Research on Microsphere Fabrication and Coolant Loop Technology for Lithium Oxide Moving Bed Fusion Power Plant

T.A. Thornton, D.C. Schluderberg, D.K. Sze, and F.A. Zenz

February 1979

UWFDM-293

FUSION TECHNOLOGY INSTITUTE
UNIVERSITY OF WISCONSIN
MADISON WISCONSIN

Research on Microsphere Fabrication and Coolant Loop Technology for Lithium Oxide Moving Bed Fusion Power Plant

T.A. Thornton, D.C. Schluderberg, D.K. Sze, and
F.A. Zenz

Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

February 1979

UWFDM-293

RESEARCH IN MICROSPHERE FABRICATION AND COOLANT LOOP TECHNOLOGY FOR LITHIUM OXIDE MOVING BED FUSION POWER PLANT

T.A. THORNTON and D.C. SCHLUDERBERG
Babcock and Wilcox, Lynchburg VA 24505

D.K. SZE
University of Wisconsin, Madison WI 53706

F.A. ZENZ
Manhattan College, Bronx, New York NY 10471

February 1979

UWFDM-293

Fusion Research Program
University of Wisconsin
Nuclear Engineering Department
Madison WI 53706 U.S.A.

RESEARCH IN MICROSPHERE FABRICATION AND COOLANT LOOP TECHNOLOGY FOR LITHIUM OXIDE MOVING BED FUSION POWER PLANT

T. A. THORNTON and D. C. SCHLUDERBERG

Babcock and Wilcox, Lynchburg, Virginia 24505, USA

D. K. SZE

University of Wisconsin, Madison, Wisconsin 53706, USA

F. A. ZENZ

Manhattan College, Bronx, New York City, New York 10471, USA

A gravity-flow bed of lithium oxide microspheres has been proposed as the tritium breeding and blanket cooling medium for both magnetic confinement and inertial confinement controlled fusion reactors using the D-T cycle. The use of the lithium oxide moving bed could help eliminate MHD, corrosion, and materials compatibility problems associated with the use of liquid lithium. Adequate particle transport mechanisms which alleviate particle attrition and erosion seem attainable. Moving bed heat transfer coefficients an order of magnitude greater than for fluidized beds are indicated. The fabrication of microspheres is complicated by the aggressive caustic nature of the lithium oxide but should be possible for commercial-scale production.

INTRODUCTION

The blanket region of a controlled thermonuclear fusion (CTF) reactor must serve the following two primary functions if the principal core energy source is the deuterium-tritium fusion reaction: 1) breed tritium, which does not occur in nature and has no other large-scale source, for recycle as fuel for the primary fusion reaction, and 2) provide a capture, transport, and recovery mechanism for the sensible heat generated as a result of the 14 MeV neutron flux emanating from this fusion reaction. Depending on the chosen blanket materials and the design philosophy, these functions could be accomplished separately — for example, lithium/lead as tritium breeder/neutron multiplier and water as blanket coolant as in the NUWMAK [1] design -- or combined in a single tritium recovery and cooling loop such as will be discussed here. The use of a single tritium breeding and reactor coolant loop significantly simplifies the blanket design but introduces many problems in the areas of materials selection, non-conventional heat transfer and transport, and perhaps more complicated tritium migration and recovery. The use of a gravity-circulated particulate solids blanket design for both Tokamak [2] and laser-fired inertial confinement [3] reactors has been proposed previously by researchers at the University of Wisconsin. These schemes involve the use of a moving bed of lithium oxide (Li_2O) microspheres as reactor blanket material and as heat transfer and transport medium whereby a number of difficult problems associated with blanket and coolant loop designs utilizing such media as liquid metals (lithium) or helium are potentially avoided. Some of the advantages which may accrue to such a design scheme utilizing a dry granular medium would include:

- elimination of MHD effects in the blanket of a magnetic confinement-type fusion reactor
- ability to de-couple or separately cool the first wall of the reactor by designing the coolant channel to minimize heat transfer between the microsphere bed and the wall
- potential capability for enhanced energy storage through the maintenance of a static storage silo of microspheres
- the high lithium atom density of lithium oxide (comparable to that of liquid lithium) in relation to other solid lithium compounds could eliminate the need for a neutron multiplier in the blanket and minimize the size of the blanket region.
- alleviation of materials compatibility problems with respect to liquid lithium as coolant/breeder.

