The Influence of Fuel Reactivity on Iron Ore Sintering

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An experimental program conducted at CSIRO showed that flame front speed (ffs) and sinter productivity increased with fuel reactivity (r) while sinter strength and fuel utilisation fell. The following relationship modelled flame front speed for a JSM style sinter mix over a fuel reactivity range of 1 x 10^-6 g/g/s to 4 x 10^-3 g/g/s:

\[
\text{ffs} = 0.2014 \cdot \ln(r) + 4.039, \quad R^2 = 0.997
\]

where ffs is flame front speed (cm/min) and r is fuel reactivity (g/g/s).

The CSIRO research concluded that rapid heating of the lower bulk density green granules and endothermic fuel gasification preceding the flame front are dominant factors that contribute to changes in flame front speed. While productivity was significantly increased, less time at temperature reduced sinter strength and gasification contributed to poor fuel utilisation under the standard sintering conditions used for the experiments. A range of changes to sinter mixes and sintering conditions are proposed to overcome the negative aspects of increased fuel reactivity while continuing to exploit the positives.

KEY WORDS: sintering; biomass; fuel reactivity; balanced productivity.

1. Introduction

Iron ore sinter, the dominant feed material for iron blast furnaces around the world, is formed by agglomerating fine iron ores, fluxes and solid fuel. Granules formed from the raw materials are layered into a thick bed (about 600 mm), the top of the bed is ignited and the flame front, fuelled with the solid fuel, is pulled through the sinter bed reacting and partially melting the granules, and forming a strong and porous sinter cake for feeding to the blast furnace.

Sintering is traditionally fuelled by coke breeze but improved efficiencies in coke production and alternative uses for coke breeze have reduced its availability for sintering. This scarcity creates opportunities to improve sintering by incorporating alternative fuels.

This paper outlines the experimental methods and results of iron ore sintering studies, conducted at CSIRO with alternative fuels and concludes with a discussion of opportunities and issues arising from the research.

2. Previous Studies

In 2004, CSIRO Minerals reported on sinter pot tests that showed how replacing coke with wood based charcoal fuels reduced greenhouse gas emissions while simultaneously reducing SO_2 and NO_x emissions by an order of magnitude and improving the balanced productivity of the sintering process. 1)

Productivity improvements suggested that fuel properties affected sintering time. A second CSIRO study reported in 2007 2) confirmed the relationship between fuel reactivity and sintering time, eliminated fuel size distribution as a cause for the changes in sintering performance leading to the development of a hypothesis that fuel reactivity effects sintering. This hypothesis was tested more rigorously in a 3rd CSIRO campaign and forms the basis of this paper.

3. Experimental

A sinter pot test program was devised to compare the sintering time and sinter properties of a typical iron ore blend fuelled with coke and more reactive fuels. Details of the iron ore blend, the experimental procedures and fuel characteristics are described below.

Previously unpublished gas analysis data from an earlier study 1) are also presented to explain the experimental results.

3.1. Raw Materials: Ores and Ore Blend

The ore blend used for the testwork was selected to simulate a typical Japanese Steel Mill (JSM) blend. The individual components of the ore blend and their chemical analyses are included in Table 1. Limestone and silica sand were used as the fluxing agents and return fines were added at a fixed rate of 30% (Ore Basis). The chemical composition of the fluxes is also included in Table 1. The fluxes were added so as to achieve a basicity level of 1.8 and fixed SiO_2 level of 5.00%.
3.2. Fuel Characterisation

The fuels chosen for pot tests included two coke breezes commonly used in sintering, two charcoal fuels and a black coal char specifically selected to assess sinter performance over a range of fuel reactivities.

3.2.1. Chemical Characterisation

The proximate and ultimate analyses of the fuels used for the test work are included in Tables 2a and 2b, respectively. Ash analyses for the fuels are included in Table 3. From the data it can be seen that:

- Charcoal fuels had higher fixed carbon content, higher volatile matter and higher calorific value than the coke or black coal char.
- Charcoal fuels had lower ash than the coke or black coal char.
- Charcoal fuel ash contained a significant proportion of calcium and iron oxides while the black coal char ash and the coke ash were dominated by silicates and aluminates.
- Coke and black coal char contained significantly greater concentrations of sulfur and nitrogen than the charcoal fuels.

