Optimization to Solidify Low-Level-Radioactive Resin Waste Via Taguchi Analysis

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ABSTRACT

Solidification of low-level-radioactive (LLW) resin was optimized via Taguchi analytical methodology. Samples that were selected according to Taguchi's method were separated into nine categories and their fabrication and development was gauged at the 7th, 21st, and 28th day after fabrication. Evaluations of the various samples focused on determined whether the compression and bending tolerances fulfilled the unique criteria of the Taiwan Power Company (TPC). Simulation results indicated that the best mixture of LLW mortar to solidify is furnace slag 30wt%, fly ash 26wt%, cement 9wt%, mixed with 13wt% of compound resin and with 22wt% of water. This recommendation of ingredients for LLW mortar was also verified with two other combinations.

Keywords: Taguchi analysis, solidification, resin, furnace slag

利用田口式最佳化分析法評量樹脂型低階核廢料固化配方

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摘要

本研究乃是利用田口式最佳化分析法評量低階核廢料固化之最佳配方，其中共有九組的核廢料試塊按照田口式最佳化的建議，而將其中組成的各成份做各種不同重量配比的變化，以求得最高的抗壓應力與抗彎應力的乘積值；評量結果顯示以百分之三十的爐石、百分之二十六的飛灰、百分之九的波特蘭二型水泥與百分之十三低階核廢料樹脂與百分之二十二的水混合，可獲得的最大的乘積值，此值並遠大於臺灣電力公司所建議之低限值。

關鍵字:田口式分析法、固化、樹脂、爐石

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Ⅰ. INTRODUCTION

Previous researchers have revealed the dominant contribution of furnace slag and fly ash in solidifying the low-level-radiowaste (LLW) resin with cement. Many components for LLW solidification have also proposed However, owing to the lack of precise investigation on these specific ingredients, the Taiwan Power Company (TPC) has always adopted conservative suggestions. The unique sea transportation process as well as the caustic off shore waste repository site have forced the TPC to reconsider all the conventional methods of processing liquid LLW waste [1].

Liquid LLW is collected from floor drain collector tanks, waste neutralizers and concentrator tanks, detergent waste drainage tanks, coolant demineralization systems, feed water purification systems, as well as equipment drain systems. To minimize their gross volume, contaminated solutions are filtered through ion-exchange resin. To improve the durability, the ion-exchange resin, which is discarded following dehydration, is solidified with portland cement as well as other essential ingredients. The LLW mortar is then poured into 53 gal, 1.2 cm thick A212-B carbon steel drums, and deposited into a tentative inland storage site for future ocean transport.

In 1990, Radiowaste Administration of the Atomic Energy Council of Republic of China (AECROC) introduced nine evaluation criteria, which a 53 gal drum that contains LLW mortar must adhere to. Among these evaluations, compression tolerance of the LLW mortar is the most essential parameter for LLW solidification [2]. Thus, consecutive surveys to identify new solidifying ingredients to fulfill the updated criteria have consistently occupied a preferential priority for Taiwanese faculties in correlated fields. However, to clarify the quantitative evaluation of solidifying ingredients within revised LLW mortar, Taguchi analytical methodology is proposed herein. For each group of three ingredients, a total of 108 samples were categorized into nine groups of various concentrations. This method of sampling corresponds with Taguchi's suggestion [3,4] that judging compression and bending tolerances preferentially determines LLW solidification optimization. Moreover, to verify this optimization, three subsequent groups are designed, which also adhere to Taguchi's recommendation as well as some extended results are also presented.

Ⅱ. INGREDIENTS FOR LLW SOLIDIFICATOIN

To fulfill the rigorous requirements, which protect against high humidity and high alkali corrosion in semitropical Taiwan, TPC has attempted replace the mortar in LLW solidification with pozzolan materials, such as fly ash or furnace slag. Generally, pozzolan reduces both the heat from hydration and the initial strength, however it improves the resistance against sulphate attack and alkali-aggregate reactions significantly, thus increasing ultimate strength. A portland-pozzolan cement can have a moderate or low hydration heat or moderate sulphate resistance[5]. Thus, the optimal ingredients to solidify LLW mortar can be fly ash[6,7], furnace slag[8,9], protland cement II, and adopted resin[10], as listed in Table 1.

Table 1. Various ingredients in LLW solidification adopted by the TPC.

