[\frac{\sigma}{g(\rho_L - \rho_v)} \right]^{\frac{1}{2}} (Ja^*)^{\frac{5}{4}} \tag{8}$$

$$Ja^* \equiv \frac{\rho_L c_p T_{sat}}{\rho_v \lambda} \tag{9}$$

4.2 Bubble stable & collapse periods

The stability of a foam is determined by a number of factors involving both bulk solution and interfacial properties, such as gravity drainage, capillary suction, surface elasticity, viscosity, electric double-layer repulsion, dispersion force attraction and steric repulsion. Among these factors, gravity drainage is the most important. Drainage is the passage of liquid through a foam. It is easily observed in daily life, in beer foam for example. Immediately after foam generation, there will always be a tendency for liquid to drain due to the force of gravity. This liquid will drain by flowing downward through the existing liquid-films, the interior of the lamellae thinning, and lead to film rupture and cause foam collapse. To describe drainage phenomenon, *Darcy’s law* is using.

$$Q = -\frac{kA}{\eta L} \Delta P \tag{10}$$

Darcy’s law is an equation that describes the flow of a fluid through a porous medium. In this equation, volumetric flow rate Q is directly proportional to permeability k, cross-sectional area A and pressure ΔP, and inversely proportional to viscosity η and length L. That implies that viscosity is the key factor of foam stability. High viscous lubricant will retard the liquid drain out and keep foam stable. In addition, higher viscosity provides drag force for bubble merging which is important phenomenon for bubble collapse, and smaller buoyancy will result in a slower rising speed. The force of viscosity on a small sphere moving through a viscous fluid is given by *Stokes Law*:

$$F = 6\pi\eta r v = (\rho_v - \rho_L) g \frac{4}{3} \pi r^3 \tag{11}$$

$$v = \frac{2}{9} \frac{\rho_v - \rho_L}{\eta} g r^2 \tag{12}$$

4.3 Overall comparison:

Key factors such as interfacial tension, vapor pressure, viscosity, solubility play as important roles at different stages of forming. Take viscosity of refrigerant-lubricant mixture for example: higher viscous liquid tends to generate small amount of bubble and create small size bubbles, it will avoid foaming phenomenon; But it’s a drawback for drainage and merging stage to collapse. We summarize these comparisons as following table 8, 9.

Table 8: Bubble generation periods

$\sigma_L \downarrow$	$\sigma_{RF-oil} \downarrow$	$J \uparrow$
$\Delta P \uparrow$	$P_v \uparrow$	
$\eta_L \downarrow$	$P_L \downarrow$	
$\chi_{Light} \uparrow$	$\eta_L \downarrow$	
	Solubility \uparrow	

Table 9: Bubble stable & collapse periods

$\eta_L \uparrow$	$v_b \downarrow$	Bubble easy accumulation	
$\eta_L \uparrow$	$v_d \downarrow$	Lamella not easy thinning	Bubble not easy collapse, easy foaming
$J \uparrow$	$v_d \downarrow$		

5. DISCUSSION

5.1 ASTM D892, air & refrigerant

In ASTM D892 foaming test, linear or branched chemical structures are not the main effect the foaming volume high or low, the factor leads high foam tendency is oil viscosity. However, POE-E is incumbent refrigeration lubricants used in R134a, the refrigerant amount dissolved in the lubricants will verify by the working condition in HVAC. ASTM D892 method is air supply, air isn't soluble in the lubricants and does not relate to real condition. If we replace air source with refrigerants, the tests showed no foam occurrence and it observed like the refrigerant dissolve in the lubricants. ASTM D892 method can't evaluate the lubricant foaming tendency in HVAC application.

5.2 Prof. Barbosa's method (Table 5,6,7,10)

Stirring effect: Comparing the ASTM D892, prof. Barbosa's method shows well-relationship with real HVAC application. We found the unstable results if testing under without-stirring condition. The liquid will keep stable and no bubble occurrence for few minutes like picture 0 sec at Table 4 even turning on the valve to release the pressure. Once stir starting, foaming inception. The reason is that pressure of liquid is not yet equal to outside pressure under without-stirring condition, even turned on the valve, and keep it at vapor pressure of $P_L = x \cdot \gamma \cdot P_{sat}$. Therefore, the drive force is only $(P_{sat} - P_L)$, it is not enough to trigger the bubble inception. After stirring, all pressure approach equilibrium to atmosphere, the drive force of $(P_{sat} - P_{atm})$ is enough for bubble formation. The results can be estimated as calculation shown on Table 10.

Refrigerant effect: Bubble size of R-1234ze is around 0.1~0.3 cm, and that of R-134a is around 0.3~0.5 cm. The reason is that although R-1234ze possesses higher surface tension, but the interfacial tension of refrigerant-lubricant is small due to its excellent miscibility, and result in small contact angle. Therefore, it leads to small size bubble generation and large bubble numbers. Regarding saturated pressure, R-1234ze saturated pressure is lower than that of R-134a, then would result in lower initial pressure. Therefore, small pressure gap will reduce the bubble numbers. The different effect on bubble numbers competes each other among interfacial tension, contact angle and pressure gap. Obviously, interfacial tension and contact angle effects on bubble numbers outweigh pressure difference effect. In addition, excellent miscibility of R-1234ze also make lower working viscosity.