Many of the fuel properties (e.g. fixed carbon, volatile matter and gross calorific value) are affected by pyrolysis/coking conditions while ash content and other chemical properties are related to the particular coal deposit or wood species and growing conditions. As knowledge of the impacts of fuel property on sintering improves there are likely to be significant opportunities to optimise fuel properties; through the supply chain as well as during preparation.

3.2.2. Physical Characterisation

Particle size distribution can have an impact on effective "reactivity" because it affects the surface area of the fuel (particularly fine particle size). In this experimental program the effect of size distribution of the fuels was minimised by dry screening the fuels at 3 mm and roll crushing and re-blending the over-sized material. Screening the 3 mm product at 1 mm produced fuel samples with similar size distributions, as illustrated in Fig. 1.

Porosity, surface area and specific reactivity are presented in Table 4. Porosity measurements were made using mercury intrusion porosimetry and can be compared with visual observations under the SEM (Fig. 2). Mercury porosimetry of the samples indicated that the charcoal sam-
samples were significantly more porous than the coke and coal char samples and the SEM images clearly show the remnant cellular structure of the black pine charcoal.

The reactivity of cokes and chars was measured using a technique developed by Roberts and Harris. One gram of each of the dried samples of particles (size range 0.6–0.8 mm) were heated in a carbon dioxide atmosphere at 700–800°C to produce carbon monoxide according to Eq. (1). Flow and temperature conditions were maintained such that the exhaust gas contained no more than 1% carbon monoxide to prevent reaction reversal. The rates and other kinetic parameters, such as the activation energy, were calculated and compared at a normalized temperature (e.g. 900°C).

\[
C_{\text{in coke or charcoal}} + CO_2 \rightarrow 2CO \quad \text{(1)}
\]

Results, summarised in Table 4, suggest that fuel reactivities vary significantly, from around \(1 \times 10^{-3} \text{ g/g/s} \) for coke and black coal char to \(419 \times 10^{-5} \text{ g/g/s} \) for the black pine charcoal.

### 3.3. Granulation

Granulation is a process used to prepare the iron ore, return fines, fluxes and fuel mix for sintering, and involves

| Fuel                                           | Porosity (%) | Specific Reactivity (x 10^2 g/g/s) | BET Surface Area (m^2/g) | Bulk Density (g/cm^3) | True Density (g/cm^3) |
|------------------------------------------------|--------------|------------------------------------|--------------------------|-----------------------|------------------------|
| Coke 1                                         | 16.5         | 2.2                                 | 16-17                    | 1.60                  | 2.06                   |
| Coke 2                                         | 16.8         | 1.09                                | na                       | 1.67                  | 1.95                   |
| Black Coal Charcoal                            | 23.0         | 1.55                                | 66                       | 1.43                  | 1.86                   |
| Industrially Produced Charcoal                 | 44.9         | 22.2                                | 915-950                  | 0.67                  | 1.85                   |
| Black Pine Charcoal                            | 58.0         | 419                                 | 228                      | 0.74                  | 1.76                   |

![Fig. 1. Size distributions of the fuel samples.](image1.png)

**Table 4.** Porosity, surface area and specific reactivity of the fuels.

![Fig. 2. SEM images of the fuels.](image2.png)
dry mixing of the raw materials in a rotating drum, followed a period of ‘granulation’ in which water is added and the rotation of the drum slowed to promote adherence of the finer raw material components to coarser ore nuclei.

The granulation step is critical to the overall sintering process as the granule shape and size distribution is a key determinant of green bed permeability. Capillary forces and liquid bridging are the principal bonding mechanisms in granulation and for a given ore blend, with sufficient nuclei to seed granule formation, granulation is optimized by varying the granule moisture.

Granulation trials in this project were based on a typical green mix consisting of approximately 3.8% fuel, 19% return fines, 7.5% moisture as a percentage of total dry mix, including the return fines (tdmb), with the balance in ore fines and fluxes. Raw materials were combined and granulated according to the methodology outlined previously.¹)

Granulating characteristics and permeability assessments indicated that granules formed with charcoal fuels were more permeable than those formed with coke and the permeability of the green bed formed with char granules was significantly less responsive to changes in moisture than those formed with coke.

