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Optimising Factors for the Production of Amorphous Rice Husk Ash via Combustion Process for Sustainable Construction: A Review

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ABSTRACT

Cement is an unavoidable material in the construction industry, as it is an essential part of concrete. Moreover, its production causes serious issues related to the environment; hence, it creates a need to search for a sustainable alternative material that can be used in place of cement. On the other hand, the disposal of agricultural waste is also becoming a significant problem, and one of the materials is rice husk. Annually, about 160 million tonnes of rice husk, considered agro-waste, turn into ash due to a lack of commercial interest. Its dumping creates land pollution that poses a risk to human life. In order to tackle these problems, using agro-waste as a replacement for cement in construction can promote a green environment and make construction sustainable. Rice husk ash (RHA) is widespread owing to the large amount of silica present. However, impurities such as alkali metal oxides and uncontrolled combustion decrease the quality of silica and promote silica crystallisation because, at higher temperatures, the alkali metals, mainly potassium oxides, dissociate and act as a crystallisation catalyst. This paper briefly reviews the factors that can optimise the production of highly amorphous silica content in rice husk ash by the combustion process. It was concluded that the important parameters that can increase the pozzolanity of RHA are leaching conditions, combustion conditions, and grinding conditions. Furthermore, it has also been figured out that some areas, like understanding the reaction kinetics of RHA, an efficient leaching process, and suitable grinding conditions, need to be explored to increase the production of highly amorphous silica.

Keywords:
Cementitious Materials; Amorphous Silica; Acid Leaching Treatment; Rice Husk Ash

1. Introduction

The necessity of developing infrastructure is increased because of a growing population, which leads to more utilisation of cement. Cement is considered an essential component of concrete production, but simultaneously, it emits greenhouse gases (CO_2) during its manufacturing, which pollutes the environment adversely [1]. Approximately 5–8% of greenhouse gases, such as CO_2, are released into the air by the cement industry [2]. Cement production is the third biggest generator of

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carbon dioxide (CO_2)[3]. As a result, researchers started working on incorporating eco-friendly construction materials to reduce the environmental pollution caused by the cement industry [4]. In addition, the quick enactment of biomass power plants gives rise to the problem of disposing of agro-waste ashes [5]. Almost half of the world's population uses rice as their fundamental food, and over 90% of the rice is used in Asian countries [6]. Around the globe, rice production is nearly 750 million tonnes per year, out of which approximately 160 million tonnes are transformed into rice husks, creating environmental pollution [7]. In some regions, rice husk (RH) is used as fuel for electricity production and steam generators [8]. Therefore, to minimise the deleterious impacts on the environment on account of agricultural waste and the cement industry, the study of different alternative materials by using agricultural waste as a cement replacement is encouraged.

Regarding agricultural waste, rice husk is believed to be a tremendous supplementary cementitious material (SCM) [9]. Identical binding compared to cement and good pozzolanic reactivity are the essential characteristics of supplementary cementitious materials (SCM) [10]. Adding rice husk ash to concrete as a supplementary cementitious material makes concrete more resistant to chemical reactions and reduces porosity [11]. Moreover, according to ASTM C618, the RHA falls under the N-type pozzolan category. The carbon content in RHA shall not exceed 10%, whereas at least 70% of aluminium oxide, iron oxide, and silicon dioxide should be used in concrete as pozzolan [12]. Implementing rice husk ash (RHA) as a substitute for cement can provide better strength and durability and keep down the problem of agro-waste disposal and greenhouse gas emissions [13]. In spite of having a good capability of being used as cement replacement material, RHA is not widely accepted in construction industries due to the non-existence of specific mixing procedures [14], however, this may happen due to the variation in the properties of RHA because of several parameters that influence the production process. Now, to encourage the sustainable construction by using RHA, it is important to optimise its manufacturing process. Hence, the objective of this article is to highlight the influence of important parameters like acid leaching treatment, combustion temperature and time, leaching conditions, and grinding conditions on the production of functionalized amorphous silica from rice husk for sustainable construction.

