# Emission of Sulphur Dioxide during Coal Briquette Burning in a Thai Traditional Cooking Stove

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## Abstract

Experiments investigating the effects of sulphur content, CaO/S mole ratio, and the amount of excess air on the emission of sulphur dioxide during the combustion of coal briquettes were carried out in a 24 cm in diameter, 30 cm high, Thai traditional cooking stove. Sulphur contents and CaO/S mole ratios of coal briquettes ranged from 1-5 %wt. and 0-4, respectively. The comparison of the use of coal briquettes with charcoal and firewood was also studied. It can be concluded from the experiments that the concentration of sulphur dioxide was high in the first 10-30 minutes of the combustion time. The concentration of sulphur dioxide increased with sulphur content in coal briquettes. The concentration of sulphur dioxide decreased when CaO/S mole ratio increased, and levelled off when CaO/S mole ratios were greater than 2. When determining the total sulphur emitted into the atmosphere, comparing to the initial sulphur content in coal briquettes (*i.e.*, %S emission), it was found that %S emission decreased when sulphur content and CaO/S mole ratio increased. The amount of excess air did not affect %S emission. The comparison of the use of coal briquettes with charcoal and firewood showed that the concentration of sulphur dioxide from the combustion of coal briquettes was significantly higher than that of charcoal and firewood. The usage efficiency of coal briquettes was found to be higher than that of charcoal and firewood. The ignition time of coal briquettes was significantly longer than that of charcoal and firewood. Recommendations were made such that coal briquettes with sulphur content and CaO/S mole ratio of 2 %wt. or lower and 2, respectively, be used for household cooking, and an oxidiser be added into coal briquettes in order to shorten the ignition time and to increase the temperature in the first stage of the combustion.

**Keywords**: sulphur dioxide, cosl briquette, traditional cooking stove

## 1. Introduction

The consumption of charcoal and firewood for household cooking in Thailand is very high, especially in rural areas. It was reported [1] that the amounts of charcoal and firewood used for household cooking in 1997 were approximately 3 and 6 million tonnes, respectively. Since charcoal and firewood mostly come from domestic wood, this leads to heavy deforestation in Thailand. Hence, it is necessary to develop alternative sources of energy to substitute the use of charcoal and firewood for household cooking. Even though there are a lot of alternative fuels, such as LPG (Liquefied Petroleum Gas) and kerosene which can be used for household cooking, such fuels are expensive and not familiar to rural people. In addition, they cannot be used with existing traditional cooking stoves. Coal briquettes, which are made of coal fines, on the contrary, are relatively inexpensive, available domestically, and can readily be used with the existing traditional cooking stoves. This is because coal is abundant in Thailand [1, 2], and the way that coal briquettes are used in cooking stoves is similar to that of charcoal and firewood. Thus, coal briquettes have become a promising choice.

However, most coals in Thailand are lignitic and sub-bituminous, which contain a high amount of volatile matter. Additionally,

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their sulphur contents are often high (>3 %wt.) [2].As a result, there are a number of problems emerging during the combustion of coal briquettes, such as the emissions of sulphur dioxide and nitrogen oxides, and the emission of a high amount of volatile matter. These make the use of coal briquettes unsafe and unattractive, since cooking stoves are generally used without chimneys. In order for coal briquettes to become more attractive, the above problems must be solved.

During the past two decades, there have been a number of studies trying to improve the quality of coal briquettes by various means, such as carbonising coal fines before briquetting [3, 4] or adding lime (CaO) into coal briquettes [5, 6]. Lime added into coal briquettes reacts with sulphur dioxide,

$$CaO + SO_2 + \frac{1}{2} O_2 \rightarrow CaSO_4. \quad (1)$$

Calcium sulphate formed in reaction (1) is stable and, thus, captured in the residual ash. It was reported [5, 6] that lime could capture 50-95% of sulphur dioxide. This, however, indicates that significant amounts of sulphur dioxide are still emitted into the atmosphere during the combustion of coal briquettes.

