Development of waste expanded polystyrene flexible coating material in concrete waterproofing

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Abstract. How to deal with the waste polystyrene and rubber has become a key research topic recently. This research attempted to process the waste rubber and waste polystyrene with chemical additives to produce the flexible coating material for concrete waterproofing. Based on the best guess on the configuration of the coating constituents by the grid-point method, the design experimental levels (3 levels) for different coating constituents could be decided by introducing the golden-section ratio. Then, the orthogonal experimental design method \( L_{27}(3^{13}) \) was adopted by choosing surface-drying time, viscosity, water permeability, tensile strength, pull-elongation rate as orthogonal indicators. Through orthogonal design method, the most influential coating constituents on different indicators were experimentally investigated. The results of the water permeability test showed that the water permeability of the coating was 0 mL and the water resistance was good. The analysis results of the orthogonal table showed that the material factors and levels affecting the tensile strength of the waste EPS flexible waterproof coating were: plasticizer (10.09%), polystyrene (17%), rubber powder (14%) and curing agent (5.1%), and the material factors and levels affecting the elongation of the waste EPS flexible waterproof coating are: plasticizer (2%), curing agent (5.1%), rubber powder (11.91%) and EPS (17%). This result can be used as a reference for adjusting the proportion of waste EPS flexible waterproof coating in the future.

1. Introduction
As known, the waste product without proper disposal will bring about the environmental pollution. For the past decades, the utilization of polystyrene and rubber as many artefacts promote the human civilization. However, with the increase of waste polystyrene and rubber, the way of burial or incineration wasn’t favoured; Instead, the physical or chemical modification of recycled polystyrene and rubber indicate a potential approach of civilization waste. In this paper, the chemical modification of waste expanded-polystyrene was developed with adding waste rubber for producing flexible coating material in concrete waterproof. In order to achieve this goal, grid-point method was adopted for obtaining the best guess of the coating constituents. Then, for finding out the optimum combination of coating constituents effectively, the orthogonal experimental design method was introduced to judge the influential significance of the coating constituents on mechanical properties (e.g. surface-drying time, viscosity, water permeability, tensile strength, pull-elongation rate) of the coating material. Based on the observation of the range analysis and variance analysis, the optimum mix for the coating material would be suggested in conclusion. And further improvement for the present constituents of coatings for concrete waterproof was also discussed.

2. Literature review
At present, for the recovery and reuse of polystyrene, four methods such as biodegradation method, physical regeneration method, decomposition recovery method and chemical modification method have been developed. The conversion of expanded polystyrene (EPS) into biodegradable materials is one of the easiest and most straightforward treatment methods to control EPS environmental pollution problems from the source. Cascades of Canada [1] has developed a special additive TDPA which can accelerate the oxidative degradation of expanded polystyrene. The EPS added with this additive has no change in the performance, but it is irradiated by oxygen, heat, ultraviolet light or mechanical stress. It can be turned into a fine powder that can be absorbed by bacteria and other microorganisms. The oxidative degradation time is about 3 years. This technology has been applied to Cascades' Bioxo series of products. Researchers at the National University of Ireland and the University of Hamburg in Germany [2] have developed a two-way technique for converting waste polystyrene into biodegradable plastics, first by pyrolyzing polystyrene into styrene oil and then using Pseudomonas putida CA-3. Bacteria convert styrene oil to polyhydroxyalkyl ester (PHA) under a nitrogen atmosphere. PHA is a biodegradable material that can be used to further produce general-purpose thermoplastics. The waste polystyrene foam is directly pulverized and used as a lightweight aggregate of concrete, and is combined with cement to form a lightweight concrete material or an external wall insulation board, which has sound absorption and heat insulation effect [3]. Because EPS has poor compatibility with cement and poor dispersibility, it needs to be surface modified to enhance compatibility. Shin C, Chase G.G and Reneker D.H. [4,5] produced sub-micron polystyrene fibres of about 500 nm by electrospinning using EPS as a raw material with excellent filtration performance. The most studied EPS decomposition catalysts are solid acids, solid bases and transition metal oxides such as HMC241, HZSM25, cerium oxide, aluminum oxide, zinc chloride and the like. Among them, the solid catalyst is superior in effect, and can simultaneously increase the yield of the lysate and the selectivity of the styrene monomer [6]. The EPS is dissolved in a solvent (toluene, xylene, halogenated hydrocarbon, etc.), and an additive such as a plasticizer and a modifier can be added to form an adhesive, and then a pigment can be used to prepare a coating, which is currently studied. The focus is on formulation design and cost reduction. Imanaka, Kutsuwada and Shitegawara [7] made a low odor gelling agent by mixing waste EPS with lemon oil and alkane mixed solvent. Yoshinari and Masato [8] prepared microcapsules using waste paper fibres as the core material and waste EPS as the wrapping material. This research hopes to study the preparation of adhesives and coatings by chemical modification. It is known from the literature that the preparation of EPS waterproof coatings requires the addition of many chemical admixtures such as solvents, plasticizers, emulsifiers and the like. However, the ductility of waterproof coatings does not seem to be the focus of the literature discussion. But in general, this is an important demand performance in building waterproof coatings. Therefore, this research developed a new type of waste EPS flexible waterproof coating for considering the ductility of building waterproof coating.