2. Rice Husk Ash

After the rice milling process, a considerable amount of waste is generated, called rice husk. This rice husk has now become a source of fuel for industries. Primarily, rice husk consists of 75% to 90% organic matter, which includes cellulose and lignin, whereas inorganic matter constitutes 10% to 25% [15]. About 20% to 25% of rice husk ash can be obtained after the complete incineration of the rice husk [16]. After the complete combustion of rice husk (RH), a quantity of around 85%–95% silica is obtained, which is in an amorphous state [17]. Rice husk ash (RHA) shows pozzolanic behaviour due to the presence of amorphous silica in greater quantity [18]. Owing to its large specific area and enough amorphous content, RHA is considered one of the suitable pozzolanic materials that can replace cement effectively [19]. Since the permeability of concrete reduces because of the pozzolanic behaviour of rice husk ash, it helps to prolong the durability of concrete [13]. However, the uncontrolled incineration of RH for an extended period affects the pozzolanic behaviour of rice husk ash (RHA) [20]. Metal oxides, such as K_2O, behave as crystallisation catalysts because they melt on the outer layer of RH and make silica non-reactive [21]. Although, by optimising the combustion process and incineration temperature, the binding properties of RHA can be improved [22]. Generally, after cultivation of the rice plant, it goes through the milling process, which separates the rice husk from the rice as waste. Furthermore, this husk is mostly burned in different industries as a source of fuel under uncontrolled burning conditions, which subsequently results in the formation of rice husk ash. Moreover, for an effective use of rice husk ash, it is combusted in a controlled environment. The schematic diagram of the production of rice husk ash via combustion is depicted below in Figure 1 [23].

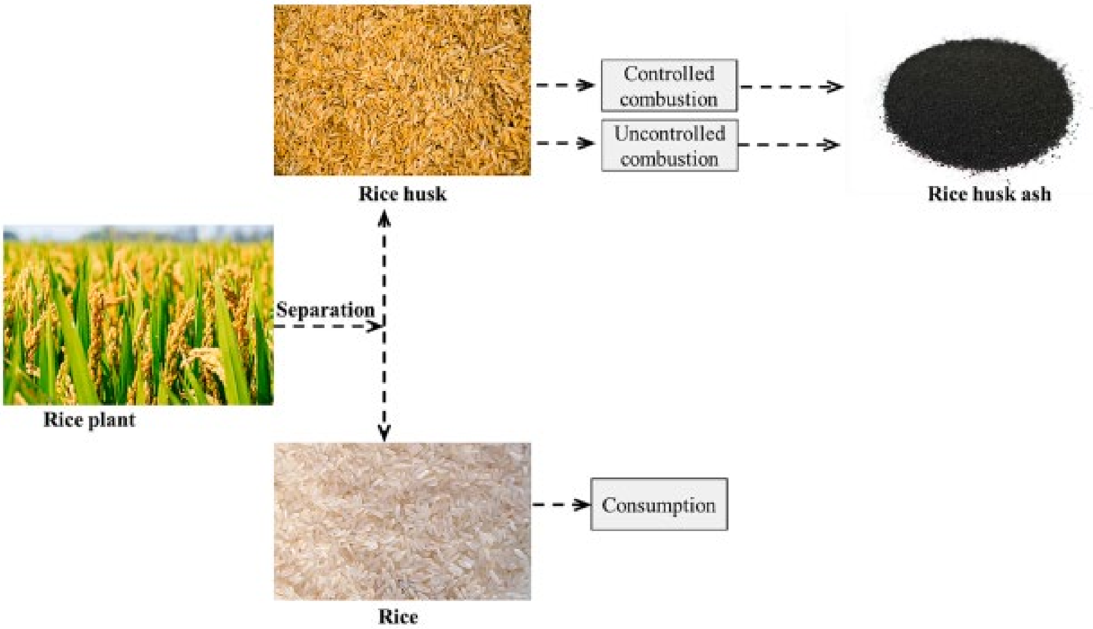

Fig. 1. Schematic Diagram of Rice Husk Ash Production [23]