Many researchers [7-9] have tried to reduce the emission of sulphur dioxide during the combustion of coals or coal briquettes, especially at industrial scales. However, few have been concerned with small scales, especially at the level of household cooking. Hence, it is the main purpose of this research to investigate the factors affecting the emission of sulphur dioxide during the combustion of coal briquettes in a Thai traditional cooking stove. Data and results obtained from this research lead to recommendations and suggestions for the production and the use of coal briquettes that are suitable for household cooking, while emitting low amounts of sulphur dioxide.

## 2.Materials, Apparatus, and Procedure

The raw coals consisted of two samples from the Banpu basin (BP1 and BP2), and two from the Mae-Moh basin (MM1 and MM2). Their proximate and ultimate analyses are summarised in Table 1. Proximate analyses of charcoal and firewood, which were bought locally, are shown in Table 2. The raw coals were crushed and ground to a size of less than 1 mm using a hammer mill. Typically, two samples of coals were mixed with each other (*e.g.*, BP1+BP2 or BP1+MM2) and lime (commercial grade, CaO content was 64.7 %wt.) to produce coal mixtures with sulphur contents and CaO/S mole ratios ranging from 1-5 %wt. and 0-4, respectively. Additionally, in order to minimise the effect of ash, which contains, *e.g.*, Ca and Mg that can capture sulphur dioxide as well, on the emission of sulphur dioxide, two samples were mixed such that the amount of ash in coal briquette samples was in the same range (approximately 25-30 %wt.). Coal mixtures were mixed with water in a mixer and then compressed to ovoid-shaped coal briquettes using a double-ring-roll-type briquetting machine. The weight of each coal briquette was approximately 15-20 g.

Each sample of coal briquettes (the sample name indicates the sulphur content and CaO/S mole ratio; *e.g.*, S12 is the sample whose sulphur content and CaO/S mole ratio are 1 %wt. and 2, respectively) was burned in a 24 cm in diameter, 30 cm high, Thai traditional cooking stove. A schematic diagram of the experimental set-up is shown in Figure 1. The mass of coal briquettes used for each test run was such that the total heat output was equivalent to that of 400 g of charcoal; this amount of charcoal is typically used with this size of stove. This corresponded to the mass of coal briquettes ranging from 658-929 g (it should be noted that, even though the mass of coal briquettes used were different, the effect of the mass of coal briquettes on the emission of sulphur dioxide was small, as reported in the previous study [2]). Oxygen and carbon dioxide concentrations were measured periodically using an Orsat analyser, which could measure both oxygen and carbon dioxide in the range of 0-21 %vol. with an approximate error of ± 2%. The concentration of sulphur dioxide and the temperature of the flue gas were measured continuously using a LANCOM 2004 gas analyser, which could measure sulphur dioxide in the range of 0-1000 ppm with an approximate error of ± 4%. Oxygen and carbon dioxide concentrations were measured every five minutes, while the concentration of sulphur dioxide and flue gas temperature were read every two minutes. The stove was placed on a

Table 1: Proximate and ultimate analyses of raw coal samples from the Banpu (BP) and Mae-Moh (MM) basins

| List of analysis | Sample | | | |
|---|---|---|---|---|
| | BP1 | BP2 | MM1 | MM2 |
| Proximate analysis (%wt.) | | | | |
| ▪ Moisture | 9.14 | 7.46 | 9.33 | 7.24 |
| ▪ Ash | 12.62 | 35.15 | 25.29 | 27.95 |
| ▪ Volatile matter | 42.91 | 33.98 | 34.69 | 34.24 |
| ▪ Fixed carbon (by diff.) | 35.33 | 23.41 | 30.69 | 30.57 |
| Ultimate analysis (%wt.) | | | | |
| ▪ C | 50.63 | 40.98 | 44.70 | 43.61 |
| ▪ H | 21.83 | 11.82 | 13.01 | 12.96 |
| ▪ N | 0.82 | 0.90 | 1.69 | 0.73 |
| ▪ S | 1.06 | 0.91 | 3.84 | 6.52 |
| ▪ O (by diff.) | 4.38 | 3.13 | 3.58 | 3.43 |
| Total sulphur (%wt.) | 1.16 | 0.87 | 3.76 | 6.73 |
| ▪ Pyretic sulphur | 0.17 | 0.31 | 0.57 | 0.62 |
| ▪ Sulphate sulphur | 0.33 | 0.04 | 1.49 | 2.72 |
| ▪ Organic sulphur | 0.66 | 0.53 | 1.71 | 3.43 |
| Heating value (MJ/kg) | 18.65 | 13.20 | 18.63 | 15.46 |
| Rank of coal | [a]sub-bit C | sub-bit C | sub-bit A | sub-bit B |

