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# Anthracite briquettes with plant byproducts as an ecofriendly fuel for foundries

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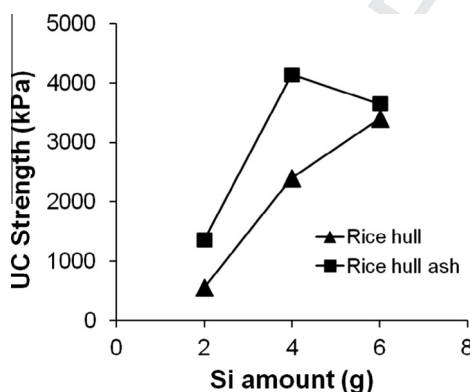
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## HIGHLIGHTS

- Anthracite briquettes were developed with waste anthracite as an ecofriendly fuel.
- Ecofriendly fuel is an alternative to conventional coke used in a cupola furnace.
- Si metal reacts with anthracite to form SiC providing mechanical strength to fuel.
- The novel briquettes showed high mechanical strength in the range of 2000–4000 kPa.

## GRAPHICAL ABSTRACT

Effect of Si metal powder on UC strength of anthracite briquettes after pyrolysis.



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## ABSTRACT

Anthracite briquettes were developed using mainly waste anthracite fines, plant byproducts of rice hulls and rice hull ash and silicon metal powder as an ecofriendly fuel and as an alternative to conventional coke used in a cupola furnace. Si metal powder reacts with anthracite to form SiC, which provides high mechanical strength to an anthracite briquette when it is exposed to a high temperature in the cupola furnace. Rice hull and rice hull ash, which are agricultural byproducts with high SiO<sub>2</sub> content were investigated as partial substitutes for silicon as the former are inexpensive relative to silicon metal. The formation of SiC was investigated under different conditions using X-ray diffraction and transmission electron microscopy. In addition, unconfined compressive strength of the briquettes was measured when the rice hull or ash substituted Si powder partially or completely. When anthracite briquettes prepared with elemental Si powder and either rice hull ash or rice hull powder were pyrolyzed at 1400 °C for 2 h, SiC formed and the briquettes showed high mechanical strength in the range of 2000–4000 kPa, which is enough to keep their structural integrity in a cupola furnace.

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## 1. Introduction

A cupola furnace is used to melt iron and it consists of three distinct regions such as dropping, heating, and melting zone [1]. In order for a carbon source to be used as a fuel in a cupola furnace, it is necessary to keep its porous structure for fast burning as it passes through the three different zones. Coke is a commonly used solid fuel to satisfy the demand in a cupola furnace of the foundry industry. Coke has high energy, low volatility, and a rigid porous structure, which facilitates fast burning as well as maintaining the structural integrity during the metal melting process in a cupola furnace [2,3]. However, coking process, which produces a foundry grade coke using bituminous coal, causes emission of pollutants such as volatile organic compounds (VOCs), polynuclear aromatic hydrocarbons (PAH), carbon monoxide, sulfur oxide, and ammonia, etc. [4]. Also, in the coking process the pyrolysis step at 1000 °C for 16–36 h leads to the loss of 10–20% energy. Thus the use of coke leads to environmental pollution through harmful emissions as well as energy loss. Besides the above drawbacks, coke is no longer cost-effective as the cost of coke increased to 400% in a short span of time, i.e., from 2002 to 2010 [1].

In order to overcome the above disadvantages we have been developing ecofriendly anthracite briquettes as an alternative fuel to replace the coke because anthracite has similar chemical properties to that of coke [1,3,5]. In our previous studies waste anthracite fines were briquetted into bricks using denatured collagen with silicon metal powder and lignin. This type of briquettes could hold their structure when they were pyrolyzed at 1400 °C for 2 h in a tubular furnace [1] and could be stored for a long time at ambient temperature without loss of structural strength via surface application of clarified neem oil and copper which prevents biological activity on bindered anthracite bricks before use for their optimal performance in cupola furnaces [6]. The structural integrity of these bricks was demonstrated effectively in a full-scale cupola furnace suggesting that the replacement of the conventional coke as a fuel might be feasible [7]. The high structural strength of briquettes in all three cupola regions was derived from the various additives used when preparing the anthracite briquettes. The use of denatured collagen contributes to the strength of bricks in the dropping zone, fused lignin contributes for sustaining their solid structure in the heating zone, and silicon metal powder added to briquettes gives very high strength by forming silicon carbide (SiC) in situ in the anthracite bricks in the high temperature region [1,3,8]. The formation of silicon carbide in the fuel briquettes is essential for their use as a fuel in the cupola because it helps the bricks to endure their physical stability up to the melting zone where temperatures are around 1600 °C.