Physically the charcoal particles handled very similarly to coke particles even though they were less dense. Visually the granules produced with the industrially produced charcoal or black coal char were indistinguishable from those produced with coke particles. Particles of black pine char did not incorporate into the granules as readily as the other fuels, possibly because they were elliptical rather than spherical.

3.4. Sintering Trials

Green granules of iron ore mix are sintered into a strong, porous cake by partial melting of the granules at high temperature. If the firing temperature is too low there may be insufficient heat to melt and bind the granules together, while too much heat can result in excessive melting and reduction in bed permeability leading to long sintering times and loss of productivity, as well as excessive magnetite formation.

Sintering trials were conducted on mixes with varying moisture and fuel level in order to maximise the sinter properties. The granules were charged to the sinter pot, ignited and fired to produce sintered aggregate.

The small-scale sinter rig (Fig. 3) consists of an ignition hood, pot, base and gas piping and suction system. The hood contains air and LPG gas burner ring mains and a suction thermocouple that measures ignition temperature. The ignition hood can be raised or lowered on to the base of a winch. The mild steel pot (150 mm depth) has a removable stainless steel grate in the base and is insulated to reduce heat losses. The base contains a pressure point (connected to a manometer) and a suction thermocouple for measuring sinter gas exit temperatures. The off-gas handling system includes a dust knockout pot, cooling sprays, pressure control valve, an in-line cartridge filter and fan connected to a stack.

Operationally, sintering is a two stage process involving ignition and sintering.

Ignition: The small-scale sinter pot was placed on to the base of the sinter rig, sealed with insulated gaskets and filled with a known mass of granules. The ignition hood, used to ignite the bed, was preheated for several minutes before lowering on to the pot and ignition of the granules initiated with 1961 Pa suction through the bed. Ignition temperature (1 300°C) was rapidly reached by adjusting the LPG/air flow rates and the suction was maintained during a 90 s ignition period.

Sintering: After ignition, the LPG/air flow was stopped, the burner was quickly lifted off the pot and the suction raised to 3 923 Pa. Ambient air was sucked through the ignited material and the flame front drawn through the granules. The flame front moves down through the bed. The gas outlet temperature was monitored and rose during the sintering process, reaching a maximum as the flame front burned through at the base of the bed. The sintering time was defined as the elapsed time between the commencement of ignition and the time at which the maximum off gas temperature was reached. The flame front speed was calculated to be the sinter bed thickness (150 mm) divided by the sintering time. Sintering time and the maximum gas temperature for a typical sinter test with each fuel type are presented in Figs. 4 and 5.

When cooled, the fired sinter cake was removed from the pot, weighed, sized, and sampled for chemistry. Tumble Index (TI) was also measured using a test methodology modified from ISO 3271¹ to allow for evaluation of small scale sinter samples.

Sinter test results (Table 5) show that a balanced sintering operation and target TI was achieved for all the fuel samples with the exception of black pine charcoal.

¹ I.e. a 1.5 kg sample of 40+10 mm sinter was tumbled in a 1 m diameter, 50 mm length drum for 200 revolutions at a speed of 25 rpm. The 65 wt%+5 mm target TI for the sinter product correlates to an acceptable industrial scale sinter plant value.
3.5. Sinter Chemistry

The chemical compositions of the sinters produced are presented in Table 6. The basicity of the samples was close to target with the exception of Coke 1 which was slightly lower than expected. The sinter chemistries were acceptable and representative of industrial sinter with the exception of the FeO analyses. Past operational experience has shown that higher fuel rates are required to achieve sinter strength and return fines balance in the small-scale sinter rig and it has been established that this results in sinters with FeO levels higher than would occur in large scale tests or at an industrial scale.

4. Discussion of Results

This section of the report compares the research with that of other researchers in the field. The results are discussed, and it is argued that solid fuel gasification has a significant influence on the temperature and width of the combustion zone, flame front speed and sinter strength. Finally, the potential for producing sinter at even higher rates of productivity with no penalty to sinter strength is discussed.

4.1. Sintering Time, Flame Front Speed and Fuel Reactivity

The pot tests described here show that fuel reactivity significantly effects sintering time (Fig. 4). For example, sintering with black pine char was 38% quicker than the average results for coke (i.e. 309 s cf. 501 s) while sintering with an industrially produced charcoal was 25% quicker than the average results for coke (i.e. 376 s cf. 501 s). Sintering time for fuels with low reactivities, i.e. black coal char and coke were similar.

