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Preparation of a Novel Water-based Acrylic Multi-Thermal Insulation Coating

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To efficiently improve the thermal insulation effect of coatings, a novel water-based acrylic multi-thermal insulation coating (multi-WATIC) combined with thermal obstruction, echo, and radiation was prepared. The category and ratio of thermal insulation functional fillers are crucial. First, water-based acrylic thermal insulation coating (WATIC) with single thermal insulation functional fillers was prepared, and the thermal insulation property tests were done. Thereafter, a novel multi-WATIC was prepared combined with the 3 thermal insulation functional fillers together, and the formula of the novel multi-WATIC was optimized based on single factor experiments by response surface methodology (RSM). Test results showed that multi-WATIC has excellent thermal insulation property, and the fitting result obtained by RSM is in good agreement with test data.

Keywords: waterborne, multi-thermal insulation, temperature difference, response surface methodology, optimization.

1. INTRODUCTION

Thermal insulation coatings control the temperature of buildings, improve indoor thermal comfort. They are a convenient and effective method to save energy [1 – 3]. Thermal insulation coatings can be divided into three different categories based on their mechanism: barrier, echo and radiation coatings. In America, Japan, Europe, and etc., thermal insulation coatings have already been studied and used widespread [4 – 7]. Researchers in China also have done much and made big improvements in this area up to now [8 – 12]. However, most research has focused on one or two thermal insulation mechanisms in tandem. As the thermal transfer of the object is always a combination of heat conduction, convection and radiation. Excellent thermal insulation coatings should incorporate more than one mechanism of thermal insulation. In fact, multi-thermal insulation coatings that combine thermal barrier, echo and radiation are a major goal, and high property multi-thermal insulation coatings should be investigated to satisfy the insufficiency of the energy-saving material market.

Here, we prepared a novel water-based acrylic multi-thermal insulation coating (multi-WATIC). First, diatomite, hollow glass beads and powder of antimony tin oxide (ATO, nanometer) were used separately to obtain barrier, echo, and radiation thermal insulation coatings based on water-based acrylic emulsion. Thereafter, how these three thermal insulation functional fillers affect the thermal insulation property of WATIC was studied. Based on single factor experiments, a new technology compound with the three thermal insulation functional fillers was developed. Meanwhile, a novel water-based acrylic multi-thermal insulation coating (multi-WATIC) was prepared for the first time, and the formula of multi-WATIC was optimized by RSM. Test results show that the highest temperature difference of the novel multi-WATIC (16.4 °C) is much higher than single thermal mechanical of WATIC (barrier is 3.9 °C, echo is 3.8 °C and radiation is 12.5 °C). This result is similar to the value of 16.8 °C predicated by response to a surface methodology model.

2. EXPERIMENTAL DETAILS

2.1. Materials

To prepare the WATIC, a waterborne acrylic emulsion was provided by Guangzhou RongDong chemical co., LTD., China. This was used as the matrix resin, the composition of the waterborne acrylic emulsion is a mixture of methyl methacrylate, 2-ethylhexyl acrylate, methyl acrylic acid, acrylamide and styrene. And the solid content of the emulsion is 47.5 % (wt.%), the pH value of the emulsion is 8.7, the viscosity of the emulsion is 300 cps and the acid value of the emulsion is 25 mg KOH/g.

To prepare the WATIC, thermal insulation functional fillers is the most important ingredients, in this study, diatomite, hollow glass beads and ATO nanometer powder were selected and purchased from Foshan AnYang Chemical Co., LTD., China. The main chemical composition of diatomite is SiO_2(≥ 86 %, wt.%). Due to its special porous structure, diatomite can barrier thermal conduction effectively. The particle size of diatomaceous is 20 – 25 μm. Hollow glass beads is a kind of super lightweight inorganic materials powder. It has a hollow sphere structure, can reflect most of the energy from visible light, it usually used as echo thermal insulation functional filler. The particle size of diatomaceous is 30 – 80 μm. The main chemical composition of diatomite is SiO_2, Al_2O_3, ZrO_2, MgO. ATO nanometer powder is a mixture of SnO_2 and Sb_2O_3, present as dark blue, the mol ratio of SnO_2 to Sb_2O_3 is 9:1. The particle size of ATO is 20 – 80 nm. It can prevent the infrared and ultraviolet radiation effectively.