Ingredient	Resource	Special feature
Fly ash	by-product of coal combustion	reduce water in solidification reduce heat of hydration
Blast furnace slag	by-product of steel making	increase denseness reduce heat of hydration reduce crack during solidification
Cement	portland cement II	low hydration heat resistance to sulfate attack
Resin		
Dowex HCR-W2 (40%)	sulfonation of a styrene divinyl-benzene copolymer	strongly acidic cation exchange resin gel type resin
Dowex SBR-P/C (60%)	porous resin incorporated with basic resins	Strongly basic anion exchange resin low crosslinking and high water retention

Ⅲ. TAGUCHI ANALYTICAL METHODOLOGY

Taguchi method is a powerful tool in designing high quality systems. It provides an efficient, systematic approach that optimizes designs for superior performance and quality. Additionally, Taguchi parameter design can also optimize performance through establishing design parameters and reducing system performance sensitivity to variation source[11,12].

A typical application of the Taguchi's method includes three parameter designation steps. The first step clarifies the dominant parameters within more than five factors, which are mutually interactive. Typically, this step requires 18 groups of samples to approach the Taguchi's criteria. However, the second step, based on recommendations from the first step, revises dominant parameter concentrations. This is conducted to ensure the precise concentration there of, while retaining the characteristics of mixed factors. Thus, in the second step, fine-tuning of quantities for the various parameters is more vital than determining the dominant parameters of the sampling process. Moreover, in contrast to the 18 sample groups in the previous step, there are 9 sample groups in this step. The third step is the last step verifying the sample process evaluation, which produces a more accurate quantity of interested parameters than if only the first step recommendations were adhered to [3,13-21].

A. Orthogonal Arrays

In this study, three ingredients, fly ash, furnace slag, and resin, as declared previously as the three dominant parameters in LLW solidification[2,22], were mixed with cement as three distinct concentrations Thus, a total of 27 ($3\times3\times3$) combinations were formed. However, according to the Taguchi Method, the samples could be organized into 9 groups and continue to yield experimental results with the same confidence as if they were considered separately. Table 2 displays the arrangement of the samples into the 9 groups, according to the Taguchi Method. The numbers indicate the experimental layout or concentrations of the distinct factors.

Alternately, Table 3 displays the manipulation of samples, which followed Taguchi's suggestion exactly. As indicated in the table, the resin contains an acidic (40%) and a basic (60%) resin component. The total weight of each sample is 5500g, which satisfies the exhaustion for nine cylindroids and three prisms in the compression and bending tolerance tests, respectively.

Table 2. Standard orthogonal arrays of 9 different groups following Taguchi's suggestion. The numbers in each column indicate that the various concentrations or layouts considered for the specific factors A-C.

Sampling no.	Factor A	Factor B	Factor C
1	1	1	1
2	1	2	2
3	1	3	3
4	2	1	3
5	2	2	1
6	2	3	2
7	3	1	2
8	3	2	3
9	3	3	1

B. Analysis of Variance: ANOVA

The evaluated compression and bending tolerance values from each group were averaged and then reorganized as follows[23]:

$$\eta = -10\log[\frac{1}{n}\sum_{i=1}^{n} y_i^{-2}] \quad (1)$$

where η was the inspection index, defined as the signal to noise ratio (S/N unit:dB). A larger value of η was considered preferable in this work, since both compression and bending tolerance for LLW solidification was expected to be higher than the suggestions from the TPC for ocean transport off the coast of Taiwan. A higher η (S/N) value also indicated a superior combination for LLW solidification, since the major signal dominated the noise. y_i was the average compression or bending tolerance in each group, i, and n was the repeating number in each group, 3 in this work.

Define S_m , S_A , S_T and S_E as follows:

$$S_m = \frac{(\sum \eta_i)^2}{9} \quad (2)$$

$$S_A = \frac{\sum \eta_{Ai}^2}{N} - S_m \quad (3)$$

$$S_T = \sum \eta_i^2 - S_m \quad (4)$$

$$S_E = S_T - \sum S_A \quad (5)$$

where S_m was the average of the squares of the sums of the η s, and η i was the specific inspection index in group i. S_A was the sum of squares correlated to various ingredients A; the η Ai was the inspection index correlated only to the specific ingredient A; and N was the number of samples in each batch. The various kinds of A were furnace slag, fly ash, and resin.