Flame front speed (ffs) is calculated by dividing the green bed height (mm) by the time taken for waste gas temperature to peak (s) indicating burn-through, and is influenced by a number of inter-related factors. These include bed permeability, permeability of the reaction zone, fuel combustion rate, convective heat transfer rate, thermal load and oxygen availability. In addition, Loo emphasised the high temperature zone of sintering as the primary limiting factor for the process because gas velocity increases with thermal expansion and pressure drop increases with the square of gas velocity.

Loo proposed that flame front speed is a function of convective heat transfer through the bed, air flow rate and temperature differential between the flame front and the un-fired bed. As convective heat transfer and air flow rates are closely related under normal sintering conditions, Loo suggests that the heat transfer flux per unit area of layer below (Q) is UAT where Q = heat transfer flux per unit area of layer below (J/s), U = heat transfer coefficient (J/s/K), A = temperature difference.

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**Table 5. Summary of sinter test results.**

| Fuel                  | % H2O | % Fuel (tdmb) | Bulk Density (kg/m³) | Sintering Time (seconds) | Flame Front Speed (cm per min) | Return Fines Balance | Balanced Productivity (cm³/d) | Tumble Index (wt%) |
|-----------------------|-------|---------------|----------------------|--------------------------|-------------------------------|----------------------|-------------------------------|------------------|
| Coke 1                | 8.15  | 2.77          | 2140                 | 481                      | 1.9                           | 1.07                 | 16.9                          | 69.0             |
| Coke 2                | 7.00  | 2.56          | 2181                 | 521                      | 1.7                           | 0.95                 | 17.6                          | 72.1             |
| Black Coal Char       | 8.46  | 2.76          | 2247                 | 500                      | 1.8                           | 0.98                 | 18.2                          | 67.6             |
| Industrially Produced Charcoal | 7.97  | 6.00          | 1908                 | 376                      | 2.4                           | 0.95                 | 22.7                          | 67.5             |
| Black Pine Charcoal   | 10.02 | 4.14          | 1752                 | 309                      | 2.9                           | 0.97                 | 19.2                          | 61.2             |

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**Table 6. Sinter chemistry.**

|                  | Coke 1 | Coke 2 | Black Coal Char | Industrially Produced Charcoal | Black Pine Charcoal |
|------------------|--------|--------|-----------------|--------------------------------|---------------------|
| Fe                | 59.02  | 58.90  | 59.02           | 59.07                          | 59.23               |
| FeO               | 7.07   | 8.96   | 7.38            | 10.40                          | 10.20               |
| SiO₂              | 5.22   | 5.26   | 4.96            | 5.18                           | 5.18                |
| Al₂O₃             | 1.71   | 1.62   | 1.66            | 1.33                           | 1.53                |
| P                 | 0.49   | 0.048  | 0.043           | 0.049                          | 0.047               |
| S                 | 0.004  | 0.003  | 0.004           | 0.002                          | 0.002               |
| CaO               | 8.57   | 9.42   | 9.41            | 9.46                           | 9.18                |
| TiO₂              | 0.09   | 0.08   | 0.08            | 0.08                           | 0.08                |
| Mn                | 0.09   | 0.09   | 0.08            | 0.09                           | 0.09                |
| MgO               | 0.17   | 0.17   | 0.16            | 0.17                           | 0.18                |
| LOI100            | -0.57  | -0.79  | -0.51           | -0.79                          | -0.92               |
| Basicity          | 1.64   | 1.80   | 1.80            | 1.83                           | 1.77                |
perature differential between flame front and the unfired bed (K).

In the current study, the most reactive fuels generated the fastest flame front speeds and produced sinter in the shortest time. A linear relationship was found between flame front speed and natural log of fuel reactivity (Fig. 6) and a linear regression of the data showed that:

$$\text{ffs} = 0.2014 \cdot \ln(r) + 4.039 \quad (R^2 = 0.997) \quad \text{(2)}$$

where ffs = flame front speed (cm/min) and \( r \) = fuel reactivity (g/g/s).

