Solidification and Stabilization (S/S) of Toxic Waste: Potential Binder for Partial Cement Replacement Materials (PCRM)

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Abstract. Solidification and stabilization (S/S) in toxic waste treatment has been used widely in transforming waste into sustainable construction materials. However, some parties are just using this treatment method in modify the toxic waste into a non-toxic form for its suitability in landfill. This action should not occur as proper waste management, should include both ‘treat’; reduce toxicity (e.g: immobilize contaminants) and ‘control’; reduce amount of waste generation (e.g: recycle). The perspective of ‘dump is cheaper than recycle’ has closed the eyes of the parties on the mesmerizing effect of S/S method in develop a sustainable construction materials. Based on the past research, this happened due to the inability of the solidified waste to meet the construction materials requirements. Strength reduction and existence of toxic elements in leachate have been identified as the main factors that limit the application of solidified waste in construction industry. Nevertheless, the issue can be mitigate by using a proper binder to immobilize and encapsulate the toxic waste. Thus, this study aims to mitigate the issue by proposing a few binders that promotes high quality and sustainability to improve the S/S product. From the analysis, selection of binder relies on the chemical (e.g; high silica content) and physical (e.g; size) composition of the materials. Hence, pozzolanic materials have been chosen as usage of additional pozzolanic materials as partial cement replacement materials is promising in increase the strength of solidified waste and reduce its porosity. The pozzolanic materials mentioned in this study are industrial waste in approach to promote sustainability. Consequently, this study recommended the combination of pre-treatment such as thermal treatment, in reduce toxicity of toxic waste before S/S process to tackle the leachant or leachate issue and further adding pozzolanic materials for further strength development.

1. Introduction
Solidification and stabilization (S/S) method is one of the alternatives that has been used widely in treating toxic waste. Stabilization refer to physical and chemical adjustment of the toxic wastes to a stable form for environmental acceptance. Physical stabilization improves the properties of the waste such as the permeability, bearing capacity, and trafficability while chemical stabilization refer to the alteration of the toxic waste in eliminate or reduce the leach ability of the toxic contents [1]. This method involve the encapsulation of the toxic waste by mixing it with cementitious materials and efficient in immobilize the toxic contents in the waste such as heavy metals and hydrocarbons [2,3]. This treatment resulted in high reduction of toxic elements leachate and safe to be dispose into landfills [2]. Furthermore, this method is well known as cost-effective [2,3,4], reduce mobility of toxic contaminants [2], and its ability to recycle waste into valuable materials such as geopolymer composite [3], construction block [5,6], and pavement [7,8].
This technique has been used to solidify liquid oil waste from pumps or turbines by the US facilities [3]. Kualiti Alam, an integrated centre of toxic waste management in Malaysia, also applied this technique in immobilize toxic contaminant in waste before its disposal into landfill. Other than that, up to 50% of Coal Bottom Ash (CBA) is being recycled using S/S method in sound filling walls along highways and for sub paddings of roads in Germany [2]. While in Netherlands, 60% of the CBA have been utilized in sub-layer of roads and asphalt’s structure [2,9]. Moreover, over 72% of CBA was transformed into cycling tracks and parking lots by using S/S method [2].

Despite the high potential of S/S method in transforming waste into valuable materials, some party is only using this method to transform waste into a better waste for its suitability in landfill. This may be due to the limitation of this technique such as porous material structure and low in compressive strength. Beside the cost benefit, application of this method for waste recyle is very mesmerizing and further development of this method is highly recommended. Hence, this review is to explore the possible potential binder for PCRM and pre-treatment method to develop a better sustainable materials that can be apply in industry.

1.1 Cement as Basic Binder in S/S Method
Ordinary Portland cement (OPC) is the most common binder due to long term stability, low cost, good impact, and good strength development [5,10]. Cement particles react with water (hydration process) resulting in solidifies, insolubilizes, immobilizes, and encapsulates the toxic contaminant, forming a non-toxic solidified waste [11]. Calcium-Silicate-Hydrate (C-S-H) is the main product of cement hydration that contribute towards the strength development and entrapment of toxic element in the waste. S/S of toxic waste with cement can successfully immobilized to almost zero value of heavy metals even with low cement content [2]. Encapsulation of oil sludge in cement matrix show decrease concentration of all heavy metals content and immobilization of up to 98% for Cu, Zn, and Ni [12]. However, slightly reduction in compressive strength was reported affect by the existences of waste in the cement matrix [1,12]. This may due to the interference such as delay of the formation of hydration products [5], waste containing organic compound is said to be a factor that inhibit the binder hydration process [3], resulting in low compressive strength.