[a] sub-bit = sub-bituminous

Table 2: Proximate analyses of charcoal and firewood

| List of analysis | Sample | |
|---|---|---|
| | Charcoal | Firewood |
| Proximate analysis (%wt.) | | |
| ▪ Moisture | 4.64 | 10.60 |
| ▪ Ash | 2.95 | 1.32 |
| ▪ Volatile matter | 23.32 | 63.45 |
| ▪ Fixed carbon (by diff.) | 69.09 | 24.63 |
| Total sulphur (%wt.) | 0.10 | 0.10 |
| Heating value (MJ/kg) | 29.01 | 18.29 |

mass balance, and the weight was read every five minutes. The concentration of sulphur dioxide was also measured when either charcoal or firewood was burned in the same stove. The amount of firewood used for each test run was 634 g (again, the total heat output was equivalent to that of 400 g of charcoal).

The total amount of sulphur emitted into the atmosphere was calculated by integrating the area under the curve of the amount of sulphur emitted with time, where the amount of sulphur emitted at each time was calculated using the data of sulphur dioxide concentration, oxygen and carbon dioxide concentrations, the burnout rate, and the flue gas temperature.

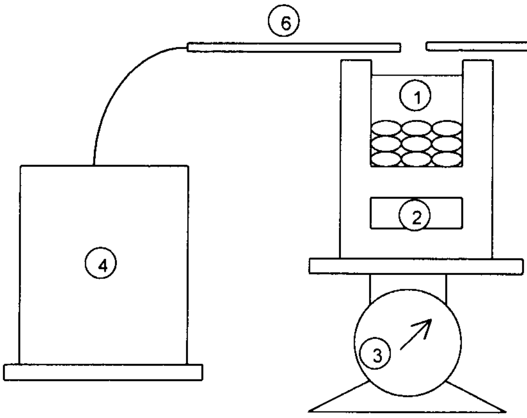


Figure 1: A schematic diagram of experimental set-up
(1) a traditional cooking stove; (2) a front window; (3) a balance;
(4) an Orsat analyser; (5) a flue gas analyser with a built-in thermocouple;
(6) a probe for the Orsat analyser; (7) A probe for the flue gas analyser.

The percentage of sulphur emission (%S emission) was calculated by dividing the total amount of sulphur emitted into the atmosphere by that of initial sulphur of each sample.

In order to study the effect of the amount of excess air on %S emission, the experiments were carried out in three different ways: 1) the front window of the stove was fully opened (this was a common practice that the experiments were carried out); 2) the front window was fully opened, but the air flow was assisted by a small electric fan; and 3) the front window was only half opened. These corresponded to the average (10-30 minutes) excess air percentages of 472, 612, and 297%, respectively.

The usage efficiencies of the combustion of coal briquettes, charcoal, and firewood were determined by heating and evaporating a known amount of water. The usage efficiency is the ratio of the total heat used for heating and evaporating water to the total heat output from the combustion of the fuel (*i.e.*, coal briquettes, charcoal, or firewood).