Mica powder and titanium dioxide were used as pigments and fillers. They were provided by Tianjing Fuchen Chemical Technology Co., Ltd., China. Mica

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powder is a kind of white powder with scaly structure, the diameter to thickness ratio higher than 70. The main chemical composition of mica powder is SiO_2 (49 %, wt.%) and Al_2O_3 (30 %, wt.%), and the particle size of mica powder is 5 – 10 μm. Titanium dioxide is the excellent fillers in coatings industry. The main chemical composition of titanium dioxide is TiO_2 (93 %, wt.%), and the particle size of titanium dioxide is 30 – 50 μm.

Necessarily additives (such as dispersant, defoamer and film formers) were necessary to improve the coating quality, provided by Tianjing Damao Chemical Technology Co., Ltd., China.

2.2. Coating preparation

To obtain the WATIC, waterborne acrylic emulsion, thermal insulation functional fillers, solvents, and additives were prepared according to Table 1. The preparation process is shown in Fig. 1.

Table 1. Formula for thermal insulation coatings based on acrylic emulsion

Materials		Contents, wt.%
Resin	Waterborne Acrylic Emulsion	35 – 70
Thermal insulation functional fillers	Diatomite	0 – 25
	Hollow glass beads	
	ATO	
Additives	Dispersant(KH570)	0 – 5
	Defoamer(PDMS)	0 – 3
	Film formers(CS-12)	0 – 2
Pigments and fillers	Mica powder	0 – 25
	Titanium dioxide	0 – 30
Medium	Distilled Water	20 – 45

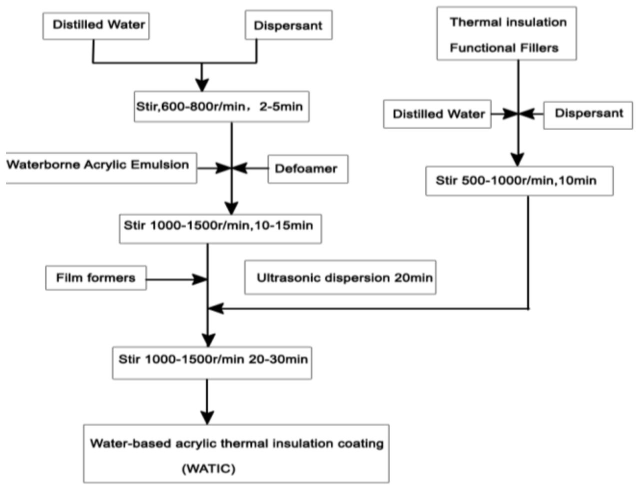


Fig. 1. Process flowchart of WATIC preparation

2.3. Temperature difference analysis

The test panel covered with WATIC was applied on a 70 × 40 × 2 mm aluminium panel (cleaned and dried). The thickness of dry coating was approximately 0.4 mm. Meanwhile, ordinary water-based acrylic coating without thermal insulation functional fillers was also applied onto the same size aluminium panel and dried for blank test. The thermal insulation property of WATIC was evaluated by temperature difference through a self-developed lamp illumination apparatus according to the Chinese “building insulation coatings” standard (JG/T 235-2008) [9, 13 – 15]. The schematic of self-made device was shown in Fig. 2. The apparatus consists of one iodine tungsten lamp (200 W) fixed on the top of ceilings, an aluminium tested panel mentioned above and an expandable polystyrene (xPS) box with two separate chambers, each chamber contained two thermocouple probes. One thermocouple probe was used to measure the surface temperatures of the test panel (T_s), and the other one was used to measure the inside environment temperature of bottom chamber (T_i). When the panel was heated to a temperature balance, the relative temperature difference between the two thermocouple probes is recorded as ΔT ($\Delta T = T_s - T_i$). To ensure the accuracy of test, blank test was need at the same time, one panel painted with WATIC and the other panel painted with non-insulated ordinary water-based acrylic coating were tested at the same time. In order to eliminate the influence of other factor, obtain the exactly thermal insulation property of the functional fillers, sample and blank sample was tested in the same environment with the same test conditions. As mentioned above, after heating to temperature balance, the temperature on the sample surface is higher than temperature on bottom chamber, the temperature difference between the above two is recorded as ΔT. To differentiate, ΔT_t represents the temperature difference for WATIC and ΔT_o for ordinary water-based acrylic coating (without any thermal insulation functional fillers) respectively. Therefore, the absolute temperature difference which represents the precise thermal insulation property of the test sample is recorded as T_d ($T_d = \Delta T_t - \Delta T_o$). In general, higher T_d represent better thermal insulation property.