By using the binders that have high sorption of organic compounds can improve the immobilization of the toxic contaminants and prevent detrimental effects on binder hydration [13]. Moreover, addition of pozzolan as PCRM also can improves the performance of the solidified waste such as provide higher compressive strength than ordinary concrete [14,15], reduce porosity [5] and save cost [14,15,16,17]. Besides that, few pre-treatment method that suitable in develop high quality of solidified waste is also reviewed.

2. Improvement of S/S Method
Many previous work had proved that S/S method can successfully encapsulate toxic waste (e.g: petroleum sludge) with the addition of other binder such as pozzolan [3,5,12,18,19]. Inclusion of pre-treatment method before encapsulation process also found to be useful to enhance the effectenss of S/S method [12,15].

2.1 Possible Partial Cement Replacement Materials (PCRM)
Pozzolanic materials is defined as material or compound that contain 70% of SiO₂+Al₂O₃+Fe₂O₃ [15]. Natural pozzolans include crushed pumice or volcanic ash, diatomaceous earth, shale, and chert. Meanwhile, artificial pozzolans is produced by man-made and typically are wastes from the industrial application [20]. Either natural or artificial pozzolans can lower the production cost, gives a synergistic effect in cement matrix, and reduce the environmental effects [6]. More than that, pozzolans give better resistance to acid attack, lower the heat of hydration, increase durability of cement [6,21], promotes workability, and high resistance to cracking [15]. However, the use of natural pozzolans is usually high in cost and not sustainable to be compared to pozzolanic waste [20]. Hence, this study focus in suggesting artificial pozzolans that are suitable to be used as PCRM for improvement of S/S method
and increase sustainability. The pozzolans that will be discuss are fly ash, rice husk ash, sugarcane bagasse ash, and palm oil fuel ash.

2.1.1 Fly Ash (FA)
Fly ash is major solid waste generated from coal-firing that have been used as additional cement material or PCRM in construction works [22]. The major components of FA are calcium-related elements that can produce pozzolanic reactions and increase the clinker performance. Low amount of free lime (CaO) in FA can lead to structural crack of concrete when long term exposure to the atmospheric CO$_2$ and moisture. Therefore, direct deployment of FA in construction materials is not recommended due to the concerns on the issue [16]. Nevertheless, FA is the common pozzolan that being used for construction materials as it reduce the pore size of the cement matrix [6].

Addition of FA in S/S of waste with cement resulted in better hydraulic conductivity, greater in reduced the leaching of hydrocarbon, and higher compressive strength than samples with cement and waste only [18]. Although the presence of waste decreased the strength of the samples, addition of FA in the mix design will gives higher compressive strength to be compared with OPC alone [18]. Apart from that, based on the study by [4], 33% increase in compressive strength was observed with the addition of 20% of FA into a concrete containing 20% drill cutting waste. Nevertheless, more than 20% of FA show in the deduction of the compressive strength [4].

Instead of the addition of FA alone, the combination of FA with other pozzolanic materials such as silica fumes could produce a synergistic effect resulting in higher compressive strength and further improve the encapsulation process [4]. By replacing cement with FA is feasible, environmental friendly, and feasible [16]. Besides reduce the environmental effect, using FA as PCRM can promotes workability, reduces cracking at early ages, and improves strength of concrete [22]. In term of effectiveness, fine FA is recommended for high deduction of pore size and decrease of permeability [6].

2.1.2 Rice Husk Ash (RHA)
Rice husk is one of the agricultural by-products that contains high amount of non-crystalline silica or amorphous silica [6]. Rice husk represents 20% of the 649.7 million tons of rice produced annually worldwide [23]. The pozzolanic activity of RHA is effective in improving the strength of construction materials and the use of RHA in plain cement concrete leads to economy and cost savings [17]. Table 1 present the content of silica in RHA to be compared with several other of plant-based ash. As the consequences of high silica content, abundant presence, and the low cost of the bio-material, it is suitable to be used in concrete and mortar production. In fact, RHA is one of the good fillers in construction block due to its fine particle size [6].