## 3. Results and Discussion

**The concentration profile of sulphur dioxide during the combustion of coal briquettes**

The concentration profiles of sulphur dioxide of various coal briquette samples are shown in Figure 2. The concentration of sulphur dioxide of each sample was high in the first 10-30 minutes of the combustion time with the peak at, approximately, 12-16 minutes. Between 10-16 minutes, even though the burning rate was high, the temperature was still relatively low since the coal briquettes had just started burning. This low temperature resulted in the low reactivity of the reaction between sulphur dioxide and calcium oxide [10, 11]. From approximately 12-30 minutes, the reactivity of the reaction between sulphur dioxide and calcium oxide increased due to the higher temperature [10, 11]. This caused the concentration of sulphur dioxide to decrease. However, due to the relatively high burnout rate, the concentration of sulphur dioxide was still high. After 30 minutes, the burnout rate of coal briquettes decreased. Meanwhile, a high amount of sulphur in coal briquettes had already been emitted during the first 10-30 minutes. Thus, after 30 minutes of the combustion time, the concentration of sulphur dioxide dropped continuously until coal briquettes had burned out.

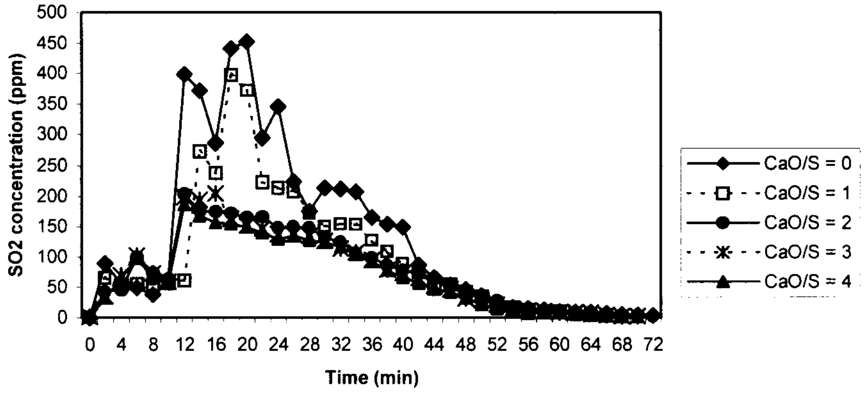


Figure 2: The concentration profiles of sulphur dioxide during the combustion of coal briquettes with sulphur content of 2 %wt. and with CaO/S mole ratios ranging from 0-4. All sulphur dioxide concentrations reported were corrected to 7% $O_2$.

**The effect of the amount of sulphur in coal briquettes on the concentration of sulphur dioxide**

The average concentrations of sulphur dioxide during the first 10-30 minutes of the combustion time of coal briquettes at various amounts of sulphur contents and CaO/S mole ratios are shown in Figure 3. It was clear that the concentration of sulphur dioxide increased with sulphur content in coal briquettes.

**The effect of CaO/S mole ratio on the concentration of sulphur dioxide**

In Figure 3, the concentration of sulphur dioxide decreased with an increase of CaO/S mole ratio, and levelled off when CaO/S mole ratios were greater than 2. This agreed with the results obtained in the previous works [5-9]. The molar volume of calcium sulphate, which is formed from the reaction between sulphur dioxide and calcium oxide, is bigger than that of the original calcium oxide. Calcium sulphate molecules then block the entrance of the pores of calcium oxide and, thus, prevent sulphur dioxide from entering into the pores to react with calcium oxide although it is still available [9]. Accordingly, the amount of calcium oxide needed must be greater than that of the stoichiometric ratio in reaction (1). The optimum CaO/S mole ratio found in this research was 2. This agreed with experiments done by other workers [5-9].

**The relation between sulphur content and CaO/S mole ratio in coal briquettes, and %S emission**

The relation between sulphur content and CaO/S mole ratio in coal briquettes, and %S emission is plotted in Figure 4. The percentage of sulphur emission (%S emission) decreased with an increase of sulphur content and CaO/S mole ratio in coal briquettes. The possible explanation might be that, when total sulphur content in coal briquettes increased, the amount of sulphate sulphur as well as the ratio between sulphate sulphur and total sulphur also increased, as shown in Table 3. Sulphate sulphur is relatively stable and, thus, not oxidised to sulphur dioxide [12]. An increase of CaO/S mole ratio caused the decrease of %S emission because calcium sulphate formed from the reaction between sulphur dioxide and calcium oxide was captured in the ash. This, therefore, reduced the amount of sulphur emitted into the atmosphere.