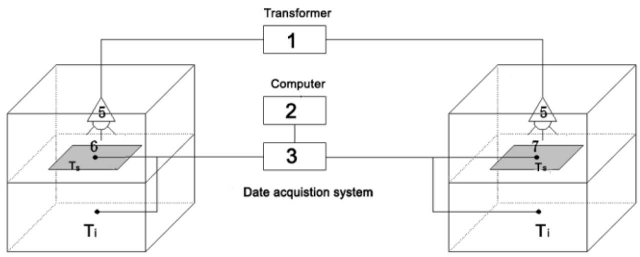


Fig. 2. Schematic illustration for the thermal insulation property measurements: 1 – transformer; 2 – data acquisition system; 3 – computer; 4 – xPS box; 5 – iodine tungsten lamp (200 W); 6 – panel painted with WATIC; 7 – panel painted with non-insulated ordinary water-based acrylic coating

3. RESULTS AND DISCUSSION

3.1. Single factor experiments of the three functional fillers

For the principle of a central composite design under response surface methodology, the factors and levels that effect the model should be confirmed based on single factor experiments first, then a series of experiments and analysis should be done according to a central composite design to obtain the optimal value of all the selected factors. Based on these, how the three thermal insulation functional fillers affect the thermal insulation property of WATIC was studied first in this study, using the principle of a central composite design under response surface methodology [16 – 17], the content and the category of the three different functional fillers were considered as significant factors to temperature difference of WATIC. So single factor experiment was studied in detail, barrier, echo and radiation thermal insulation coatings were prepared with certain thermal insulation functional fillers respectively. For barrier water-based acrylic thermal insulation coating means in this coating, the thermal insulation functional filler is just diatomite, echo water-based acrylic thermal insulation coating means in this coating, the thermal insulation functional filler is just hollow glass beads, radiation water-based acrylic thermal insulation coating means in this coating, the thermal insulation functional filler is just ATO nanometer powder. The temperature difference analysis was done to evaluate thermal insulation property of these 3 functional fillers. Variation rules of absolute temperature differences are shown in Fig. 3.

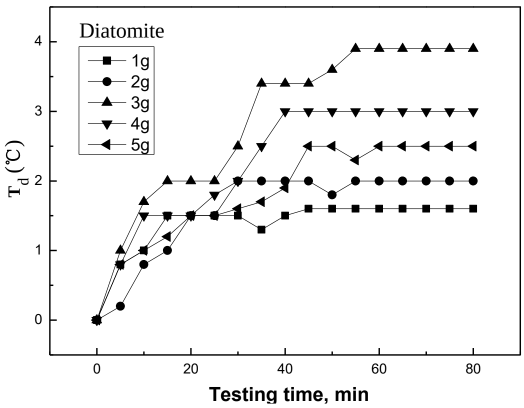
Fig. 3 a – c shows the rules of absolute temperature differences of WATIC with different weight contents of thermal insulation functional fillers. It clearly indicates that the T_d of WATIC contained any category of the 3 functional filler is higher than ordinary water-based acrylic coating without functional fillers. That means either functional filler (diatomite, hollow glass beads and ATO) has exceptional thermal insulation property.

Fig. 3 and Fig. 4 clearly show that for all the 3 functional fillers, curve of temperature difference (T_d) change with the weight content of the functional fillers, but the rules between the T_d and the weight content of either functional fillers are non-linear. For barrier thermal insulating coating, the T_d dropped after initially increasing. The highest T_d can be up to 3.9 °C when the weight content of diatomite is 3 g.