SiC is a versatile ceramic material due to its many useful properties including extremely high oxidation resistance, thermal resistance and mechanical strength [9]. In general SiC has been synthesized through a variety of methods such as chemical vapor deposition of Si powder [10,11], direct carbonization of hydrocarbon with Si [12], decomposition of organic silicon compounds [13], and carbothermal reduction of SiO<sub>2</sub> [14–16]. Our group also reported previously that silicon carbide nanowires could be grown in situ on the anthracite fine surfaces in anthracite briquettes with Si metal powder through vapor–solid mechanism [8] under high-temperature carbothermal conditions. Several previous studies were focused on the use of silica-containing plant-based agricultural biomass or wastes such as cotton fibers [17], rice hulls [18–20] and rice hull ash [21] for the synthesis of SiC which are more cost-effective than Si powder. For the synthesis of SiC with rice hulls, Janghorban and Tazesh [22] used sodium silicate while others used some metallic compounds such as Fe<sup>0</sup> [23], FeCl<sub>2</sub>

[24], CoCl<sub>2</sub> [25] and NiCl<sub>2</sub> [26] as catalysts to produce SiC. However, synthesis of SiC was also reported from this plant-based Si source without any catalysts [27].

Therefore, the objective of this study was to investigate the formation of SiC in anthracite fuel briquettes with/without the addition of different catalysts when rice hulls or rice hull ashes were used to replace silicon powder completely or partially. This study reports the preparation of anthracite briquettes using agricultural byproducts like rice hulls or rice hull ash for the first time.

## 2. Materials and methods

### 2.1. Raw materials

Raw anthracite was obtained from Jeddo Coal Company (Hazelton, PA). This was Jeddo's No. 4 anthracite. It was dried at 110 °C for 1 day to remove its moisture. Then it was packed into a cylindrical mold (2.86 cm diameter, 4.76 cm long) and crushed using a pressing unit at 69 MPa (10,000 psi) before being used to prepare fuel briquettes. Dry, small, granular form of collagen was provided by Entelechy, representing Hormel Foods Company (Austin, MN). Elemental silicon lumps (98.4% purity, Alfa Aesar) were grounded into powders with a SPEX 8000 mixer/mill from A.O. Smith Corp. (Milwaukee, WI) and then sieved with a U.S. #100 mesh. Soft lignin derived from pine was supplied from Innventia (Stockholm, Sweden). It was dried at 60 °C for 1 day and subsequently sieved with a U.S. #40 mesh. Rice hulls and rice hull ash were obtained from Riceland (Stuttgart, AR). Rice hull ash was sieved with a U.S. #100 mesh before use. Rice hulls were dried in an oven overnight at 60 °C and then ground using a coffee grinder (model PG13658) to prepare rice hull powder. After that, it was sieved with a U.S. #40 mesh.

### 2.2. Preparation of anthracite briquettes

For a typical preparation of anthracite briquette or pellet, 100 g of the precrushed anthracite and 1.5 g of collagen were used with a variety of additives including different Si sources to increase strength of anthracite pellets. First, 1.5 g of collagen was put in 9–20 ml of water in a nickel beaker and then it was dissolved in a water bath at 70 °C to form a gelatin solution. If there is any dissolvable additive, it would be added to the beaker along with collagen. Appropriate amounts of Si-containing materials including elemental Si powder and lignin were added into the gelatin solution and subsequently all materials were mixed thoroughly with 100 g of the precrushed anthracite. After that, the final mixture was packed into the mold with 69 MPa in order to make a pellet or briquette. After extruding it from the mold, the pellet was dried overnight under ambient conditions.

The pellets with catalysts were prepared in the same way as normal anthracite briquettes after adding the catalysts. As a catalyst, 3 g of NiCl<sub>2</sub> (Aldrich, 98%), CoCl<sub>2</sub>·6H<sub>2</sub>O (J.T. Baker, 99.6%), FeCl<sub>2</sub>·4H<sub>2</sub>O (Aldrich, 99.9%), 2–6 g of elemental Fe (Aldrich, 99.9%) or 2.5 g of sodium silicate (Alfa Aesar, 44.8–47.6% SiO<sub>2</sub>, 49.5–51.5% Na<sub>2</sub>O) was used to facilitate the growth of Silicon carbide by an increase in the reaction rate during pyrolysis. The catalysts were used to find out whether or not they would lead to the formation and growth of SiC nanowires without the addition of Si powder. The metal salts were first dissolved in 10–20 ml of deionized water and then mixed with the precrushed anthracite particles, while Fe metal powder was directly blended with precursors of the anthracite briquettes. Sodium silicate solution was prepared through dissolution of 2.5 g sodium silicate in 100 ml of deionized water when it was used as a catalyst. First, rice

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