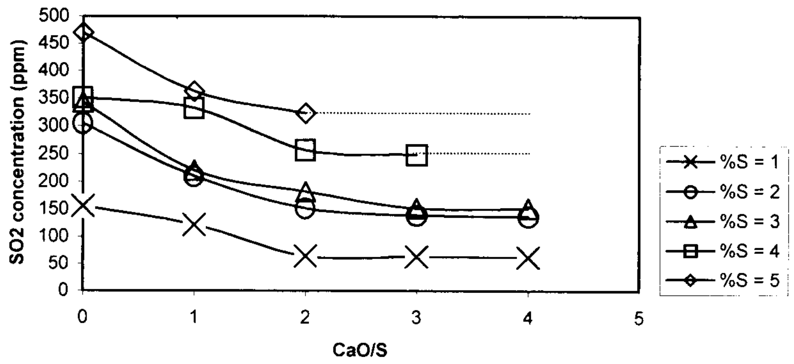


Figure 3: The average (10-30 minutes) concentrations of sulphur dioxide during the combustion of coal briquettes with sulphur contents and CaO/S mole ratios raging from 1-5 %wt. and 0-4, respectively (note: dot lines illustrate predictive trend). All sulphur dioxide concentrations reported were corrected to 7% $O_2$.

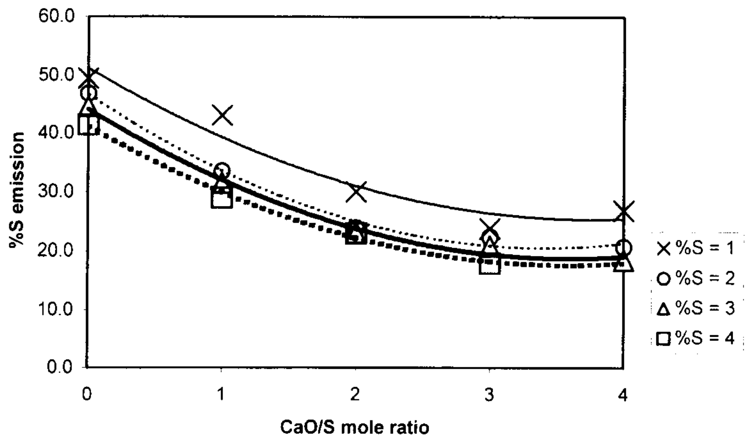


Figure 4: Percentages of sulphur emission (%S emission) of coal briquettes with sulphur contents and CaO/S mole ratios ranging from 1-5 %wt. and 0-4, respectively; each line in the Figure represents %S emission of each sulphur content calculated from Equation (2).

Table 3: Sulphate sulphur contents and percentages in coal briquettes at various amounts of total sulphur contents

| Total sulphur content in coal briquettes (%wt.) | Sulphate sulphur content in coal briquettes (%wt.) | Percentage of sulphate sulphur content to total sulphur content in coal briquettes (%) |
|---|---|---|
| 1 | 0.2 | 20 |
| 2 | 0.6 | 30 |
| 3 | 1.1 | 37 |
| 4 | 1.6 | 40 |
| 5 | 2.1 | 42 |

The correlation between sulphur content (%S) and CaO/S mole ratio (CaO/S) in coal briquettes, and the percentage of sulphur emission (%S emission) can be presented as the following empirical equation:

$$\%\text{S emission} = \frac{49.5 - 1.77[\%S]}{\exp(0.253[CaO/S])} \quad (2)$$

Each of the lines in Figure 4 represents %S emission at different sulphur contents calculated from equation (2).

**The effect of the amount of excess air on %S emission**

Table 4 summarises percentages of sulphur emission (%S emission) at different amounts of excess air. It seemed that the amount of excess air did not affect %S emission. This might result from the reason that the chemical equilibrium of the reaction between sulphur dioxide and calcium oxide was not affected by the amount of excess air.

