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Coal and Biomass based Fuels in Rural India: Emissions and Possibility of their Control

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Emissions from solid fuels used in the rural areas have been recognized as a major environmental and health hazard in India and many other developing countries. Use of these fuels is expected to continue in near future, and it is necessary to find techno-economically feasible solutions for emission control. Monitoring of CO and PM emissions as well as temperature measurements for rural cook-stoves have been done at actual emission sources, for some common solid fuels used in India. Based on these data, a catalytic approach for emission control from rural cook-stove has been explored by study of catalytic materials for CO oxidation.

1. Introduction

Indoor air quality is influenced due to emissions from various sources like new materials, and products of building materials, insulation, bonding resins, fabrics, cleaning materials, personal care products, pesticides, and in addition, from the combustion process of cooking, fuel heating and tobacco smoking. However, emissions from solid fuels used in the rural areas has been recognized as a major environmental and health hazards in India and many other developing countries, especially among the poor people. According to a previous WHO report[1], among the people living on less than one dollar a day, over 75 per cent of them use solid fuel for cooking, in India. Every year smoke from burning solid fuels in the home kills a large number of children under five years. The World Health Organization (WHO) estimated that about 420 thousand premature deaths were caused annually in India by indoor solid fuel air pollution[2]. In this way, the most significant issue that concerns indoor air quality in household environments of developing countries is that of exposure to pollutants released during combustion of solid fuels including biomass (wood, and crop residues), dung or coal, frequently used for cooking and heating. Majority of rural household burn these simple solid fuels in inefficient earthen or metal stoves, or use open pits in poorly ventilated kitchens, resulting in very high concentrations of indoor air pollutants. It is estimated that use of open fires with these fuels exposes nearly 2 billion people with enhanced concentrations of particulate matter and gases, up to 10-50 times and even higher than health-based guideline values available for typical urban outdoor concentrations[3]. Although biomass makes up only 10-15 percent of total human fuel use, since nearly half the world's population cooks and heats their homes with biomass fuels on a daily basis, indoor exposures[4] likely exceed outdoor exposures to some major pollutants on a global scale[5]. Such exposures have serious health consequences for household members, particularly for the women involved in cooking and young children spending time around their mother. The traditional cook-stoves used for cooking are not energy efficient, which leads to incomplete combustion. The incomplete combustion of biomass and other solid fuels releases complex mixture of pollutants, which include suspended particulate matter (SPM), carbon monoxide (CO), polyorganic material (POM), poly aromatic hydrocarbons (PAH), formaldehyde etc. These fuels may also contain intrinsic contaminants such as sulphur, trace metals, etc.

Acute and chronic respiratory diseases, which are causally linked to exposure of indoor air pollution in developing countries, are the leading cause of global burden of disease. Indoor air pollution is responsible for more than 1.6 million annual deaths and 2.7 % of global burden of diseases[6]. Acute respiratory infections (ARI) are the leading cause of the global burden of disease and account for more than 6 % of the global burden of disease and mortality, mostly in developing countries[7]. Between 1997 and 1999, acute lower respiratory infections (ALRI) were the leading cause of mortality from infectious diseases, with an estimated 3.5-4.0 million deaths worldwide[7-9]. According to a study undertaken on the correlation of eye and respiratory symptoms among those exposed to wood smoke emitted from indoor cooking, concluded that these are significantly associated with wood use[10]. Another study showed an independent effect of indoor air pollution on birth weight[11]. Several studies have been undertaken in various Indian locations mainly in slums or rural households to estimate various indoor air pollutants like particulates, VOCs, SO_x, NO_x, CO and CO_2[12]. Various other studies also looked into health aspects of women exposed during cooking in kitchens. In India, many exposure related studies focused on bio-fuel combustion for cooking in rural and slum areas[13,14]. Similar studies have also been carried out in other parts of the world, where health concerns were given a primary focus[15,16]. Poor living conditions have been identified to significantly aggravate the impacts of indoor air pollution.

Exposure to indoor air pollution, especially to particulate matter, from the combustion of biofuels (wood, charcoal, agricultural residues, and dung) has been implicated as a causal agent of respiratory diseases in developing countries[16,17]. This association, coupled with the fact that globally more than 2 billion people rely on biomass as the primary source of domestic energy, has put preventive measures to reduce exposure to indoor air pollution high on the agenda of international development and public health organizations[7,18].