The corresponding number, N, was all three in this work. S_T was the sum of squares of the variance and S_E was the sum of squares of the errors correlated only to the specific ingredients A. Define F_{A0} as the index of F-test for checking the specific ingredient, A, and given as:

$$F_{A_0} = \frac{S_A / f_A}{S_E} \quad (6)$$

where f_A was the degree of freedom for that specific ingredient, which values were all two for furnace slag, fly ash and cement. $\frac{S_A}{f_A}$ was also defined as the variance; V_A ; of the specific ingredient A. The F-test which was first proposed by Dr. Fisher[24] was an auxiliary tool for inspecting the dominant factors involved in interactions. The contribution of the specific ingredient dominated the LLW solidification if F_{A0} was approximately equal to or greater than the index $F_{0.05,n1,n2}$ ($n1, n2$ were the number of degree of freedom in the numerator and denominator, respectively)[25]. Thus, the larger the F_{A0} value , the more dominant the ingredient in LLW solidification.

Ⅳ. EXPERIMENTAL PROCESS

A. Sampling Preparation

Nine groups of samples each containing twelve individual specimens were prepared for testing. Table 3 lists the combination of ingredients in each group. The twelve specimens were fabricated simultaneously for consistency and, then divided into four batches for both the compression and the bending tests. To ascertain the repeatability as well as to minimize fluctuation, each batch included three specimens. The timing and configuration of each batch for the tests was as follows: (1) the first three batches were cylindroids ($\pi\times 2.5^2 \times 10\ cm^3$) and underwent compression testing at the 7th, 21st, and 28th days after following fabrication. (2) the remaining batches were prism ($5^2 \times 10\, cm^3$) and underwent a bending tested on the 28th day following fabrication.

B. Compression and Bending Tolerance Test

For the compression and bending tolerance tests, specimens from each batch were placed into a universal testing machine. Table 4 lists the measured tolerances and corresponding S/N ratios [cf. Eq. 1] from the final batch of each group. Notably, to ensure well-developed rigidity, the final batch of each group was held until the 28th day following fabrication. The multiplication of compression and bending tolerances was also an significant parameter in optimizing LLW solidification. There are rigorous criteria for ocean transport off Taiwan. Thus, if this study does indeed improve both compression and bending tolerance as is required, this value should be larger.

T able 3. Orthogonal arrays of various groups following Taguchi's suggestion. Each quantity is given in g.

sampling	water	Furnace slag	Fly ash	Portland cement II	resin	Total weight
1	1447	1447	1158	579	868	5500
2	1310	1310	1310	524	1048	5500
3	1196	1196	1435	478	1196	5500
4	1196	1674	957	478	1196	5500
5	1250	1750	1250	500	750	5500
6	1146	1604	1375	458	917	5500
7	1310	1571	1048	524	1048	5500
8	1196	1435	1196	478	1196	5500
9	1250	1500	1500	500	750	5500

Table 4. Compression and bending tolerance with the corresponding S/N ratio for each group of samples. Data were evaluated from the last batch of specimens (the 28th day after fabrication). The multiplication of the compression and bending tolerance was also listed.

Sampling no.	compression [1] tolerance (kg/cm^2)	S/N ratio (dB)	bending [2] tolerance (kg/cm^2)	S/N ratio (dB)	[1]×[2]	S/N ratio
1	31.3±2.4	29.9	16.4±2.1	24.3	512.4±75.2	54.2
2	26.9±3.8	28.6	14.4±1.8	23.2	386.3±74.1	51.7
3	21.3±6.0	26.6	14.3±1.6	23.1	303.0±91.7	49.6
4	26.2±3.9	28.4	11.6±0.4	21.3	304.5±46.0	49.7
5	20.2±1.2	26.1	9.1±0.6	19.2	184.7±15.8	45.3
6	48.1±6.4	33.6	13.1±0.7	22.4	631.8±89.4	56.0
7	27.6±0.9	28.8	7.8±1.7	17.8	213.6±47.2	46.6
8	38.4±0.6	31.7	9.0±1.6	19.1	346.1±63.1	50.8
9	46.4±5.3	33.3	11.4±3.0	21.1	528.4±153.2	54.5

Table 5 lists the correlated parameters for the dominant factors in LLW solidification obtained through both compression and bending tolerance. The $F_{0.05,n1,n2}$ is also cited in the last column for reference. Furthermore, Fig. 1 presents the mean S/N response graph for each case. As illustrated, the top, middle and bottom graph represent compression, bending tolerance and tolerance inspection of both, respectively. Since in all three cases, a larger expected tolerance is better, the best recommendation for solely a compression base, solely a bending base, and both compression and bending is level 3-3-1, 1-3-1, and 1-3-1 for furnace slag, fly ash and resin, respectively [cf. Tab. 2]