Figure 6, a plot of flame front speed vs. fuel reactivity suggests that the rate controlling step shifts from a combination of chemical and diffusion control to being principally controlled by diffusion as fuel reactivity increases. It is possible that sintering reaction with reactive fuels are approaching the mass transfer limit for the sinter blends used in this study and it follows that further increases in sintering time could be achieved with reactive fuels by increasing the air flow rate across the bed or increasing the oxygen concentration in the sinter gas.

4.2. Reactivity and Flame Front Speed

Solid fuel reactivity, gasification and combustion are well researched topics. Key physical fuel properties that affect fuel reactivity include porosity, active surface area, gas diffusion rates, concentration of active sites and hydrogen and oxygen content. Of these factors, porosity has a direct effect on bulk density of the bed, bed porosity, permeability and heat transfer in the sintering process.

The more reactive fuels tested have a significantly lower density than less reactive fuels (Table 4) and consequently, reactive fuels like charcoal produced green granule beds with lower bulk densities (Table 5). For example, the industrially produced charcoal fuelled green granule beds are 12% lighter i.e. (1 908 kg/m³ cf. 2 161 kg/m³) and the black pine charcoal sinter mixes are 19% lighter than the average coke fuelled mixes i.e. (1 752 kg/m³ cf. 2 161 kg/m³). A significant proportion of this difference can be attributed to the very low bulk density of the chars and the green bed bulk density was exacerbated by the higher fuel requirement of these mixes.

Low bulk density granules have a lower thermal load and they will reach combustion temperatures more quickly than denser granules enabling the flame front to advance faster. Figure 7 illustrates the drastic reduction in flame front speed associated with increasing bulk density of the sinter bed. A similar finding was reported by Loo who suggested that flame front speed was ultimately determined by the time required for particles to reach combustion temperatures and that flame front speed would decrease when the bulk density of the bed increased.

4.3. Temperature of the Combustion Zone

Figure 8 is a schematic of the sintering reaction. It shows the ambient cool fresh air entering the top of the sinter cake. Initially the air exchanges heat with the sinter as it is pulled through the sinter bed by a suction fan. In the combustion zone, oxygen in the hot air combusts unburnt fuel and heat is carried down the bed by convection into the sintering zone by the combustion gasses.

Above the sintering zone, air drawn into the system is heated by hot sintered material before reacting with unburnt fuel. The reaction depletes \( O_2 \) and increases \( CO_2 \) in the gas stream and the moving gas stream transfers heat through the bed by convection.

Gas and solid temperatures continue to rise as the hot gas moves through the combustion zone of the bed and more fuel is burnt. The combustion zone is broader when unreactive fuels burn to extinction more slowly and melting of some phases begins at about 1 373 K.

Further down the bed, falling oxygen availability and high temperatures promote a series of endothermic reactions that cool the bed. Traditionally, endothermic reactions
associated with calcination of limestone and other fluxes and evaporation of water have been considered. However, endothermic gasification of reactive fuels is also likely to absorb thermal energy, possibly at temperatures as low as 900 K. Table 7 presents heat of reaction for significant reactions likely to occur during sintering.

When oxygen levels are low (i.e., in the pre-sintering, combustion, calcination and gasification zone, Fig. 8) it is likely that the fuel gasification is produced by the high temperature, endothermic Boudouard Reaction (i.e., the reaction of C and CO₂ to form CO) and that such a reaction:
- Occurs more rapidly with reactive fuels
- Reduces the amount of fuel available for sintering
- Reduces the thermal energy available for heating the green granules
- Cools the bed and narrows the thickness of the pre-sintering, thereby reducing pressure drop across the combustion zone and the sinter bed.

Figure 9 plots previously unpublished CO levels for sinters fuelled with reactive (red gum charcoal) and unreactive (coke) and a 50:50 red gum and coke blend. The increased CO levels associated with increasing reactive fuel supports the gasification hypothesis.

A considerable variation in the maximum off-gas temperature is noted and results suggest that this is not directly related to fuel level or total calorific value when fuel reactivity changes (Fig. 5 and Table 2a). While the highest off gas temperatures (791°C) was recorded for sinters produced with reactive industrially produced charcoal (6 wt% and a calorific value of 34.5 MJ/kg) the second highest breakthrough temperature (730°C) was recorded for the sinter fuelled with unreactive coke 2 at the lowest fuel rate (2.56 wt% and a calorific value of 28.5 MJ/kg). At the lower end of the breakthrough temperatures, sinters produced from the reactive Black Pine Char and less reactive Coke 1 recorded the lowest maximum off-gas temperatures (approximately 570°C and 605°C, respectively).