Indoor biomass combustion often generates huge amounts of indoor air pollutants and a lot of efforts have been devoted for measurements of these pollutants. Measurements of 24 h mean indoor levels of PM_{10} (particles with mass median aerodynamic diameter of <10 μm) have been reported between a very wide range of 300 and 30,000 μg/m³ and CO between 2 to as high as 500 ppm. These levels are comparatively 2-200 times higher than the U.S. Environmental Protection Agency regulations for outdoor air pollutants[19-21]. PM_{10} can bypass the filtering system of the nasal and oral cavity to either deposit on the mucosa of large- and medium-sized airways (coarse PM) or deposit deep in the alveoli (fine PM), and thus is able to affect respiratory health[22]. Human exposure to high levels of outdoor PM_{10} is independently correlated to lung cancer and cardiopulmonary mortality[23]. According to one estimate, the global burden of disease, attributed to indoor air pollution from biomass combustion can account for approximately 3 % of worldwide disability- adjusted-life-years (DALY) lost. This is predominantly due to pneumonia in children and chronic obstructive pulmonary disease and lung cancer in women[16]. The gravity of the problem can also be gauzed with the fact that prolonged exposure to even very low concentrations of these pollutants can not be considered as safe in health point of view[24,25]. It is also important to mention here that in addition to severe health impacts, the environmental impacts of solid fuel emissions are now better understood and recognized as a significant contributor with considerable greenhouse impacts per unit energy delivered. This is mainly from the emissions of nitrous oxide, methane, CO_2 and other important greenhouse pollutants, although not a large overall contribution as compared to other sectors. In addition, solid fuel combustion emits significant amount of other shorter-lived pollutants like black carbon that has been correlated to reduction of crop yields and to accelerated melting of glaciers.

Considering the slow decline in the ratio of the Indian population using solid fuels (about 0.7 % per year between1998-1999 and 2005-2006[27]) and the population growth expected for the country (currently, about 1.5 % per year), the overall number of users of solid fuel will not change significantly in the next decade, baring some major intervention by the government. This is also closely related to the income level of rural population in India. In this way, the provision of clean cooking energy for the poor people continues to be a major challenge, in the era of renewable energy, which is recognized with practically no environmental impacts. Considering the projected use of solid fuels as the cooking energy source for a

Table 1 Distribution of household cooking fuel use in India in 2005 (from reference 26)

Fuel	Urban (%)	Rural (%)	National (%)	Household (%)
Dung	2.80	14.40	10.60	24.00
Biogas	0.50	0.40	0.50	1.10
Crop waste*	1.30	13.00	9.20	21.00
Wood	22.00	61.80	48.70	110.50
Charcoal	0.50	0.30	0.40	0.90
Coal	4.30	0.80	1.90	4.30
Kerocene	8.20	0.80	3.20	7.20
LPG	58.70	8.20	24.70	56.00
Elec.	0.90	0.10	0.40	0.90
Others	0.80	0.20	0.40	0.90
Biomass Total	27.00	90.00	69.00	156.00
Solid Fuel total	31.00	90.00	71.00	160.00

*includes the category entitled "straw/grass/shrubs"

majority of Indians, there remains the need for ensuring the emission control from this prominent source. In the present study, indoor emission monitoring of CO and PM emissions have been carried out at source, mainly to check the CO and PM concentrations, in a view to design the simulated conditions for catalyst evaluations. Work has been initiated to explore the catalyst based solutions as well as on development of improved cook-stoves, for the control of emissions from these solid fuels used for the cooking and heating requirements of rural households. However, considering the various constraints including the cost, it will not be possible to use noble metal based catalysts for the target application.

Non-noble metal based perovskites type catalysts seems to be the potential candidates for this application, mainly due to their thermal stability, tailoring capabilities and low-cost. $LaCoO_3$ is an extensively reported catalyst for CO oxidation and the activity range was found be 280-350 °C depending on synthesis procedures and evaluation conditions[28-31]. However, the reported articles do not study the catalytic activity in presence the simulated exhaust composition of a rural stove. As the present work is targeted for control of CO emissions from solid fuel combustion, it is very important to study the active compositions for CO oxidations such as $LaCoO_3$ under the actual conditions in rural homes. Therefore, this composition was selected as a typical low cost catalyst. Additionally improved cook stove is also being developed with effective insulation to enhance the combustion efficiency of the soild fuels used in the rural areas of India as well as in many other developing countries.

Experimental

2.1 Technical approach

Under this research, attempt is being made to develop catalyst coated mesh and an improved design of cook stove, which can effectively control the emissions from these solid fuels. This involves development of low cost catalysts, their characterization and laboratory evaluations for their catalytic activity. The selected catalysts are proposed to be coated on suitably designed mesh for final applications.