Table 1. Ash and silica contents of various plants [14]
Optimum combustion temperature for obtaining highly reactive RHA is 600°C as it has the highest amount of silica, fineness particle, and high surface area [14]. RHA as PCRM can replace up to 20% weight of cement without a decrease in the strength while replacement of 10% lead to a significant increase in strength [23]. Addition of RHA in cement slurry can improved compressive strength, reducing cost, and can prevent disposal of waste that can lead to environmental problem [15].

As RHA can improves the strength of mortar, RHA is expected to be a good PCRM for S/S method since lesser pores and better immobilization could be achieved. Based on Yin et al [24], the combination of OPC with RHA in S/S of toxic waste fulfil the requirement of Unconfined Compressive Strength (UCS) and leach ability of metals of the product. Incorporation of RHA in the binder system reduced the leach ability of metal and the UCS results of the samples is sufficient to be used as construction materials [24]. S/S of petroleum sludge in cement matrix by using RHA as PCRM gives better compressive strength results than the solidified sludge with 0% of RHA [5]. Despite of compressive strength, addition of RHA also reduce the porosity of samples which make RHA as a promising materials for PCRM [5].

The use of RHA in cement matrix is also assists in solving the disposal problem of rice husk heaps in Asian countries [24]. Not only low in cost, RHA improves the durability of cement and increase the performance of concrete [14]. For the optimum effectiveness, increase the RHA fineness can increases its reactivity and efficient as a pozzolanic material [17].

2.1.3 Sugarcane Bagasse Ash (SCBA)

SCBA is one of the agriculture wastes generated by processing industry in produce sugar. Almost 1500 Million tons of sugarcane are produced worldwide and leaves around 40-45% of bagasse after juice removal which is a total of 600 Million tons of bulky waste [25]. Table 2 describe the composition of OPC vs SCBA. Even though the silica content in SCBA is not as much as in RHA, it is still considered as high and have pozzolanic property that make it suitable to be used as substitute of cement in construction material [17].

Table 2. Composition of OPC vs SCBA [26]

| Oxide | OPC | SCBA |
|-------|-----|------|
| SiO₂  | 20.44 | 77.25 |
| Al₂O₃ | 2.84  | 6.37  |
| Fe₂O₃ | 4.64  | 4.21  |
| CaO   | 67.73 | 4.05  |
| K₂O   | 0.26  | 2.34  |
| MgO   | 1.43  | 2.61  |
| Na₂O  | 0.52  | 1.38  |
| P₂O₅  | 0.10  | 0.59  |
| TiO₂  | 0.17  | 0.58  |
| MnO   | 0.16  | 0.27  |
| SO₃   | 2.20  | 0.11  |
| LOI   | 7.14  | 1.40  |
| SiO₂+Al₂O₃+Fe₂O₃ | - | 87.83 |

According to Asma et al [26], up to 30% of SCBA in concrete can increase the workability and compressive strength of the concrete, better than the sample with OPC alone. Other than that, SCBA enables pozzolanic reaction and improve the microstructure that lead to high early strength caused by its ultra-fine particle size [26]. Moreover, the use of super-plasticizer is not essentiial as incorporation of SCBA in concrete increase workability of fresh concrete and gives higher compressive strength as compared to the commercial concrete [25]. Despite it usage as PCRM, SCBA as sand replacement for mortar production also can develop a better mechanical properties of mortar [27] and the addition of SCBA in compressed earth block did not affect the mechanical strength and water absorption which proves the feasibility of SCBA as replacement of construction materials [28]. Other than that, the combination of SCBA and RHA can form lighter brick with good compressive strength [17].
In term of S/S application, few studies have emphasized the capability of SCBA in effectively immobilize heavy metals in cement matrix. It has been observed that the hydration characteristics including setting time, compressive strength and the porosity of SCBA blended cement were enhanced and can successfully immobilize heavy metals [29]. Moreover, Saiful et al. [30] indicated that 100% concentration of heavy metals had been successfully removed with the optimum mix design of 5% OPC and 15% SCBA. In conclusion, SCBA can be utilized as PCRM for S/S of toxic waste [29,30], production of building bricks [17], and as PCRM to improve the mechanical properties of construction materials [27]. The use of SCBA in increased the strength of samples, reduced the leach ability, and immobilize heavy metals, not only marks a novelty in the S/S but also reduce the amount of this bulky waste [29].

