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AIP | Journal of Renewable and Sustainable Energy

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Citation: J. Renewable Sustainable Energy **5**, 039101 (2013); doi: 10.1063/1.4807607
View online: http://dx.doi.org/10.1063/1.4807607
View Table of Contents: http://jrse.aip.org/resource/1/JRSEBH/v5/i3
Published by the American Institute of Physics.

JOURNAL OF RENEWABLE AND SUSTAINABLE ENERGY 5, 039101 (2013)

Alumina reduction by laser sustained plasma for aluminum-based renewable energy cycling

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(Received 30 January 2013; accepted 29 April 2013; published online 21 May 2013)

A novel alumina (Al_2O_3) reduction technique for a renewable energy cycling system based on aluminum is proposed. Al_2O_3 powder was fed into laser-sustained plasma and thermally dissociated. The produced Al was expanded to supersonic speeds through a nozzle. From the Al and argon line distributions in the flow direction, it was found that Al remained in the dissociated state. A water-cooled copper tube was inserted in the flow to collect Al. X-ray analysis indicated that elemental Al was observed on the surface of the tube. The maximum value of the estimated reduction efficiency was 5%. *© 2013 AIP Publishing LLC*.
[http://dx.doi.org/10.1063/1.4807607]

Apart from concerns over global warming, renewable energy sources such as solar and wind power have received increasing attention because of the impact of the Fukushima Daiichi nuclear disaster. Since electric power generation from renewable sources is subject to weather and seasonal variations, the development of large-scale storage systems has become crucial for maintaining stable power supply. The prototypical storage system, Li-ion batteries, is already in use in household applications. Because of their high energy density and reliance on rare earths, Li-ion batteries are, however, not suited for power plants that produce several MW to GW of electricity. Hydrogen, another major storage material, suffers from the same disadvantages even though high-pressure tanks have been introduced.

In the search for viable alternatives, we have focused on aluminum as the basic energy storage material. It is well known that Al production consumes a considerable amount of electric energy, and indeed common beverage cans are sometimes referred to as "cans of electricity." On the other hand, however, this implies that Al is able to store energy as chemical potential. A comparison of energy densities of commonly used energy storage materials is listed in Table I. The energy density of Al is a few orders of magnitude higher than that of Li-ion batteries or hydrogen and almost equals that of fossil fuels. In addition, Al is stable under standard conditions and shows negligible degradation with aging. This allows storing the renewable energy in aluminum on a suitable site and transporting it to a consuming region, where electricity is extracted by a thermal power plant or a future Al-air battery.[1] Figure 1 shows the concept of our cycling system for renewable energy based on Al. The cycle makes it not only possible to store the electricity generated by renewable energy sources but also reduces power generation costs. The large-scale storage of renewable energy makes it possible to generate

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TABLE I. Energy storage density of common energy storage materials.

Material	kJ/g	kJ/cm^3
Aluminum	15.5	41.9
Coal	41.3	57.8
Oil	60.4	47.2
LNG	76.9	33.7
H_2 (100 MPa)	0.60	0.05
Lead-acid battery	0.13	0.29
Li-ion battery	0.36	0.9

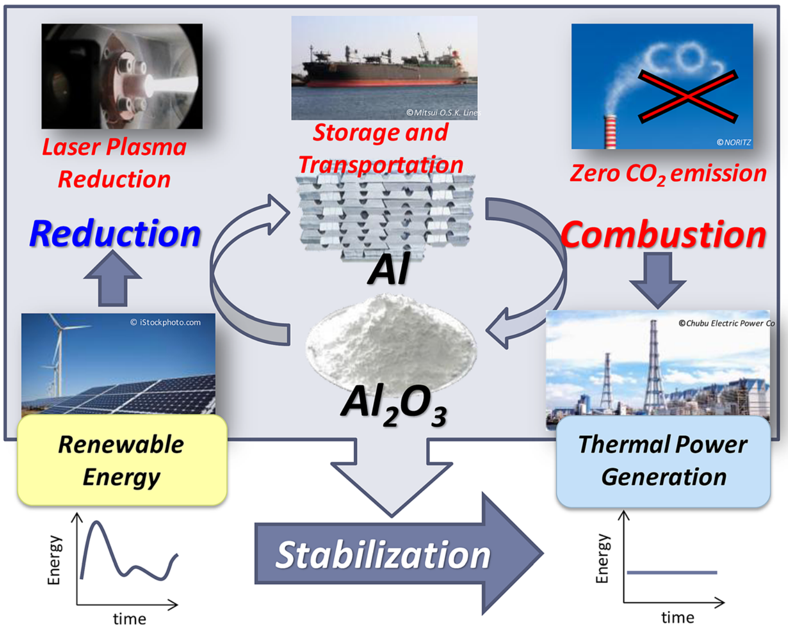

FIG. 1. Renewable energy cycling system based on aluminum as a storage material.