2.1 Physical Properties

The physical properties of rice husk ash vary with different conditions of burning and grinding. Depending on the amount of unburnt carbon, the color of rice husk ash can be white, black, or grey. Figure 2 below illustrates different colours of rice husk ash produced by burning non-treated husk and leaching with different concentrations of HCl (0.01M, 0.1 M, and 1M, respectively) before burning at 800^0C for 2 hours [24]. Moreover, the separation of the organic content of rice husk during the incineration process is the reason for the porous nature of rice husk ash [25]. In general, rice husk ash has a density of about 180–200 kg/m^3, which is evident by its high porosity, lightweight, and fine nature [26]. Furthermore, the pozzolanic behaviour of rice husk ash is related to the surface area, fineness, and amorphous silica, and these parameters can be controlled by suitable burning and grinding conditions to implement in structural concrete [27].

Fig. 2. Different colors of Rice Husk Ash [24]

2.2 Chemical Properties

The chemical components in rice husk ash depend on geographical, geological, and atmospheric conditions [28]. However, rice husk ash generally contains deleterious alkaline earth metals in the form of oxides, mostly K_2O and silica dioxide. The chemical composition of rice husk ash reported by various researchers is highlighted in Table 1.

Table 1
Chemical components of RHA reported by different authors

References	LOI	SiO_2	K_2O	Na_2O	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	P_2O_5	TiO_2
[14]	-	72.81	5.71	0.22	0.56	0.78	2.33	3.77	0.59	1.279	0.076
[29]	-	76.8	1.82	-	-	-	2.6	0.10	-	-	-
[30]	0.53	89.85	1.38	-	4.79	0.86	0.79	-	1.34	0.22	-
[31]	4.81	96.7	0.91	0.16	1.01	0.05	0.49	0.19	-	-	-
[32]	-	96.23	0.45	0.054	0.281	1.366	0.57	0.269	0.202	0.361	-
[33]	3.90	90	4.60	-	0.46	0.43	1.10	0.77	-	2.43	-
[34]	1.39	91.45	1.39	0.11	0.44	0.18	0.99	0.36	0.04	-	-
[35]	-	89.43	3.67	1.60	0.99	0.65	2.67	-	-	0.99	-
[36]	7.35	86.29	2.30	0.12	0.57	0.57	-	0.62	0.27	-	-
[37]	2.67	91.94	1.69	0.11	0.29	0.25	1.05	0.44	0.37	0.44	0.07
[38]	-	93.5	1.4	0.1	0.55	0.23	1.11	0.31	0.03	-	-
[39]	2.61	88.07	2.02	1.15	1.35	0.22	1.04	0.74	0.49	0.12	-
[40]	1.06	87.22	1.12	0.20	0.70	1.68	2.12	1.18	0.04	-	0.46

It can be figured out from the reported compositions of rice husk ash that the amount of metallic impurities, primarily K_2O, affects the quantity of silica. Furthermore, the silica content produced in RHA is also influenced by the carbon content, which is determined by loss on ignition. Hence, to improve the silica percentage in rice husk ash, it is required to minimise the number of unburnt carbon and alkali metals, mainly K_2O, during its production. Furthermore, on the silica surface, two kinds of functional groups are associated: siloxane (Si-O-Si) and silanol (Si-OH). Silanol groups further have three classifications: vicinal, germinal, and isolated. Among all these types, the most reactive group is isolated silanol. However, the siloxane group is considered nonreactive [41]. Moreover, potassium oxide is regarded as a crystallising agent as it is the reason for the surface melting of silica during combustion [20]. Metallic impurities in rice husk, like sodium and potassium, react with silica during incineration. As a result, eutectic processes started occurring, yielding eutectic compounds with a low melting point. Therefore, a large amount of potassium or sodium reduces the actual crystallisation temperature of silica to 700^0C [42]. The eutectic reaction between silica and alkali metals, mainly potassium, produces a glass layer that prevents oxidation at high temperatures. Hence, this situation promotes the crystallisation of silica with a higher percentage of loss on ignition [43]. R. V. Krishnarao *et al.,* [44] discussed in their research that alkali oxides, mainly K_2O, affect the purity of silica. As the temperature rises, potassium causes the surface of rice husks to melt. Subsequently, the unburnt carbon from organic matter dissolves in it; hence, amorphous silica changes its state to crystalline form. Furthermore, by gradual heating, all carbons can be removed before the dissociation temperature of K_2O, which helps produce pure amorphous silica. Furthermore, Jia Xiao *et al.,* [45] reported that during the formation of cement paste, internal curing starts because of RHA, hence cement hydration rate and pozzolanic reaction of RHA increases. As a result, more C-S-H gel formed which mitigates the porosity and improve strength.