2.2 Catalyst synthesis

$LaCoO_3$ was synthesized by combustion method using urea as fuel. Nitrate salts of metals precursors and urea were taken in 1:2 molar ratio, and dissolved in requisite quantity of DI water. The solution so obtained was heated at 100 °C till foamy mass is obtained. The material was homogenized and then calcined at 750 °C for 8 h. The catalyst thus synthesized was coated on cordierite powder, following a dip coating method. The loading of catalyst was optimized at 5 wt % with respect to support weight. The catalyst was pre-treated in He flow as described above, and catalytic activity was measured as a function of temperature.

2.3 Catalyst characterization

The X-ray diffraction analysis was carried out using Rigaku Miniflex-II Instrument with Cu Kα1 radiation at 30 kV and 15 mA with monochromator. The samples were scanned in 2θ range of 10-80° with scanning speed of 3 °C per min. Diffraction peaks were compared with standard database reported in the Joint Committee on Powder Diffraction Standards (JCPDS) cards. $LaCoO_3$ compound synthesized can be indexed to rhombohedral system, corresponding to JCPDS card number 84-0848. The morphological details of the material were studied by scanning electron microscopy (SEM), while the BET surface area was measured on a Quantachrome equipment. The sample was degassed at 250 °C before the nitrogen adsorption experiment.

2.4 Catalytic activity evaluations

The evaluation of the catalyst was carried out by using a steady state, fixed bed type catalyst evaluation assembly, equipped with mass flow controllers and high temperature reactors. 100 mg of powdered catalyst was loaded in a fixed bed reactor and pre-treatment was carried out in a continuous helium flow at 300 °C for 1 h for desorption of adsorbed gases. The feed composition containing 500 ppm CO with 10 % oxygen and balance nitrogen was passed through the pretreated catalyst bed from inlet of reactor and the outlet gas stream was monitored using a Gas Chromatograph equipped with thermal conductivity detector (TCD). The catalyst activity was evaluated at different temperatures and under different conditions of space velocity. A schematic of the catalyst evaluation set up is given in Fig. 1.
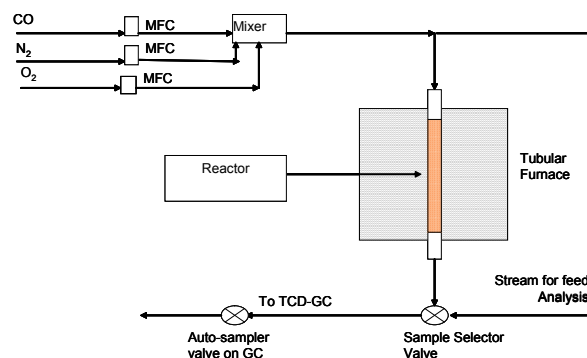

Fig. 1 Steady state, catalyst evaluation assembly.

3. Results and discussion

The emission monitoring data from rural households suggest CO emissions in a wider concentration range, depending on many factors like the fuel used, fuel condition, stove type, ventilation, season etc. The CO concentration at the source (cook-stove exhaust) was mainly in the range of 100-400 ppm, however, many reports suggest even higher concentration of CO under different conditions for different cook-stoves. A higher concentration of 500 ppm was therefore, considered for catalytic evaluations, considering any failure mode.

The temperature profile studies of cook-stove exhaust and cook-stove walls, suggest the temperature availability in the range of 200-600 °C, which is very suitable for catalytic activity. Fig. 2 gives the temperature profile data, where, F.T, P.T. and I.W.T. represent fuel temperature, pot temperature and its inner wall temperature respectively.
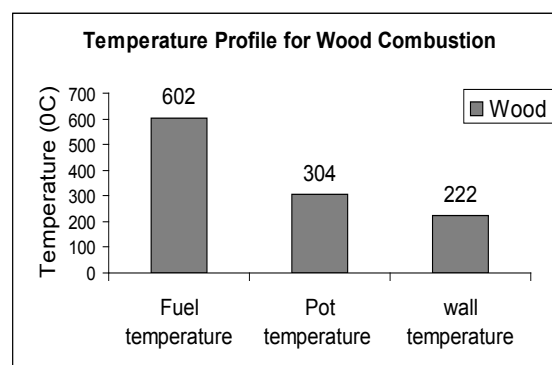

Fig. 2 Temperature profile results for wood as a fuel.