3. Experimental program

3.1. Materials

In this research, waste EPS, waste rubber, mixed solvent, curing agent and plasticizer were chosen as the coating constituents of the target coating material. Here, for increasing the solubility of polystyrene, the mixed solvent were composed of methylbenzene, ethyl acetate as well as #200 solvent refined oil, in which methylbenzene can easily dissolve EPS, ethyl acetate is soluble in #200 solvent refined oil and the mixing compound of methylbenzene and ethyl acetate ease the toxicity of methylbenzene or ethyl acetate. And the selected curing agent is carboxyl polymer and di-n-butyl phthalate (DBP) used as plasticizer.

3.2. Mixing procedure

For producing the target coating material, the mixing procedure is illustrated as follows: a) to calculate the amount of the coating constituents by initial guess which was empirically decided by trial-error tests, as listed in Table 1; b) to add EPS and methylbenzene in agitator as A-agent for swelling EPS within methylbenzene; c) to prepare the pre-mixed solvent, which were composed of curing agent,
ethyl acetate and #200 solvent refined oil, at 76°C for 60 minutes; d) to mix A-agent with the prepared mixed solvent at 60°C for 30 minutes; e) afterward, to add plasticizer in the above mixed solvent at 60°C for 150 minutes; f) finally, to pour and stir the waste rubber powder in c)–ready solution uniformly.

3.3. Testing
In order to evaluate the performance of the coating material in concrete waterproofing, the experimental items including test of surface-drying time (CNS 10756), viscosity test (ASTM D4212), water permeability test (CNS 10757), test of tensile strength (CNS 3553-1996), test of pull-elongation rate (CNS 3553-1996) were carried out to obtain the surface-drying time, coefficient of kinematics viscosity, water permeability, tensile strength and pull-elongation rate individually.

Table 1. Grid-point design of target coating material.

| Initial Guess | Item                  | Weight(g) | Weight ratio(%) |
|---------------|-----------------------|-----------|-----------------|
|               | waste EPS             | 7.0       | 12              |
|               | waste rubber          | 8.2       | 14              |
|               | methylbenzene         | 15.84     | 26              |
|               | #200 solvent refined oil | 9.56   | 16              |
|               | ethyl acetate         | 15.02     | 25              |
|               | curing agent          | 3.28      | 5               |
|               | plasticizer           | 1.09      | 2               |
| Best Guess    | waste EPS             | 6.80      | 17.0            |
|               | waste rubber          | 5.28      | 13.2            |
|               | methylbenzene         | 14.15     | 35.4            |
|               | #200 solvent refined oil | 2.31    | 5.8             |
|               | ethyl acetate         | 6.60      | 16.6            |
|               | curing agent          | 2.04      | 5.1             |
|               | plasticizer           | 2.82      | 7.0             |