2.1.4 Palm Oil Fuel Ash (POFA)

Palm oil fuel ash (POFA) is a waste materials produced by the oil palm industry through the burning of fiber, palm oil shells (POS), and empty fruit bunches (EFB) until it can be collected as ash [31]. It is a pozzolanic materials due to its amorphous nature and high silica content [31]. Table 3 describe the composition of OPC vs POFA. Based on the table, it can be observed that the amount of silica in POFA is higher than OPC, but very low in CaO which is only 4.8% from OPC content. The silica contents in POFA promotes pozzolanic reaction during hydration; produce calcium aluminate hydrates (CAH) and CSH that increase the durability of samples [32]. Other than that, the high silica content in POFA enhance the chemical bond between heavy metal and binder, resulting in heavy metal immobilization [33].

Table 3. Chemical Composition of OPC vs POFA [34]

| Element | OPC | POFA |
|---------|-----|------|
| CaO     | 63.0| 3.0563 |
| SiO₂    | 20.0| 43.492|
| Al₂O₃   | 5.70| 3.188 |
| MgO     | 0.99| <LOD |
| Fe₂O₃   | 2.90| 11.413|
| SO₃     | 3.50| 0.0389|

Based on the study by Sooraj et al. [32], 20% replacement of cement by POFA can produce concrete with more than 30N/mm² of compressive strength while higher percentage of replacement caused deficiency in strength. Moreover, Noorafizah et al. [33] also obtained the same results which is the optimum percentage of POFA replacement is 20% for 30% of petroleum sludge S/S as it gives the best result in immobilize the heavy metals content from leach out of the mortar. However, greater than 20% of POFA replacement showed an unsatisfactory result which is increase concentration of heavy metal in leachate [33]. This may cause by the lack of hydration since higher percentage POFA lead to the low content of CaO in the mixture, resulting in the weak structural formation of sample [33]. Proper ratio of POFA will improve the durability of cement through pore refinement and reduce the formation of calcium hydroxide (Ca(OH)₂) that further improve the resistance corrosion of concrete [21].

The low profitable commercial value of POFA have been one of the reasons of its disposal into the landfills. However, the disposal of POFA is not only required high cost but, it also may give negative impact to the environment [21]. Since the efficiency of POFA as PCRM in S/S have been proved by previous researcher, alternatives in using POFA in construction materials have attract major interest due to its abundant accessibility and ability to reduce cost of disposal, space of landfill, and the usage of natural resources (e.g: cement).

3. Discussion and Recommendation for Development Research

The mesmerizing ability of pozzolans as PCRM in improve the S/S of toxic waste is proved by the previous researcher. Various type of pozzolans have been tested in study their effectiveness in S/S of
toxic waste. Based on the pozzolans list from the previous chapter, SCBA is recommended to be used as a PCRM in S/S of high organic compound waste (e.g: Petroleum sludge) for further research. This is because, previous research has proved the captivating performances of SCBA in immobilize heavy metals however, very less study mentioned its effectiveness in encapsulate toxic organic compound. All of the pozzolans mentioned in previous chapter is suitable to be used as PCRM in S/S process. Based on the previous research, each of the pozzolans have its own advantage, for example RHA provides better strength than FA but, FA is more resistance to corrosion attack to be compared to RHA. In choosing the PCRM for S/S process, each characteristic of the pozzolans need to be considered in obtain the desired quality of product.