### 4.4. Tumble Index

Tumble Index (TI) is an important measure of sinter strength and it is broadly accepted that TI increases with the volume of melt phase produced during sintering. Results presented in Table 5 shows that Tumble Index of the sinter product falls as fuel reactivity increases, a finding that suggests less melt is formed when fuel reactivity increases in sintering.

It is anticipated that the reduction in TI is primarily due to a reduction in time above melt-forming temperatures that arises as a consequence of increased flame front speed and a narrowing of the combustion and sintering zones (see Sec. 4.2).

Furthermore, the reduction in bulk density associated with increasing fuel reactivity is also likely to reduce sinter strength, as a more porous sinter cake is produced.

### 4.5. Moisture Level

Moisture level is a well known and important factor in optimising green granule bed permeability. Too little moisture restricts green granule growth while too much water can saturate and collapse the lower bed and result in decreased permeability.

In this study, no evidence that flame front speed was retarded by moisture addition was observed. Reactive fuels, particularly those formed by conventional pyrolysis of biomass tend to be very porous and require significantly higher moisture levels in granulation than less reactive fuels and at its most extreme, an increase in flame front speed of nearly 40% when moisture levels increased by 40% was observed i.e. Coke 2 (7 wt% H₂O) compared with Black pine charcoal (10 wt% H₂O).

### 5. Further Improvement of Sintering Performance

Significantly increased flame front speed and the likelihood of increasing productivity by more than 1/3 is the principle advantage of increasing fuel reactivity. Sourcing the fuel from a sustainable agricultural system will also allow a wide range of environmental, social and industrial benefits that include a closed carbon cycle as well as reduction of nitrous and sulfur oxides by an order of magnitude.

These benefits could significantly improve the contribution of the iron and steel industry to sustainable development.

Poor fuel utilization and reduced TI are issues that need to be addressed to promote the use of reactive biomass fuels in the sintering process. However, it is anticipated that increasing the availability of oxygen, by oxygen enrichment or increased air flow rates, would increase the oxygen potential of the off-gas and prevent gasification of C with CO₂. Returning CO levels to close to zero would simultane-

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Table 7. Heats of reaction for significant reactions during sintering.

| Reaction | Estimated Heat of Reaction at 25°C (kJ/mol) |
|----------|---------------------------------------------|
| C₁₀₈ + O₂(10) → CO₂(10) | -405.9<sup>11</sup> |
| C₁₀₈ + ½ O₂(10) → CO(10) | -123.1<sup>11</sup> |
| C₁₀₈ + CO₁₂ → 2CO(10) | -159.7<sup>11</sup> |
| C₁₀₈ + H₂O → CO₂ + H₂(10) | -118.9<sup>11</sup> |
| CaCO₃(10) → CaO(10) + CO₂(10) | 183.0<sup>9</sup> |
| 2Fe₂O₃(10) → Fe₃O₄(31) + H₂O(10) | 53.2<sup>9</sup> |

<sup>11</sup> Testing was conducted in a pilot scale sinter rig in a cylindrical pot (300 mm diameter×500 mm depth). Ignition was conducted at 1300°C for 90 s with a suction pressure of 7845 Pa, and sintering conducted at 15 691 Pa. Fuel rates were 6.09% for 100% coke, 7.64% for 50% coke/50% charcoal and 9.23% for 100% coke.
ously increase the heat available for sintering and improve fuel utilisation and further increase flame front speed.

The improved viability of heat through oxygen enrichment or increased air flow rates may partially reduce the need for additional melting. However, it is considered most likely that TI could be increased by the addition of more reactive iron oxides (e.g. goethitic, porous hematite ore types and martitic ores) or a reduction in unreactive ores (e.g. dense hematite).

On-going iron ore sintering research at CSIRO addresses these issues so that integrated steel mills around the world can continue to improve energy efficiency, reduce global green house gas emissions, and contribute towards a sustainable future.

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