3.4. Grid-point method (formula experiment design) for best guess
In order to evaluate the influential significance of the coating constituents, the proper experimental upper/lower limits of experimental variables need setting. For starting from a proper guess of the initial combination of experimental guess on coating constituents, the grid-point method was introduced. Through this method, the regression of the dependent variable (or called performance) of the mixture can be reasonably built up, based on a small amount of mixture constituents. For this research, \{3,2\} condition index of the grid-point method was utilized for choosing surface-drying time, viscosity and tensile strength individually as performance. Here, \{3,2\} means that six test levels would be executed for 3 experimental variables. Then, the optimum mixing proportion can be found based on the regression and the constraint conditions of the mixture constituents for each performance. Through the preliminary test of the target coating material, the best guess by considering the result of tensile strength as dominant performance was also shown in Table 1.

3.5. Orthogonal experimental design method
For efficiently finding out the influential significance of the experimental variables, the orthogonal experiment design method (or called as Taguchi method) was introduced by selecting \(L_{27}(3^{13})\) orthogonal table for arranging the combination of experimental variables. Here \(L_n^{rxm}\) means \(L\) is the table code in Taguchi orthogonal tables; \(n\) is the rows(or amount of experiments); \(r\) is the number of the test levels and \(m\) is the columns(or number of variables). Then, based on the experimental results of the arranged combination tests, range analysis and ANOVA (Analysis of variance) would be executed to understand the influential significance of each variable. Here, range means the difference between maximum and minimum value of test results. By comparing the range values, the significant
of variables on performance would be judged. ANOVA is performed by F-test, based on the calculated quadratic sum of standard deviation, degrees of freedom and variance. Based on the planning of the experimental plan, the results of the formula experiment design method and the preliminary test indicated the main material factors affecting tensile strength and elongation are plasticizer, curing agent, rubber, EPS, etc. The lower limit and the medium value are inferred by the golden ratio method. Table 2 indicates the judging level of the significance. Table 3 shows the level values of the experimental variables, in which the upper/lower limits of mixing proportion were decided by adopting golden-section ratio after the best guess from grid-point method. As a result, there are a total of 27 sets of test proportions, each set of 3 test pieces, a total of 81 sets of test pieces for the water permeability test, tensile strength measurement test, pull-elongation rate measurement individually.

| Item                | Influential Significance       |
|---------------------|-------------------------------|
| $F > F_{a=0.01}$   | Highly significant            |
| $F_{a=0.05} < F < F_{a=0.01}$ | significant                   |
| $F_{a=0.1} < F < F_{a=0.05}$ | Fairly significant           |
| $F < F_{a=0.1}$    | Not significant               |

Table 3. Level values of experimental variables

| Variables    | Upper Limit (%) | Medium Value (%) | Lower Limit (%) |
|--------------|-----------------|------------------|-----------------|
| plasticizer  | 10.09           | 7.0              | 2.0             |
| curing agent | 5.54            | 5.1              | 4.38            |
| waste rubber | 14.0            | 13.2             | 11.91           |
| waste EPS    | 20.09           | 17.0             | 12.0            |

4. Result and discussion
The main objective of this research was to develop the flexible coating material in concrete waterproofing by utilizing waste EPS and waste rubber, i.e. to investigate the use of waste to replace the raw materials of traditional synthetic polymer waterproof coatings and to address physical properties and waterproof properties. Hence, the water permeability, the tensile strength and the pull-elongation rate were chosen as the performance indices of the target coating material.

![Figure 1](image)
4.1. Water permeability
From the observation of Figure 1(b), the water-repelling effect of the target coating material could be recognized. In addition, according to the test results following CNS 10757 (for water permeability), the water permeability for all the 27 tests indicated 0.