Viscosity effect and lubricant type effect: In the same ISO grade, the linear and branched type POE have similar miscibility and working viscosity, when foaming occurred, the foaming height and decay time were similar. Check the difference phenomenon of higher viscosity POE-E in R134a, POE-E had different results at max foaming height and decay time lasted longer than low viscosity lubricants. We observed the bubble size is different between low and high viscosity lubricants. When bubble generating, the bubble size are similar, low viscosity lubricants and R134a mixture bubbles merged together easily than POE-E in R134a. That's why the max foaming height images could see the difference size bubbles at Table 5 and POE-E foaming height was lower and last longer than other lubricants. Because of viscosity of POE-E was higher the bubbles not easily form one big bubble. When the foaming collapsed, all pictures showed the similar bubble size. Converting R134a to R1234ze, the foaming generation of POE-E in R1234ze is higher than that of POE-E in R134a. Observing the foaming process of POE-C and POE-E, although POE-C mixture viscosity is higher than POE-E mixture in R1234ze, when refrigerants desorbed from the mixture, the viscosity of POE-C is smaller than POE-E at 25°C. POE-C bubble easily merged together becoming bigger bubble to the surface, however POE-E bubble was small and not easily merged and stacked together led to higher foaming height and last longer to collapse.

Solubility effect: The most meaningful method of solving foaming issue in HVAC system is controlling solubility of refrigerant in lubricant (Table 7). Solubility is affinity between refrigerant and lubricant at high temperature, that may be different with miscibility (low temperature affinity), and can be optimized through well-designed chemical structure of lubricants.

Table 10: Bubble generation period calculation

	Unit	R-1234ze / POE-E		R-134a / POE-E	
		Initial	Stir-Final	Initial	Stir-Final
k	m ² *kg/(s ² *K)	1.38E-23	1.38E-23	1.38E-23	1.38E-23
NA		6.02E+23	6.02E+23	6.02E+23	6.02E+23
Atmospheric pressure	Pa	101325	101325	101325	101325
σ	N/m	0.008	0.008	0.0072	0.0072
η	Pa-s	0.0062	0.0062	0.0112	0.0112
Oil MW	g/mol	866	866	866	866
RF MW	g/mol	114	114	102	102
m	Kg	1.89E-25	1.89E-25	1.69E-25	1.69E-25
wt%	%	33	33	33	33
molar ratio x		0.789098741	0.789098741	0.807014571	0.807014571
N	1/m ³	1.74E+27	1.74E+27	1.95E+27	1.95E+27
T	K	298	298	298	298
B		0.92969958	0.734417208	0.935671524	0.717427134
Psat	Pa	4.99E+05	4.99E+05	6.65E+05	6.65E+05
Pliquid	Pa	3.93E+05	1.01E+05	5.37E+05	1.01E+05
Rcr	m	1.52E-07	4.03E-08	1.12E-07	2.55E-08
Wcr		7.76E-16	5.44E-17	3.79E-16	1.97E-17
Exp(-Wcr/kT)		0	0	0	0
τ	s	5.88E-12	5.23E-12	5.88E-12	5.15E-12
Homogeneous bubble generation					
$J\eta$	1/(m ³ *s)	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Heterogeneous bubble generation					
Contact angle θ		20	20	26.2	26.2
m'		0.939692342	0.939692342	0.897257898	0.897257898
S		0.030153829	0.030153829	0.051371051	0.051371051
F		0.002672925	0.002672925	0.00764582	0.00764582
$J\eta l$	1/(m ³ *s)	0.00E+00	2.80E+10	0.00E+00	7.77E+09
Bubble growth & rising velocity					
Rd,max	m	0.001	0.001	0.002	0.002
Growth time	s	2.10E-06	6.40E-07	3.40E-06	9.00E-07
Vapor density	Kg/m ³	26.32	26.32	32.35	32.35
Liquid density	Kg/m ³	1027	1027	1038	1038
Velocity	m/s	0.3517444	0.3517444	0.782730917	0.782730917
Height	m	0.07	0.07	0.07	0.07
Rise time	s	0.199008143	0.199008143	0.089430478	0.089430478

6. CONCLUSIONS

In this study, we try to give a preliminary look on parameter effects. Such as refrigerant solubility, interfacial tension, contact angle, vapor pressure, viscosity, density, and temperature, we find each parameter can play a different role on bubble generation and collapse stage. After checking foaming phenomenon of refrigerant-lubricant mixture through conducting experiments, we found that the path of linear or branched structure of POE influencing foaming phenomenon is through altering solubility and working viscosity of refrigerant-lubricant mixture, then following resulting different foam characteristics. Since the most important effect on the foaming issue appear on solubility of the refrigerant and lubricant, the reasonable way to improve foaming issue is controlling solubility through structure

designed. Converting HFCs to HFO, the characteristics of lubricant in HFO mixture are much different than lubricant in HFC. We need to generate the different lubricants to solve the problem may happen in the HVAC application.

NOMENCLATURE

A	Cross-sectional area
C_p	Specific heat of liquid
J	Bubble number generation rate, $1/(m^3*s)$
$J_{\eta,homo}$	Homogeneous bubble number generation rate, $1/(m^3*s)$
$J_{\eta,heter}$	Heterogeneous bubble number generation rate, $1/(m^3*s)$
k	Permeability
L	Length
PAFS	Per- and Polyfluoroalkyl Substances
P_L	Pressure outside bubble (Liquid pressure)
POE	Polyolester
P_V	Vapor pressure inside bubble (equal to P_{sat})
ΔP	Pressure
Q	Volumetric flow rate
R_0	Bubble radius at $t=0$
R_{cr}	Critical radius of bubble inception
$R_{d,max}$	Maximum radius of bubble in refrigerant/oil mixture
T	Temperature
T_{sat}	Saturated temperature of fluid
v_b	Bubble rising velocity
v_d	Velocity of liquid drainage in lamella
χ_{light}	Molar fraction of light (refrigerant)
η	Viscosity
η_L	Liquid viscosity
θ	Contact angle
λ	Latent heat of vaporization
σ_L	Interfacial tension (surface tension) of liquid

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