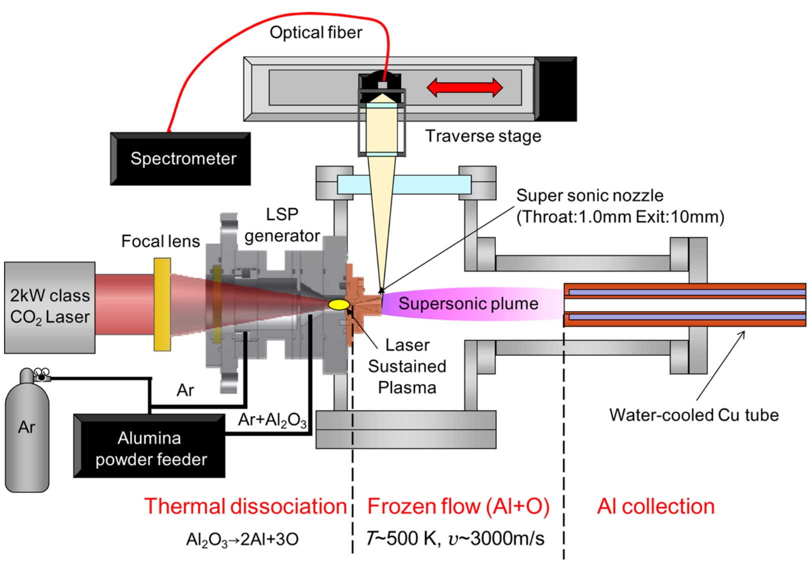

FIG. 2. Schematic representation of the experimental setup. The system consists of three parts, which are involved in thermal dissociation, frozen flow, and aluminum collection.

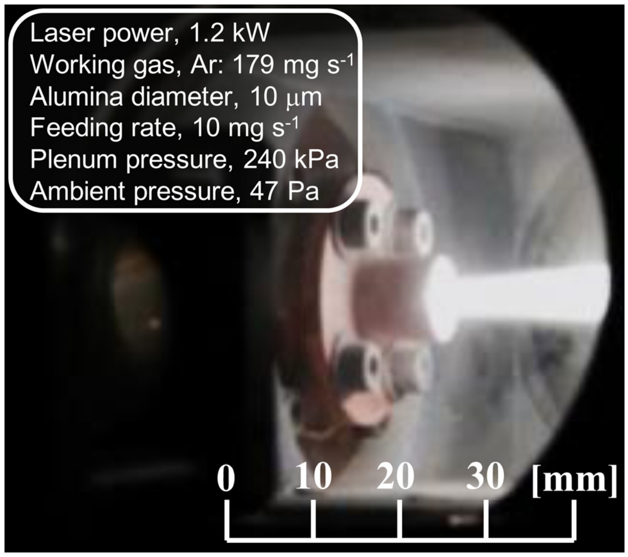

FIG. 3. Photograph of the LSP in operation. The LSP is stable and expands through the nozzle.