3. Processing of Highly Amorphous RHA

As previously mentioned, the factors that may affect the pozzolanic behaviour of rice husk ash are the amount of active silica, alkali metal oxides, heating conditions, and particle size. Hence, by optimising all these factors can enhance the production of amorphous silica from rice husk ash. In order to increase the performance of rice husk ash, heating conditions play a vital role [14]. There are few other ways of producing RHA, but thermal treatments yield silica with less unburnt carbon. Therefore, it gets the attention of most researchers [46]. Subsequently, B. A. Tayeh *et al.,* [47] highlighted that an understanding of the reaction kinetics of rice husk ash should be developed to improve the productivity of active silica obtained from silica. The burning temperature between 600^0C to 800^0C is suitable for getting amorphous silica. However, if the temperature exceeds 800^0C, the amorphous silica phase starts converting into crystalline silica as cristobalite.

Furthermore, if the temperature increases up to 1000^0C, then the formation of tridymite occurs [48]. The combustion process in the presence of oxygen produces active silica of about 98.32%. Moreover, heating rice husk in the range of 600^0C to 800^0C improves the content of active silica from 83.66% to 92.90% [49]. The burning temperature of rice husk, from 600^0C to 800^0C for 2 hours, is the optimum condition for getting active silica [50].

To eliminate or reduce the crystallisation process of silica, researchers adopted the chemical treatment of rice husk to eliminate deleterious alkali. Acid leaching treatment before combustion can potentially remove metallic impurities from rice husks [51]. Owing to the elimination of metallic impurities, the temperature conditions do not affect the oxidation process, and the formation of black particles in rice husk ash does not occur [44]. Silica obtained from acid-leaching pre-treatment is purer and highly amorphous (over 95%) [52]. Furthermore, acid leaching treatment produces rice husk ash in a white colour, which means having less unburnt carbon, which increases the purity of silica as pozzolanic material [53]. In addition to this, R. V. Krishnarao *et al.,* [44] shared a study related to black particles in rice husk ash that appear due to different conditions of combustion. Three samples of rice husk were investigated, which were raw husk and treated husk with different acid concentrations (RRH, TRH1, and TRH2, respectively). All samples were put in different heating conditions: sudden heating and slow heating. It was found that the ash produced by slow heating conditions was whiter than the ash produced by sudden heating conditions, as shown in Figure 3. It was concluded that the slow heating allowed the carbon to oxidise completely before melting the silica surface, whereas it did not happen with sudden heating because K_2O dissociated earlier and reacted with silica. At last, the remaining unburnt carbon is trapped in the eutectic compound produced by the eutectic reaction.