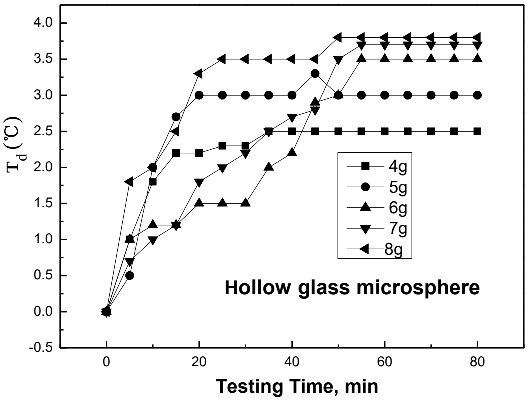
Hollow glass beads have low thermal conductivity due to its hollow structure, is used to prepare the echo thermal insulating coatings [9, 18, 19]. Test results show that the T_d of WATIC with hollow glass beads can be up to 3.8 °C when its weight content is 7 g.

When the weight content of ATO is 2.5 g, the T_d of the radiation WATIC can be up to 12.5 °C, much higher than the other two functional fillers. This probably due to ATO can absorb both visible and near infrared radiation, when used as thermal insulation functional fillers, it shows excellent thermal insulation performance [10, 20].

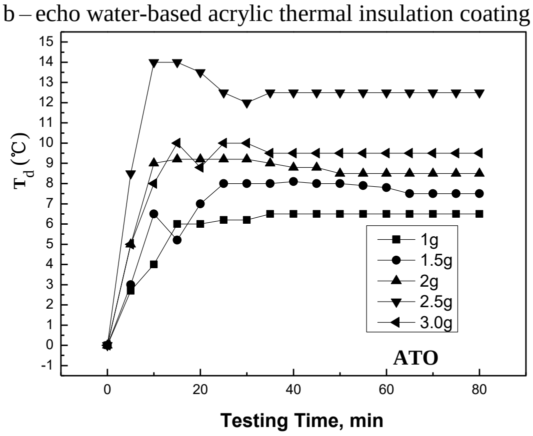
As mentioned above, temperature difference analysis results showed that the thermal insulation property of WATIC not only affected by the weight content of functional fillers, the category of functional fillers can also affect the thermal insulation property of WATIC at the same time. Test data indicate that, for all the 3 functional fillers, the temperature difference does not always increase while more functional fillers were added into coating system. There is a maximum T_d in the experiment range for each single functional filler as shown in Fig. 4.



a – barrier water-based acrylic thermal insulation coating



b – echo water-based acrylic thermal insulation coating



c – radiation water-based acrylic thermal insulation coating

Fig. 3. The absolute temperature differences of WATIC based on single insulation mechanism

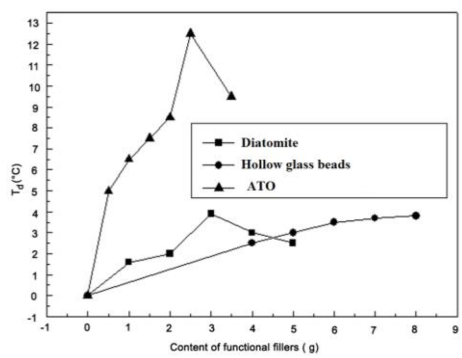


Fig. 4. Variation rules of T_d on the basis of category and weight content of thermal insulation functional fillers

3.2. Formula optimized by response surface methodology

According to the results of single factor experiments, the category and weight content of functional fillers were considered significant to the thermal insulation property of WATIC.

In order to obtain the optimal formula of multi-WATIC, all the 3 single factors were optimized by RSM through a full-factorial central composite design. Thereafter, a three-factor, three-level experiment design was adopted as shown in Table 2 and Table 3 taking the weight content and the category of functional fillers as the variations.

Table 2. Factors and levels in RSM design

Variables	Values			Code value
	– 1	0	+ 1	
Diatomite, g	1	2	3	*A*
Hollow glass beads, g	4	5	6	*B*
ATO powder, g	1.5	2.5	3.5	*C*

According to the rule of full-factorial central composite design, three-factor experimental variables – diatomite, hollow glass beads and ATO – in three-level required 17 experiments. For all these 17 experiments, the optimization of variables, sum of squares, degree of freedom, mean square, *F*-value, values of "$P > F$" and etc. were all determined by Design-Expert Software 8.0. The actual values of temperature difference for the variables are given in Table 4.