**The comparison of the use of coal briquettes with charcoal and firewood**

Since charcoal and firewood are widely used for household cooking, in order to evaluate the suitability of the use of coal briquettes for household cooking, they should be compared with charcoal and firewood. Thc comparison of the use of coal briquettes with charcoal and firewood in the aspects of the concentration of sulphur dioxide, the usage efficiency, the ignition time, and the burnout time is summarised in Table 5.

The concentration of sulphur dioxide from the combustion of coal briquettes was significantly higher than that of charcoal and firewood. However, the average concentration of sulphur dioxide during the first 10-30 minutes of the combustion time of coal briquettes with sulphur content of either 1 or 2 %wt. and with CaO/S mole ratio of 2 was lower than the U.S. emission standard of this gas, at which the maximum acceptable level of 150 ppm (corrected to 7% $O_2$) is set [13].

When considering the usage efficiency, it was found that the usage efficiency of coal briquettes was higher than that of charcoal and firewood. This might result from the longer burnout time of coal briquettes comparing to that of charcoal and firewood.

The ignition time of coal briquettes, however, was significantly longer than that of charcoal and firewood. This was a drawback and, thus, needs to be improved.

Table 4: Percentages of sulphur emission (%S emission) at various amounts of excess air

| Characteristics of the front window of stove and the air flow into the stove | Corresponding excess air percentage (% excess air) | Percentage of sulphur emission (%S emission) |
|---|---|---|
| ▪ The window was fully opened, and the air flow was assisted by a small electric fan | 612 | 22.0 |
| ▪ The window was fully opened, and the air flowed into the stove by natural convection | 472 | 23.6 |
| ▪ The window was only half opened, and the air flowed into the stove by natural convection | 297 | 23.0 |

Table 5: The comparison of the use of coal briquettes (sample S12 and S22)[a] with charcoal and firewood

| Fuel | The average (10-30 min.) concentration of $SO_2$ (ppm)[b] | Usage efficiency (%) | Ignition time (min) | Burnout time (min) |
|---|---|---|---|---|
| ▪ S12 | 65 | 34.5 | 8-10 | 68-70 |
| ▪ S22 | 149 | 35.9 | 8-10 | 70-72 |
| ▪ Charcoal | 9 | 32.9 | 3-4 | 54-60 |
| ▪ Firewood | 5 | 25.2 | 2-3 | 42-45 |

[a] Sample Sxy = the coal briquette sample with sulphur content and CaO/S mole ratio of x %wt. and y, respectively

[b] All sulphur dioxide concentrations reported were corrected to 7% $O_2$.

## 4. Conclusions and Recommendations

1) The concentration of sulphur dioxide was high in the first 10-30 minutes of the combustion time. The concentration of sulphur dioxide increased with sulphur content in coal briquettes.
2) The concentration of sulphur dioxide decreased when CaO/S mole ratio increased, and levelled off when CaO/S mole ratios were greater than 2.
3) The percentage of sulphur emission (%S emission) decreased when sulphur content and CaO/S mole ratio in coal briquettes increased. The relation between sulphur content (%S) and CaO/S mole ratio (CaO/S) in coal briquettes, and %S emission can be presented as follows:

$$\%S\ \text{emission} = \frac{49.5 - 1.77[\%S]}{\exp(0.253[CaO/S])}.$$

The amount of excess air did not affect %S emission.
4) The concentration of sulphur dioxide from the combustion of coal briquettes was significantly higher than that of charcoal and firewood. However, the average (10-30 minutes) concentration of sulphur dioxide of coal briquettes with sulphur content of either 1 or 2 % wt. and with CaO/S mole ratio of 2 was lower than the U.S. emission standard (*i.e.*, lower than 150 ppm).
5) The usage efficiency of coal briquettes was found to be higher than that of charcoal and firewood.
6) The ignition time of coal briquettes was significantly longer than that of charcoal and firewood.
7) Recommendations were made such that coal briquettes with sulphur content of 2 %wt. or lower and with CaO/S mole ratio of 2 be used for household cooking, and an oxidiser be added into coal briquettes in order to shorten the ignition time and to increase the temperature in the first stage of the combustion. The higher temperature results in the higher reactivity of the reaction between sulphur dioxide and calcium oxide.

## 5. Acknowledgements

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