V. DATA INTERPRETATION AND EXPERIMENTAL RESULTS

Figure 1 depicts the best recommendation of LLW mortar for solidification as furnace slag 30wt%, fly ash 26wt%, cement 9wt%, mixed with 13wt% of compound resin and 22wt% of water. This partially adheres to Pan's previous recommendation[22]. That is, from 18 groups of various, LLW mortar combinations, he recommended an optimization as furnace slag 24wt%, fly ash 24wt%, and cement 24wt%, mixed with 10wt% of compound resin and 17wt% of water. In addition, according to his estimation the approximate compression and bending tolerance for such a mortar should exceed 167.3 Kg/cm^2 and 18.4 Kg/cm^2 , respectively[22]. However, to ascertain its superiority the best recommendation from this study still requires verification. Table 6 lists the three LLW mortar combinations that were mixed specifically to confirm our experimental findings. As displayed, Sampling no. 1 follows Taguchi's recommendation exactly, whereas the others are differ slightly in their concentration. Moreover, Table 7 also lists both compression and bending tolerances for these three various combinations. In addition, the last array lists the maximum values of each evaluation that were produced from measuring the nine original orthogonal arrays. As Table 7 clearly depicts, the best recommendation of ingredients attained the highest compression tolerance and the multiplication value, but only the second highest bending tolerance. The inspection index of this evaluation was attaining the highest multiplication value for compression and bending tolerance. Thus, although the Taguchi recommendation was maintained, the bending tolerance that it produced was not the highest one.

Table 5 Table of ANOVA and F-test for compression, bending and their product.

Ingredient	Compression[1]			Bending[2]			[1]×[2]			$F_{0.05,n1,n2}$
	S_A	V_A	F_{A0}	S_A	V_A	F_{A0}	S_A	V_A	F_{A0}	
Furnace slag	13.25	6.63	3.98	26.53	13.26	78.70	3.91	1.96	1.05	19.00
Fly ash	10.34	5.17	3.10	4.45	2.22	13.20	27.75	13.88	7.47	19.00
Resin	32.44	16.22	9.74	6.94	3.47	20.58	67.66	33.83	18.21	19.00