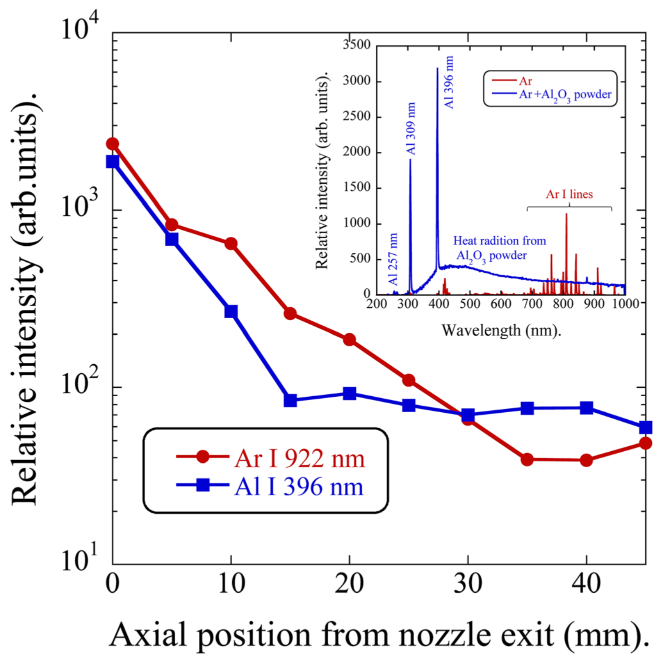

FIG. 4. Emission spectra at the nozzle exit (inset) and emission line distributions in the direction of the nozzle axis.

power in deserts near the equator, where the amount of solar radiation is 2–3 times higher than in Japan or Europe, while vast areas are available at minor land costs.[2]

A key technology in the cycling process is the reduction of alumina (Al_2O_3). The conventional electrolysis method (i.e., the Hall-Heroult process) is widely used in industry, but is not suitable for application in a renewable energy-cycling system, as it emits considerable amounts of CO and CO_2, which results from the use of carbon electrodes as the reducing agent.

In this study, we employed laser-sustained plasma (LSP) to reduce Al_2O_3. The use of stationary plasma for alumina reduction enables a higher processing capacity than direct heating by a focused laser beam, as reported by Yabe *et al.*[3] LSP, as a plasma production method, has the advantage of electrode-less heating and operation at atmospheric pressures. In the

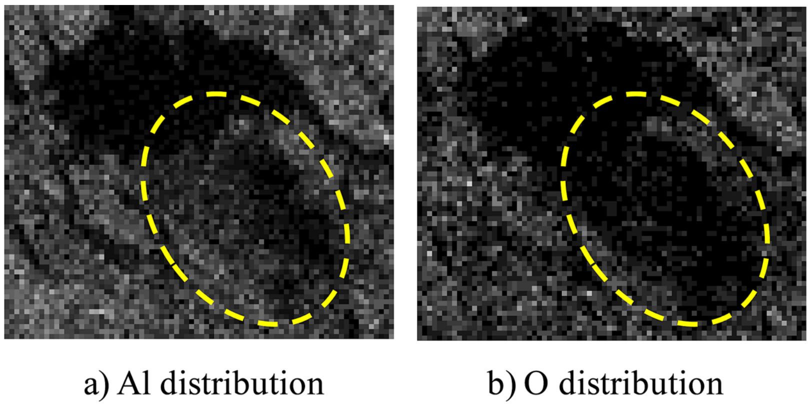

FIG. 5. Aluminum (a) and atomic oxygen (b) distribution in the copper collection tube as observed by EDS. White color indicates the presence of the element under investigation.

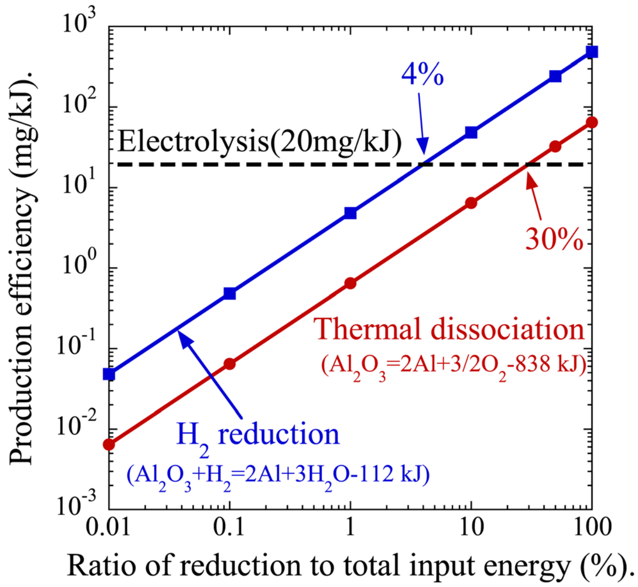

FIG. 6. Aluminum production efficiency of the LSP process as a function of the ratio of reduction energy to total input energy. The efficiency of the Hall-Heroult electrolysis process is also shown.