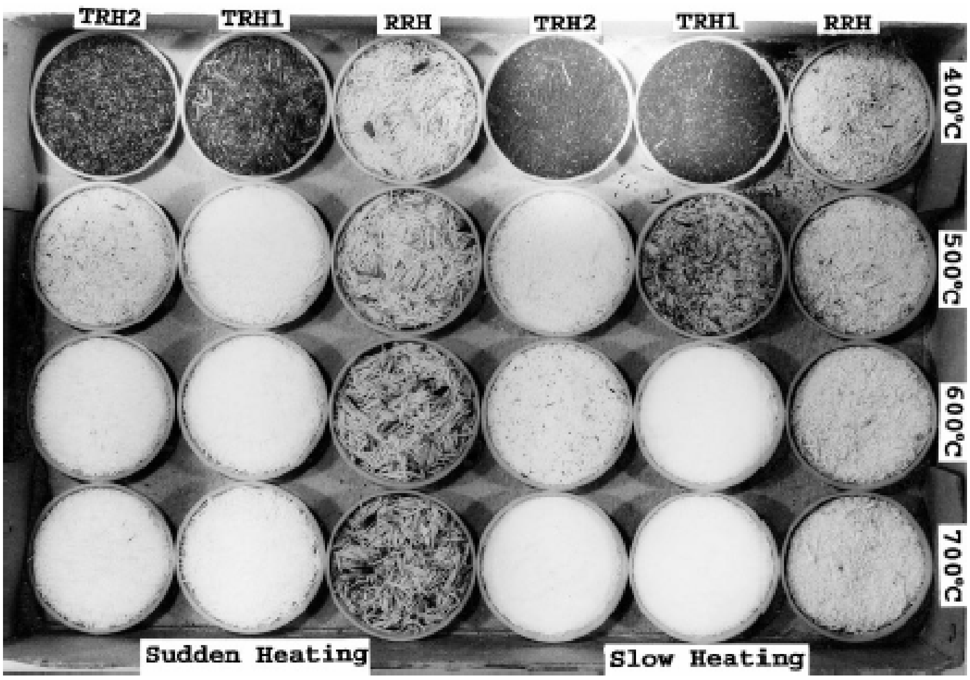

Fig. 3. Black Particles in RHA at different conditions [44]

It was researched by Y. S. Wong *et al*., [54] that a high percentage of silica can be produced by using hydrochloric acid (0.01 to 1.0M) as a leaching agent and with a burning temperature of 800^0C in the presence of oxygen. In conclusion, the maximum silica content of around 99.49% was obtained by 48 hours of leaching with 0.1M acid, while the burning duration was 2 hours.

In the manufacturing of rice husk ash, A. Gholizadeh-Vayghan *et al.,* [55] reported that the grinding time, acid leaching, and incineration retention time are essential to its wide application. The combustion temperature, with an increasing rate of 10^0C/min up to 700^0C for a specific duration, then well pulverization, is favourable for RHA production. Although the retention time of combustion is set according to the type of leaching acid, it was also emphasised that the oxidation process is affected by the low oxygen supply in the furnace. As a result, combustion time increases, ultimately raising unburned carbon content.

The investigation done by W. Xu *et al*., [56] showed that the crystallisation temperature of silica after leaching rice husk with different acids (like sulfuric acid, hydrochloric acid, and nitric acid) increases to 1200^0C and remains in the amorphous phase for about 2.5 hours. It was found that the hydrochloric acid leaching treatment with an acid concentration of 1.0 N and a leaching time of 2.5 hours can produce active silica of over 96% by heating at 600^0C for 2 hours. However, it was observed that annealing at 900^0C for 6 hours after chemical treatment still shows an utterly amorphous structure. It was also figured out that the presence of potassium in rice husk is the main reason for promoting the crystallisation of silica during combustion. During heating, the eutectic compounds of potassium and silica do not allow carbon to release; subsequently, carbon is trapped in the melted part of silica. Thus, when temperature increases, trapped carbon causes a black particle appearance, indicating silica's changing state from amorphous to crystalline.

Y. S. Wong *et al*., [24], shared that rice husk ash can be leached using various acids like sulfuric acid, nitric acid, and hydrochloric acid; however, hydrochloric acid is more efficient than any other acid as a leaching agent. This study analyses the effect of acid-leaching pretreatment at different concentrations. As a result, it was determined that 1.0M of HCl for leaching at 800^0C for 2 hours is enough to yield white RHA containing a high percentage of silica (99.28%) that shows pozzolanic properties to a great extent if it is followed by a combustion temperature of 800^0C for a short time of 2 hours.