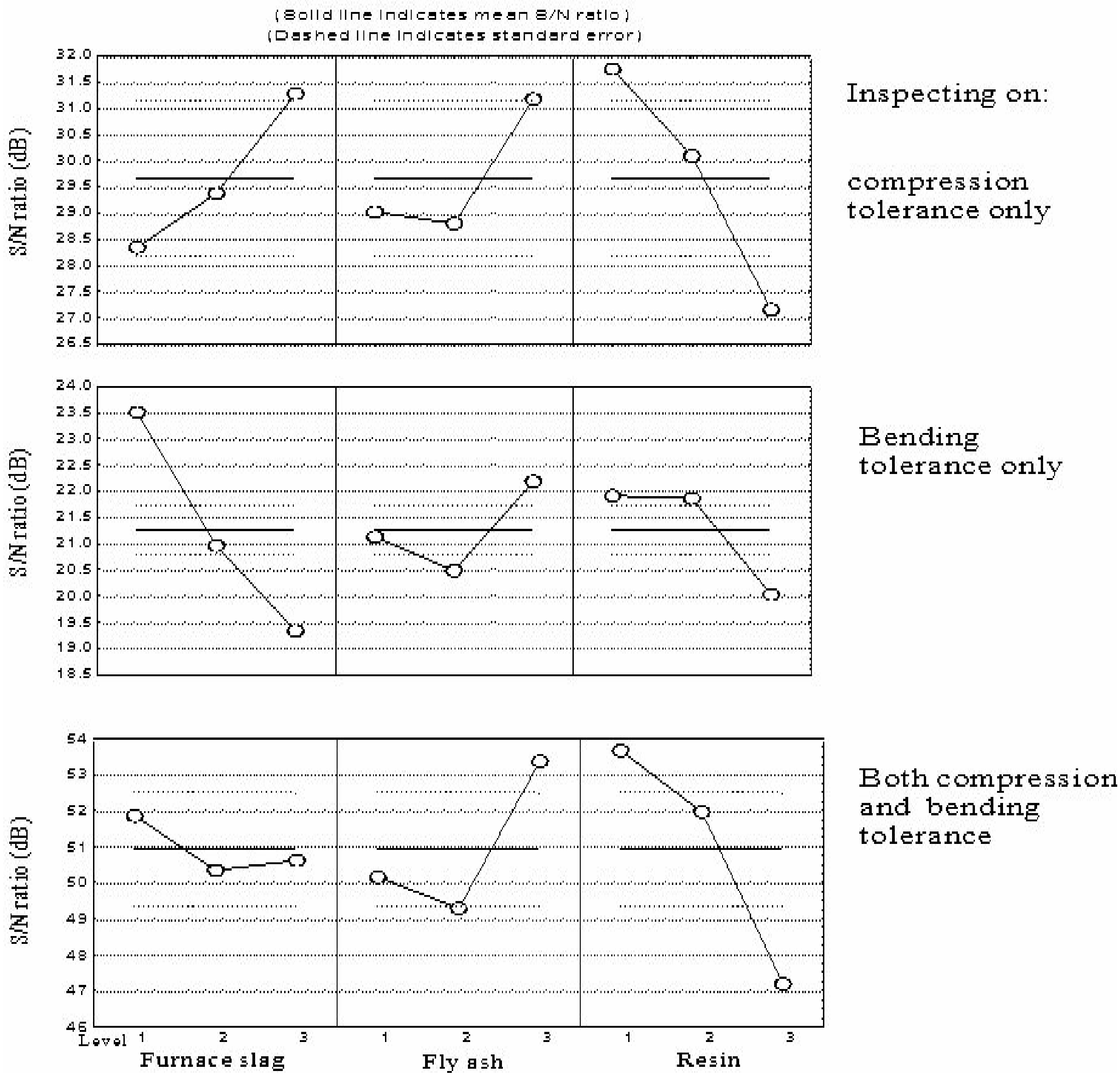

Figure 1 The mean S/N response graph for only compression, bending and both compression and bending tolerance.

Since this unique requirement was only proposed to fulfill rigorous criteria for ocean transport off the coast of Taiwan, the inspection index of Taguchi's analysis did not have to be only the compression and bending tolerance multiplication. The inspection index can be interpreted as either an evaluation judgment or a scheme destination. Thus, the index can be any quantified value. Notably, if the performance of LLW solidification is judged from an economical viewpoint, thus, as much resin as possible should be mixed into the mortar. Hence, to fulfill the requirement of this new criterion, the inspection index shall also be redefined as another quantified value. This new quantified value shall be focused only on compression tolerance and just barely above the minimum AECROC requirement, which is 15 Kg/cm^2 for underground repository. Since the destination of this new scheme is set as a fixed[2], neither too low nor too high the requirement of quantified inspection index is fulfilled this new criterion. Furthermore, an ideal definition of the inspection index implies the requirement in reality, whereas, an inaccurate setting might alter the trend dramatically in evaluation. Since both are equally essential in shipping, compression and bending tolerance multiplication, as proposed herein, can be a compromised setting. Nevertheless, LLW solidification quality relies on high compression or bending tolerance as well as other essential evaluations. Hence, a comprehensive study that includes all possible considerations is still required.

Table 6 Verification for Taguchi's best recommendation. Sampling no. 1 is exactly followed the Taguchi's recommendation in this work.

Sampling no.	water	Furnace slag	Fly ash	Portland cement II	resin	Total weight
1	1196	1674	1435	478	717	5500
2	1833	917	917	917	917	5500
3	1447	868	1447	868	868	5500

Table 7 Both compression tolerance, bending tolerance and their multiplication value for the three different combination and each the maximum value quoted from measuring the original nine orthogonal arrays are also listed.

Sampling no.	compression [1] tolerance (Kg/cm^2)	bending [2] tolerance (Kg/cm^2)	[1]×[2]
1	73.8±5.6	12.5±1.2	922.5±112.9
2	31.4±1.1	7.2±0.4	226.1±14.8
3	49.7±5.6	10.8±0.5	536.8±65.4
Max.	46.4±5.3	16.4±2.1	631.8±89.4

Ⅵ. SUMMARY

Optimization of low-level-radioactive resin solidification has been investigated herein via Taguchi's analytical method. To determine the best combination of mixed LLW mortar, various samples that followed Taguchi's suggestion were created. The rigorous criteria for future ocean transport off the coast of Taiwan require that LLW mortar attain both a higher compression tolerance and a higher bending tolerance. Thus, Taguchi's inspection index is defined as the highest multiplication value of both compression and bending tolerance. The best recommendation for LLW mortar solidification is furnace slag 30wt%, fly ash 26wt%, cement 9wt%, mixed with 13wt% of compound resin and 22wt% of water. The combination of ingredients in mortar might be modified slightly if the inspection index of Taguchi's is redefined to fulfill various requirements.

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