3.2 Pre-Treatment of S/S Method

Despite of adding pozzolan as PCRM in S/S, it is observed that the application of pre-treatment of waste before S/S can also improve the effectiveness of the encapsulation process. This is because, some waste such as petroleum sludge contain a high number of organic compounds. The organic compound in the waste is said to be one of the reason that hinder the hydration process of cement; producing a high porous structure and low in compressive strength product [5]. Other than that, even at low concentrations, organic materials can produce a significant micro and macro structural changes to the properties of hydrated cement [3]. Hence, it is suggested to reduce the organic compounds in the oily waste to prevent the limitation of the S/S method. One of the pre-treatment that can effectively reduce organic compound in oily sludge is the thermal treatment. Common applied thermal treatment methods include incineration and pyrolysis that offer benefits in S/S by reduce the amount of toxic in the waste and further improves in properties of solidified waste [35]. Since many researches reported that only few amount of oil sludge is able to be encapsulate in the cement matrix, transforming the waste into ash form using incineration process as pre-treatment is highly recommended.

S/S of oil sludge ash is identified as more effective against metal release than oily sludge which shown that chromate is only detected in oil sludge sample and none from the oil sludge ash sample [12]. This proved that S/S method of toxic waste did reduced the amount of toxic leachate but, application of pre-treatment have modified the chemical compound of the waste, leading in better result gained. Apart from that, addition of oily sludge ash in cement matrix can contribute towards the formation of hydration product which is C-S-H that can further improves the strength of solidified waste. Furthermore, petroleum sludge ash contained the same substances as cement such as free lime, calcium oxide, and sulphur dioxide [36]. This is supported by Panova et al. [37] that stated that the main constituents of the mineral part of the oil sludge are quartz sand (63.1 %), albite (16.5 %), silicates (6.9 %), magnetite (6.4 %), muscovite (4.1 %), and kaolin (2.9 %). The content of the oil sludge ash investigated found to be highly suitable to be used in construction of building materials to be compared with the compositions of oily sludge that is high in toxic substances [37].

In addition, advantages of using pyrolysis process includes minimizes waste volume and its ability in yields valuable products [35]. Since the pyrolysis process can remove a high amount of organic content in oily sludge, this process is suitable as pre-treatment to enhance the effectiveness of S/S method. Other than that, pyrolysis can break down the high-chain organic matter into small molecular matter through three phase which are solid (inorganic minerals and residual carbon), liquid (fuel and water), and gas (CH₄ and H₂, etc.) [38]. Instead of S/S in cement, certain product of the pyrolysis of oily sludge have the asphalt-like emulsion or bitumen properties, make it suitable to be used for S/S in road or pavement [39]. Moreover, not only as pre-treatment, the application of pyrolysis as single treatment for toxic waste is as of interest since the residue fulfil the requirements for direct disposal into the landfill [40].

Moreover, despite of the pre- treatment for the waste, pre-treatment of the selected binder also can be applied in boosting the capability of the binder to bind the waste. For example, washing of RHA by using hot water as a pre-treatment improve the reactivity of the RHA resulting in better properties of cement slurry and compressive strength to be compared with regular RHA [15]. Other than that, the carbonation of fly ash before using as PCRM could develop the strength of cement mortars resulting in 20% higher than sample of cement and accelerating the strength development by up to 28% [16].
Moreover, FA contains a minor amount of free-CaO that can cause alkaline aggregate reaction resulting in PH reduction and further cause structural defects when deploying in construction materials. The carbonation of FA has turned out to increase the amount of CaO content, making it more effective as PCRM [16]. Application of pre-treatment not only produce better solidified waste but also can increase the percentage of encapsulated waste, hence, further reduce the abundance of the waste, avoid its disposal into the landfill, and prevent environment impact.

The result gained from the application of pre-treatment of toxic waste before encapsulation are highly fascinating. However, very few studies are done in combine both of the improvement to highly secure the place of solidified waste as a sustainable construction material. Hence, it is recommended to combine the addition of pozzolan as PCRM in solidify of pre-treated toxic waste for the future research development.

4. Conclusion
Consequently, this review was performed to mitigate the problem and advancement in capability of solidified waste as sustainable construction materials. These recommendations are expected to guide on a better alternative in improving solidification and stabilization (S/S) product by a proper selection of pre-treatment method and PCRM that promotes high quality and sustainability. The addition of pozzolan as PCRM and application of pre-treatment, have shown a fascinating result in develop better solidified waste. Application of both alternatives in the future research is recommended as it may secure the ability of solidified waste as sustainable construction materials.

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