conventional arc-discharge process, dissociated oxygen causes severe erosion of the electrode, resulting in short operation times.[4] Inductively coupled plasma, a conventional electrode-less heating process, is limited to operation at pressures below the atmospheric pressure; this results in low processing efficiency because the thermal conductivity depends on the pressure.[5]

A schematic of the experimental setup is shown in Figure 2. The system consists of three parts, which are involved in thermal dissociation, frozen flow, and Al collection (see Ref. 6 for details on the LSP generator) and is operated as follows. First, the Al_2O_3 powder is fed into the LSP at atmospheric pressure. Although the peak temperature of the LSP core (where most of the laser beam is absorbed) is ca. 9000 K, the surrounding region is ca. 4000 K.[6,7] Nevertheless, a temperature of 4000 K is high enough to dissociate Al_2O_3. The produced Al is accelerated to supersonic speeds by a convergent-divergent nozzle to prevent Al from recombining with oxygen. After expansion, the plasma is cooled to ca. 500 K and the velocity is increased to ca. 3000 m/s.[8] This type of supersonic flow, a "frozen flow," is generally in non-equilibrium, and Al and atomic oxygen exist in the dissociated state. Al is then selectively collected. A water-cooled copper tube is used as a collector, preventing formation of a shock layer, which could lead to an increase in pressure and a return to equilibrium, resulting in reformation of Al_2O_3.

Figure 3 shows a photograph of the LSP in operation. The LSP could be stably maintained and expanded through the nozzle, even when Al_2O_3 powder was being fed. A typical emission spectrum at the nozzle exit as well as the emission intensity distributions in the flow direction are shown in Figure 4. Feeding alumina powder gave rise to the appearance of strong Al lines. The observed broad continuum band spectrum is presumably heat radiation originating from un-melted Al_2O_3 powder. The emission intensity distributions at the 396 nm (Al I) and 912 nm (Ar I) lines show a concurrent decreasing trend. The emission intensity depends on the number density and the excitation temperature. Considering that the argon number density in the flow region is almost constant, the decrease is mainly thought to result from a decline in excitation temperature. Hence, this result shows that the Al number density is constant as well and, thus, that the flow in this region is frozen. Figure 5 shows the Al and atomic oxygen distribution on the surface of the collection tube as measured by energy dispersive X-ray spectrometry (EDS). As is clear from this figure, Al regions can be observed without atomic oxygen, indicating that the Al observed is reduced Al rather than Al_2O_3.

Finally, the reduction efficiency of the system was estimated. In our previous research, the energy conversion efficiency of the LSP was investigated,[9] where it was found that more than 90% of the power laser beam is absorbed by the LSP. The absorbed energy is used for heating of the working gas as well as its ionization, the heating and dissociation of Al_2O_3, and for thermal energy that is lost in the wall through conduction and radiation. Although the energy allocation depends on operating conditions such as working gas flow rate, laser beam intensity, and plenum pressure, the percentage that is used for heating Al_2O_3 is estimated to be 10%–50% (assuming a relative percentage of 10%–30% for heating the working gas, 10%–20% for ionization, 20%–30% for thermal loss, and 10% for other processes, including transmitted beam energy). Figure 6 shows the production efficiency, defined as Al production per energy unit, as a function of the ratio of reduction energy to total input energy. Here, the electrolysis value is that of a typical Hall-Heroult process. Since the beam production efficiency of the CO_2 laser is 20%, the fraction that contributes to Al_2O_3 heating is 2%–10%. In addition, taking into account the specific and latent heat of Al_2O_3, the fraction used for reduction is 1%–5%. This value is one order of magnitude lower than that of electrolysis. Assuming the use of a high power diode laser, of which the plug-in efficiency can be 60%, the resulting efficiency would approach that of electrolysis. Alternatively, the efficiency can be increased by using hydrogen as the reducing agent. However, since hydrogen production requires electric energy as well, its contribution to the total efficiency of the process should be investigated.

In summary, we have proposed a novel alumina reduction technique for a renewable energy cycling system based on aluminum. Alumina powder was successfully reduced by laser sustained plasma, and the maximum reduction efficiency was estimated to be 5%. We expect that the efficiency can be increased to 15% by using a high-power diode laser and even further by employing hydrogen as the reducing agent.

This research was partially supported by the Ministry of Education, Science, Sports and Culture, via a Grant-in-Aid for Young Scientists (A) (No. 22686079) in 2010.

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