3.1 Characterization of catalyst

The XRD pattern for the synthesized catalyst is given in Fig. 3, which suggest the formation of crystalline $LaCoO_3$ phase. $LaCoO_3$ compound synthesized can be indexed to rhombohedral system, corresponding to JCPDS card number 84-0848. The combustion synthesis has resulted in highly porous material with voids as seen in SEM pictures (Fig. 4). This is due to the sudden release of gases during combustion synthesis. The catalyst shows surface area of 10.2 m^2/g, which is quite high for the perovskite phase.
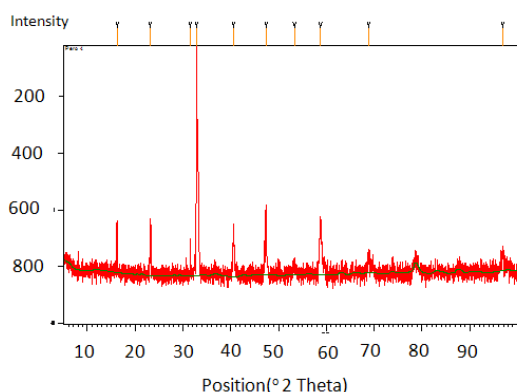
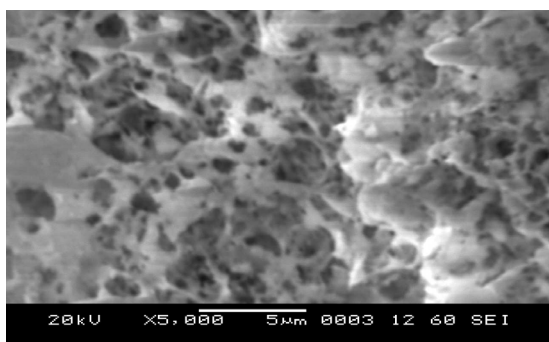**Fig. 3** X- Ray diffraction pattern for perovskite catalyst.

Fig. 4 Scanning Electron Micrograph for $LaCoO_3$.
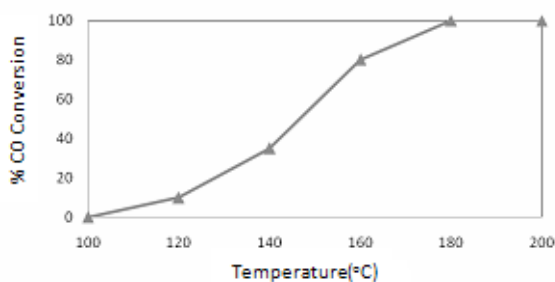

Fig. 5 CO conversion as a function of temperature (Conditions: CO = 500 ppm + 10 % O_2 + balance N_2, Flow rate = 100 ml / min, Catalyst wt. = 100 mg).

Table 2 CO oxidation activity results for catalyst coated ceramic honeycomb

Temperature (^{o}C)	% CO oxidized
350	100
320	100
245	100
224	100
189	85
164	73
120	30
70	0

3.2 Catalytic activity results

The Fig.5 provides the catalytic activity data, confirming the 100 % CO conversion at 180 ^{o}C and substantial conversion even below that temperature. A stream of the actual exhaust was also used for catalytic activity of coated catalyst, which provided the similar results on CO oxidation. Further the catalyst coated ceramic honeycomb was also tested for CO oxidation, which once again suggested the similar results under the actual conditions of practical use. Table 2 presents the CO oxidation activity results observed with catalyst coated ceramic honeycomb. These experiments under the simulated and actual conditions of cook-stove applications infer the initial feasibility of catalytic approach for CO emission control from the solid fuel based cook-stoves. We have now developed a range of other low cost catalysts and their testing are under progress. It will be however, important to study the long term stability of select catalysts as the exhaust of solid fuels often contains a large number of so called "dirty compounds" with respect to catalyst stability.

4. Conclusion

The present study suggests the adequate exhaust temperature at cook stove and possibility of catalytic intervention for control of CO emissions. These fuels produce extremely high emissions as they are combusted in highly inefficient cook-stoves. This leads to severe health and environmental impacts inclduing the high health burden mainly with respect to women and children. The perovskite type catalyst has been synthesized owing to its low cost. The catalyst has been characterized and supported on ceramic material, cordierite. The catalyst evelution data confirm the high activity of catalyst and effective conversion of CO even under the realistic conditions of its use. Further work is required mainly to study the durability of catalyst under the harsh conditions of solid fuel exhaust. It is however, required to continue exploring various options for emissions control from cook-stove as it is envisaged that use of solid fuels will continue in near future.

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