Correlating the thermal insulation property of WATIC with functional fillers is a basis for optimization the formula of multi-WATIC.

The variance on the absolute temperature difference of multi-WATIC, which was analysed by the Design Expert 8.0 software, is shown in Table 4. Based on these, a polynomial equation was set up as following:

$$Y = 9.00 - 0.19A + 0.13B - 0.063C + 0.62AB + 0.50AC - 0.13BC - 0.75A^2 - 0.38B^2 - 1.25C^2. \quad (1)$$

In the equation above, *Y* is the predicted response (T_d), meanwhile, *A*, *B*, *C* are represented the weight content of diatomite, hollow glass beads and ATO, respectively.

Table 3. The design of RSM experiments and the corresponding T_d of the WATIC samples

RUN	Variables/Levels			Response, T_d/°C
	A	*B*	*C*	
1	1	1	0	13.5
2	-1	0	-1	12.5
3	0	1	-1	12.0
4	0	0	0	16.5
5	1	0	1	12.5
6	1	0	-1	14.0
7	-1	1	0	15.5
8	-1	-1	0	14.5
9	-1	0	1	15.3
10	0	-1	-1	13.0
11	0	0	0	16.5
12	0	0	0	16.4
13	0	0	0	16.6
14	0	1	1	13.0
15	0	-1	1	15.5
16	0	0	0	16.8
17	1	-1	0	12.5

Usually, "$P > F$" value was used to validate the regression coefficient in RSM, if "$P > F$" value lower than 0.05 means model terms are excellent. In this case, as seen in Table 4, *A-A*, *B-B*, *AB*, A^2, B^2, C^2 are crucial model terms. They had a significant effect on the temperature difference of multi-WATIC. It is noteworthy that *F*-values farther from unity, this verified that the factors can explained the variations properly. That is, the estimated factor effects are real. Fig. 5 shows the interaction of the AB (diatomite and hollow glass beads), AC (diatomite and ATO) and BC (hollow glass beads and ATO) on the temperature difference of multi-WATIC.

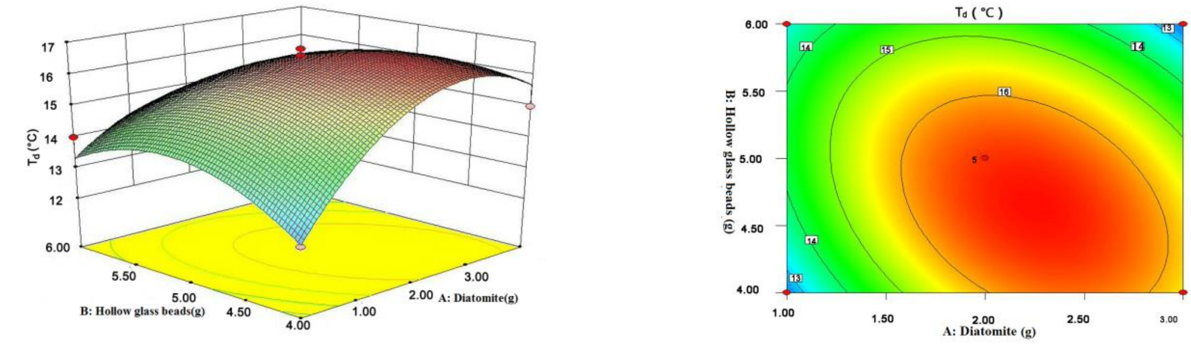
According to the polynomial equation, drawing the response surface plot showing the interactive effects of either two variables on the temperature difference of multi-WATIC while the third variable at it is fixed level.

Based on the single factor experiments of the three functional fillers (3 variables), the constant value of the third variable setted at fixed level "0". The plots are shown in Fig. 5.