Furthermore, the leaching treatment was performed before the combustion by R. A. Bakar *et al*., [57], using hydrochloric acid and sulfuric acid as leaching agents. Due to the removal of alkali metals by leaching treatment, the eutectic reaction between alkali metals and SiO_2 does not occur. As a result, a complete amorphous form of silica is yielded at a combustion temperature of 600^0C. Both acids were efficient in producing around 99% silica, but the hydrochloric acid leached ash has a larger surface area than the others.

A study reported that the pozzolanic activity of rice husk increases, and it shows adequate quality when well pulverised and burned under controlled conditions. Furthermore, it also investigated the pozzolanic properties of hydrochloric acid pretreated rice husk using a rapid evaluation method; that is, the more efficient the rice husk ash appears, the greater the change in the conductivity of $Ca(OH)_2$. It was observed that the treated RHA has a higher value of change in the conductivity of saturated $Ca(OH)_2$ than the untreated RHA. However, this change in conductivity descends remarkably as the incineration temperature rises above 700^0C, which concludes that the pozzolanic behaviour of treated RHA may be affected by not maintaining a desired temperature [58].

To produce active silica from rice husk R. Prasad *et al.,* [59] performed several pre and post-incineration chemical treatments (with HCl, HNO_3, NaOH, H_2SO_4 etc.) along with combustion temperatures ranging from 773 – 1673 K. Moreover, it was found that the formation of crystalline silica in the form of cristobalite and tridymite begins when the temperature goes up to 1073 K.

Although, by controlling the incineration temperature, more amorphous silica can be obtained from rice husks.

C. Real *et al.,* [60] Showed in his research that RHA obtained by acid leaching treatment before the combustion of rice husk is more efficient (99% pure) than RHA obtained without leaching treatment or leaching treatment after annealing. However, interestingly, it was observed that this RHA changed its state from amorphous to crystalline at 1300^0C.

In continuation, P. Chen *et al.,* [43] discussed some insights into the leaching process for producing pure silica in rice husk ash. It was noticed that there are two phases of the leaching process: the rapid leaching phase and the slow leaching phase. In the beginning, the hydrogen ion from an acid rapidly reacts with metals to perform an ion exchange process. After a specific time (till 30 minutes), the process slows to achieve an equilibrium state. It was reported that leaching time above 120 minutes shows no further changes.

Moreover, it was also discussed that the removal of alkali metals is done more efficiently by mineral acids (like HCl) than organic acids. Additionally, P. P. Nayak *et al.,* [61] highlighted that more appropriate studies on the leaching process should be done to get favourable leaching time and temperature conditions because the leaching duration and temperature vary significantly. Furthermore, leaching process conditions adopted by different authors are presented in Table 2, which shows that the leaching duration and temperature used in different studies are different; hence, there is a need to increase the study area that also covers the efficient way of leaching treatment.

Table 2
Leaching treatment Conditions followed by different authors

Reference	Acid Concentration	Leaching Duration	Leaching Temperature
[62]	500ml of 0.1M HCl to leach 50g RH	1.5 h	-
[63]	10% HCl	2 h	100^0C
[64]	500ml of 3N HCl to leach 100g RH	4 h	80^0C
[65]	500ml of 3M HCl to leach 50g RH	24 h	Room Temperature
[66]	0.5N HCl	0.5 h	60^0C

It was discussed earlier that the purity of RHA does not only depend on the acid leaching and combustion processes; the grinding conditions also influence the efficiency of the production of RHA. In order to improve the pozzolanic behaviour, the degree of fineness of the material is crucial. S. Mirmohamadsadeghi *et al.,* [41] Discussed that in the manufacturing of RHA, to attain the purity of silica, it should be well ground. Furthermore, the scanning electron microscopy (SEM) analysis done by A. Rivas *et al.,* [67] shows some interesting images of rice husks, which conclude that the outer surface of the rice husk contains more silica. In contrast, the significant impurities are situated on the inner surface of the rice husk. Hence, to increase the efficiency of chemical treatment, the exposure of impurities to the leaching solution should be enhanced by well-pulverised rice husk.