Table 4. Range analysis of experimental variables on tensile strength.

| Variables       | Range Ratio(%) |
|-----------------|----------------|
| Plasticizer     | 21.49          |
| plasticizer x EPS | 15.60        |
| EPS             | 13.41          |
| curing agent    | 6.50           |
| waste rubber    | 3.09           |

x: reciprocal action

Table 5. ANOVA of experimental variables on tensile strength.

| Variables       | S    | φ    | V    | F    | F_{α=0.01} | F_{α=0.05} | F_{α=0.1} | Sig. Dif. |
|-----------------|------|------|------|------|------------|------------|-----------|-----------|
| plasticizer     | 6.35 | 2    | 3.18 | 17.50| 6.93       | 3.89       | 2.81      | Highly sig|
| plasticizer x EPS | 3.94 | 4    | 0.98 | 5.42 | 5.41       | 3.26       | 2.48      | Highly sig|
| EPS             | 2.23 | 2    | 1.11 | 6.14 | 6.93       | 3.89       | 2.81      | Sig.      |
| curing agent    | 0.57 | 2    | 0.29 | 1.58 | 6.93       | 3.89       | 2.81      | Not sig.  |
| waste rubber    | 0.13 | 2    | 0.06 | 0.35 | 6.93       | 3.89       | 2.81      | Not sig.  |

S: quadratic sum of standard deviation; φ: degree of freedom; V: variance; x: reciprocal action

Figure 2. Influence of plasticizer on tensile strength.

4.2. Tensile strength
The result of range analysis for tensile strength is shown in Table 4. It indicates that the order of the influential significance for tensile strength is plasticizer, plasticizer x EPS, EPS, curing agent and rubber. Such findings agree with the result of ANOVA, as shown in Table 5. In Figure 2, it is realized...
that the tensile strength significantly increases from 0.61 MPa till 1.50 MPa as the amount of plasticizer increases from 2% to 7% while the tensile strength gradually increases to 1.73 MPa as the amount of plasticizer increases to 10.09%. This phenomenon might be attributed to anti-plasticization, which means the adding of plasticizer into polymer may sometimes decrease elastic modulus, tensile strength and increase the pull-elongation rate while it may sometimes also cause the curing of resin and enhance the tensile strength. As for the reciprocal action of EPS and plasticizer on tensile strength, from the observation on Figure 3, it shows influence for selection of EPS in the case of 2% plasticizer on tensile strength is unclear but the impact from the reciprocal action of EPS and plasticizer on the tensile strength becomes stronger with the increase of plasticizer. It may resort to that the bonding strength of the target coating material can be effectively reinforced under the co-existence of a certain amount of EPS and plasticizer while the amount of EPS over 17% will weaken the bonding strength of the target coating material due to the anti-plasticization. This phenomenon can also be observed in Figure 4. As a result, how to choose the proper combination of EPS and plasticizer is one of the key points for considering the need of tensile strength during the production of the target coating material.

**Figure 3.** Reciprocal action of plasticizer and EPS on tensile strength.

**Figure 4.** Influences of EPS on tensile strength.
4.3. Pull-elongation rate
The result of range analysis for pull-elongation rate is shown in Table 6. It indicates that the order of the influential significance for pull-elongation rate is plasticizer, plasticizer x curing agent, curing agent, rubber and EPS. Such findings agree with the result of ANOVA, as shown in Table 7. In Figure 5, the plasticizer is added from 2% to 7%, the pull-elongation rate decreases from 71.14% to 3.43% while the plasticizer is added to 10.09%, the elongation is 2.36%. Due to the low content of plasticizer, the viscosity is low, the volatilization rate of the mixed solvent is slow, and the reaction is relatively complete. However, the elongation rate is not improved to the specification standard due to insufficient plasticizer content. As for the plasticizer and curing agent on pull-elongation rate, from the observation on Figure 6, when the plasticizer is 2% interacting with the curing agent, the change in the elongation rate is the largest, the difference is 38.29%, but when the plasticizer is increased to 7% and 10.09%, the reciprocal action with the curing agent for the little change of the elongation rate, it is thought that when the plasticizer is added to 7% or more, the coating material has become brittle, so the elongation rate has little influence.