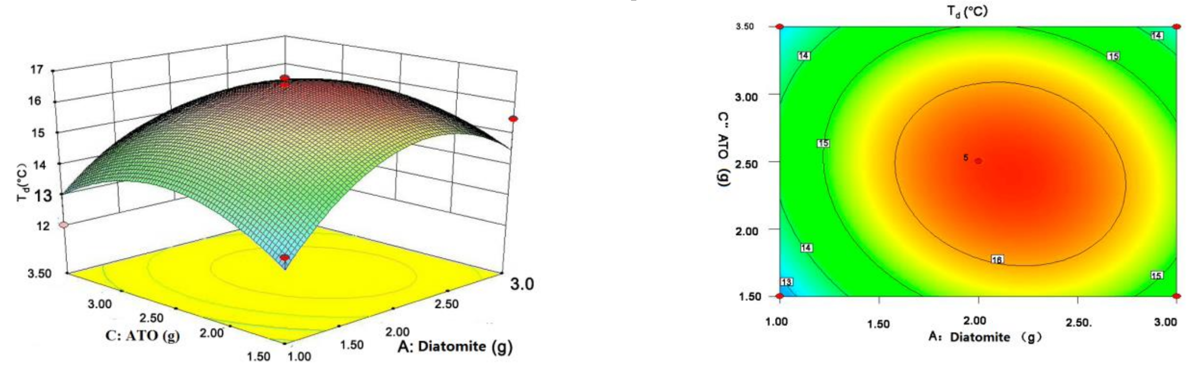
Fig. 5 a shows that when the weight content of ATO is constant, the interaction effects between the diatomite and the hollow glass beads on the thermal insulation property is significant.

Table 4. The analysis results of variance by central composite quadratic model

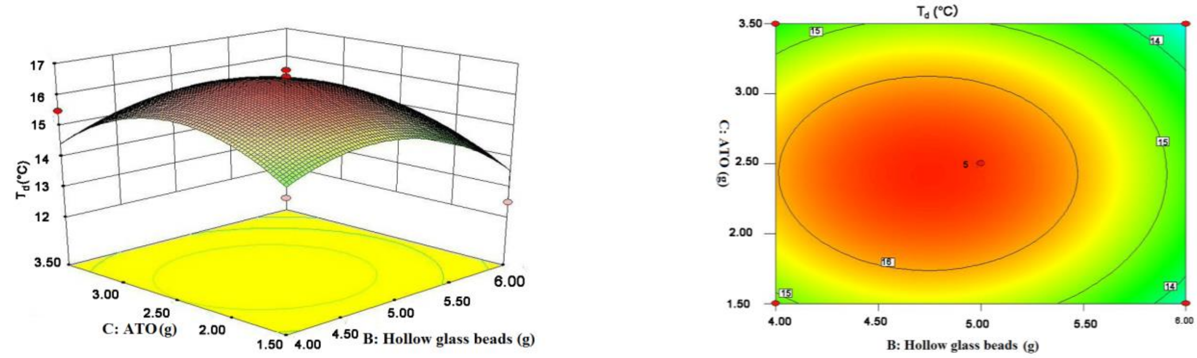
Source	Degree of freedom	Sum of squares	Mean square	*F* value	*P>F*
Model	9	41.78	4.64	5.62	0.00165
A-A	1	2.53	2.53	3.07	0.1234
B-B	1	3.13	3.13	3.78	0.0928
C-C	1	0.28	0.28	0.34	0.5778
AB	1	4.00	4.00	4.84	0.0636
AC	1	0.56	0.56	0.68	0.4364
BC	1	0.00	0.00	0.00	1.0000
A^2	1	14.29	14.29	17.31	0.0042
B^2	1	6.24	6.24	7.56	0.0285
C^2	1	7.59	7.59	9.19	0.0191
Residual	7	5.78	0.83		
Lack of Fit	3	5.69	1.90		
Pure Error	16	47.56			
Corrected Total	9	41.78	4.64	5.62	0.00165



a – the interaction of the AB on the temperature difference of multi-WATIC



b – the interaction of the AC on the temperature difference of multi-WATIC



c – the interaction of the BC on the temperature difference of multi-WATIC

Fig. 5. Analysis of variance based on full-factorial central composite quadratic model

The response value-- absolute temperature difference (T_d) rise with the content of diatomite and the hollow glass beads increases. When the content of diatomite and the hollow glass beads rise to a certain degree, the response value T_d reached the maximum value. Keep on increase the content of diatomite and the hollow glass beads, the value of T_d decrease. The results indicate that while the content of diatomite and the hollow glass beads is too low or too high would both depress the response value of T_d, only when they get to a moderate value, the thermal insulation property of multi-WATIC (T_d) can reached to a maximum value.