4. Optimising Factors

Previous studies have proved RHA to be a potential cement replacement material because of the presence of highly amorphous silica. However, the existing literature highlights the key variables that can influence the pozzolanity of RHA as an alternative material. Burning conditions of RHA are one of the most critical parameters for the synthesis of RHA. A combustion temperature ranging between 600^0C to 800^0C for duration of 2 hours with proper oxygen supply can provide white ash, which is good as a cement replacement material. Moreover, the pozzolanic behaviour of RHA also depends on the large surface area and porous structure of RHA. Beside the heating conditions, grinding conditions also affect the pozzolanity of RHA, as it is correlated with the particle size and surface area. In addition to this, to optimise the properties of RHA, one crucial factor is the acid leaching treatment of rice husk. It can be concluded that most of the researchers considered using HCl as a leaching agent with a concentration of 0.1M to 1M, while having a leaching temperature of up to 80^0C for 2 hours is enough for the manufacturing of amorphous RHA. The schematic diagram of yielding optimised rice husk ash is represented in Figure 4.

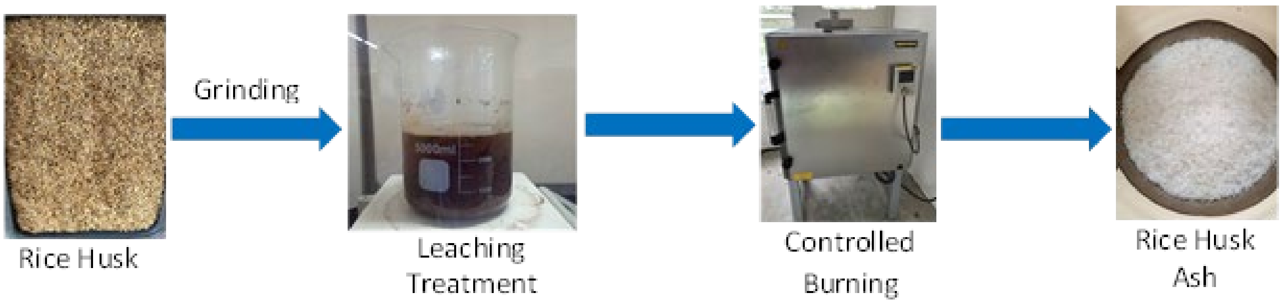

Fig. 4. Graphical representation of Optimized Rice Husk Ash Production

5. Conclusion

Rice husk ash can be used as a supplementary cementitious material. It not only increases the strength and durability of concrete but also provides a solution to reduce the carbon emissions created by the cement industry and helps the rice-producing sector manage rice waste efficiently.

The extensive research review on the production of functionalized rice husk ash shows that the significant influencing variables of amorphous silica production from rice husk are combustion temperature, combustion time, metallic impurities, predominantly potassium, leaching process, and grinding conditions. The formation of carbonaceous matter during the combustion of rice husk is due to the presence of alkali metals, mainly potassium. The dissociation of potassium oxide should be avoided before complete oxidation by incinerating rice husk in the presence of oxygen. Because if the eutectic process begins, then the remaining carbon will be trapped by eutectic compounds, which influence the purity and promote silica's crystallisation. Most of the studies agreed that acid leaching treatment followed by the incineration process with a temperature of up to 700^0C for 2 hours is suitable for achieving high-quality amorphous silica. Mineral acids like HCl are more suitable as a leaching agent and provide more amorphous silica than other acids.

Although, in the previous investigations, critical factors like combustion temperature and leaching treatment were discussed thoroughly, nevertheless, there are still some areas that should be investigated in the future: (1) understanding the reaction kinetics of rice husk ash by adopting advanced characterization methods (2) evaluate the effect of grinding and leaching conditions, as these significantly vary in almost all investigations. It is believed that these research gaps will definitely upgrade the knowledge regarding rice husk as a supplementary cementitious material and increase the production of highly amorphous silica via the combustion of rice husk.

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