Table 6. Range analysis of experimental variables on pull-elongation rate.

| Variables             | Range Ratio(%) |
|-----------------------|----------------|
| plasticizer           | 45.18          |
| plasticizer x curing agent | 8.66          |
| curing agent          | 8.53           |
| waste rubber          | 3.85           |
| EPS                   | 2.45           |

x : reciprocal action

Table 7. ANOVA of experimental variables on pull-elongation rate.

| Variables             | S    | \( \Phi \) | V | F  | \( F_{\alpha=0.01} \) | \( F_{\alpha=0.05} \) | \( F_{\alpha=0.1} \) | Sig. Dif. |
|-----------------------|------|------------|---|----|----------------------|----------------------|----------------------|------------|
| plasticizer           | 27948.193 | 2 | 13974.096 | 256.79 | 18.00 | 6.94 | 4.32 | Highly sig |
| plasticizer x curing agent | 1562.176 | 4 | 390.544 | 7.18  | 15.98 | 6.39 | 4.11 | Sig.          |
| curing agent          | 794.279  | 2 | 397.140  | 7.30  | 18.00 | 6.94 | 4.32 | Sig.          |
| waste rubber          | 155.057  | 2 | 77.529   | 1.42  | 18.00 | 6.94 | 4.32 | Not sig.     |
| EPS                   | 62.618   | 2 | 31.309   | 0.58  | 18.00 | 6.94 | 4.32 | Not sig.     |

S: quadratic sum of standard deviation; \( \Phi \):degree of freedom; V:variance; x: reciprocal action

4.4. Role of waste rubber
The motivation of this research is to develop the flexible coating material in concrete waterproofing by waste EPS and waste rubber. Through the above discussion, the role of EPS was clarified for its relevancy of the tensile strength. Desirable tensile strength can be achieved if over-or-less adding of EPS can be exempted as shown in Figure 4. However, how about the role of rubber in this research? According to the experimental observation of Figure 7 for the influences of rubber on tensile strength, it is clear to say there is no significance of rubber adding, from the viewpoint of chemical reaction. But, why is the reason for this unexpected result? If this is true, it shows there will be no feasibility for adding waste rubber in producing flexible coating material according to our proposed approach. Based on our discussion, from the viewpoint of physical participation, this deviation of experiment results may be solved by diminishing the particle size of waste rubber. In order to effectively be participated in the reaction of the target coating material for rubber in this research, the size of rubber should be...
small enough to pass through the interlayer pores, as schematized in Figure 8. If it can be done, the role of the rubber in the coating material will be 3-dimension rather than zero-dimension material, which means the rubber can offer its function for either pulling or breaking. In fact, by taking SEM photo of the target coating material as shown in Figure 9, the distribution of rubber within the resin shows it is insoluble in the present mixed solvent. It is worthwhile to consider how to promote the solubility of mixing solvent by proposing another kind of mixed solvent.

![Figure 5](image1.png)

**Figure 5.** Influences of plasticizer on pull-elongation rate.

![Figure 6](image2.png)

**Figure 6.** Reciprocal action of plasticizer and curing agent on pull-elongation rate.
5. Conclusion
Based on the above experimental discussions, several findings can be concluded as follows.
- The roles of EPS, plasticizer and curing agent on tensile strength and pull-elongation rate were experimentally observed; however, due to the complicated reciprocal action of each other, the
proper combination of them needs considerate care during the production of the target coating material. The decided desired mixing proportion for tensile strength can be referred to the result of the finding in this research.

- The result of tensile strength found in this research meets the requirement of the coating material of the specification in general; yet, the result of pull-elongation rate wasn’t so satisfactory partly due to the need of improvement of rubber particle size.

- In order to obtain the required particle size of waste rubber for the above request, the fine particle technology for rubber will be also the new research issue for producing the target coating material. In addition, for the time being, the long-term performance of the developed coating material wasn’t fulfilled. It will be also the new research issue for assuring the stable quality of the target coating material.

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