Fig. 5 b is the response surface plot showing the interactive effects of the content of diatomite and ATO on T_d of multi-WATIC when the weight content of hollow glass beads is constant. The response value – T_d rise with the content of diatomite and ATO increases until reached at the maximum value when the content of diatomite and ATO rise to a certain degree. Keep on increase the content of diatomite and ATO, the response value of T_d decrease. The plot indicates that while the content of diatomite and ATO is too low or too high would both depress the value of T_d, only when they get to a moderate value, the thermal insulation property of multi-WATIC (T_d) can reached to a maximum value.

Fig. 5 c shows that when the weight content of diatomite is constant, the interaction effects between the ATO and the hollow glass beads on the thermal insulation property. The response value – T_d rise with the content of ATO and the hollow glass beads increases. When the content of ATO and the hollow glass beads rise to a certain degree, the response value T_d reached the maximum value. Keep on increase the content of ATO and the hollow glass beads, the value of T_d decrease. The results indicate that while the content of ATO and the hollow glass beads is too low or too high would both depress the value of T_d, only when they get to a moderate value, the thermal insulation property of multi-WATIC (T_d) can reached to a maximum value. Meanwhile, the iso-response 3D surface graphic Fig. 5 (a) is steeper than Fig.5 (b) and Fig. 5 (c). This suggests that the interaction between diatomite and the hollow glass beads has the greatest influence on the thermal insulation property of the multi-WATIC.

Through moving both the major and minor axis along of the contour line to determin the statistical optimal values (T_d), the maximum T_d can be obtained when the response arrived at the center point. Thereafter, from the 3D plots shown in Fig. 5, the optimal concentration values for *A*, *B*, and *C* (diatomite, hollow glass beads and ATO) were identified as 2.3, 4.6 and 2.4 g, respectively. Thus, the formula of the multi-WATIC optimized by response surface methodology is as seen in Table 5. The maximum T_d can be up to 16.8 °C as predicated by the model. This is much higher than the T_d of multi-WATIC with just one single mechanism functional fillers (*A*, *B*, or *C*) as seen in Fig. 3 and Fig. 4.

Table 5. The formula of the composite multi-WATIC

Materials		Contents, wt.%
Resin	Waterborne acrylic emulsion	35 – 70
Thermal insulation functional fillers	Diatomite	2 – 5
	Hollow glass beads	5 – 8
	ATO	0.5 – 3
Additives	Dispersant(KH570)	1 – 5
	Defoamer(PDMS)	0.5 – 3
	Film formers(CS-12)	0.5 – 2
Pigments and fillers	Mica powder	0 – 25
	Titanium dioxide	0 – 30
Medium	Distilled water	20 – 45

3.3. Confirmatory test of the model

This study shows that the formula of a novel multi-WATIC system was optimized by response surface methodology. For the novel multi-WATIC, three different categories of functional fillers were contained. As a result, it can promote the collaborative thermal insulation effects including obstruction, radiation, and echo. The absolute temperature difference T_d could be up to 16.8 °C as predicated. Thus, to verify this prediction, a multi-WATIC is prepared according to the formula seen in Table 5. The T_d of the novel multi-WATIC was tested with the thermal insulation property measurements mentioned in Fig. 2. The result showed that the T_d of the multi-WATIC is 16.4 °C, which agrees well with the predicted result.

4. CONCLUSIONS

1. The three thermal insulation functional fillers: diatomite, hollow glass beads, and ATO nanometer powder, has significant effect the thermal insulation property of WATIC.
2. In the formula for a novel multi-WATIC optimized by the response surface methodology, the content of diatomite, hollow glass beads and ATO nanometer powder are 2.3 g, 4.6 g, and 2.4 g, respectively.
3. Temperature difference analysis showed that the T_d of the optimized multi-WATIC is 16.4 °C. This is much higher than the T_d of the WATIC with just one single mechanism functional filler. This means that the multi-WATIC has excellent thermal insulation property as expected.
4. For the absolute temperature difference of the optimized multi-WATIC, the predicted value of 16.8 °C agrees very well with the experimental value of 16.4 °C. That is, the response surface methodology is very effective in formula optimization.

Acknowledgments

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