The tritium handling and neutronics considerations for a solid moving bed of lithium oxide microspheres as well as some of the mechanical and thermal hydraulic design considerations [2] have been treated in previous papers [4,5,6]. This paper is a report of further analytical studies of these mechanical and thermal hydraulic considerations; as well as analytical and experimental studies concerning the physical-chemical characteristics of lithium oxide microspheres and their fabrication methods; moving bed heat transfer characteristics; and particle transport and attrition properties.

FABRICATION OF LITHIUM OXIDE MICROSPHERES

The dominant characteristic which lithium oxide has exhibited in the research and experimentation on microsphere fabrication is its capacity for rapid and substantial absorption of moisture and carbon dioxide to form lithium hydroxide and lithium carbonate. The presence of these substances in solid solution with the lithium oxide in the microspheres to be used as fusion power reactor coolant/breeder material should be minimized since their presence decreases the sintering temperature of the microsphere bed, thus compromising its capability to maintain free flow properties in the reactor blanket environment. A prime consideration in the evaluation of methods for the fabrication of lithium oxide microspheres has therefore been the capability of these processes to retard the formation of lithium hydroxide and lithium carbonate in the final product. Some other important considerations found to be required during the experimental work concerning the fabrication of lithium oxide microspheres were the chemical purity of available starting materials, their chemical toxicity, and their cost vis-a-vis production of lithium oxide from alternate materials in the laboratory.

The fabrication of lithium oxide microspheres was seen to be accomplished in two general steps; the preparation of a sized pregranulate powder of lithium oxide or lithium peroxide, and the firing of this pregranulate powder to microspheres. During the firing step, the lithium peroxide would decompose to lithium oxide while simultaneously spheroidizing. The starting materials were fine lithium oxide and peroxide powders, both bought and generated in the laboratory from commercially available lithium hydroxide via an aqueous precipitation technique.

Numerous pregranulation techniques for the fine powder starting materials were considered and evaluated, including fluidized bed granulation, high intensity mixing, aqueous and non-aqueous gelation, pellet pressing and crushing, direct precipitation of granular peroxide, and powder bed sintering and crushing. Although all these methods excepting aqueous gelation may provide an adequate pregranulate lithium oxide material on a commercial or more advanced scale, the required simplicity of the lab-scale techniques available under this study eliminated all but the pellet pressing and bed sintering techniques from further consideration.

The quality of the microspheres depends to a large extent on the chemical quality of the available starting material. Analyses for lithium hydroxide and lithium carbonate levels in the three starter materials revealed that none provided an ultrapure lithium oxide base for the subsequent processing steps. Table 1 presents the results of these analyses for these materials; two as-received from vendors, and the third for the lithium peroxide generated by an aqueous precipitation technique.

TABLE 1.
LITHIUM HYDROXIDE AND CARBONATE IMPURITY ANALYSES IN MICROSPHERE FABRICATION STARTING MATERIALS

Source	Component (Weight Percent)				
	Li_2O_2	Li_2O	LiOH	$LiOH \cdot H_2O$	Li_2CO_3
AS-RECEIVED LITHIUM OXIDE	—	89.7	10.3	—	—
AS-RECEIVED LITHIUM PEROXIDE	94.9	0.3	—	4.8	—
PRECIPITATED LITHIUM PEROXIDE	92.2	0.4	—	7.4	—

In addition to these as-received contamination levels, subsequent pelletization, granulation, and plasma firing operations, having been carried out under controlled ventilation, but not controlled atmosphere conditions, resulted in further water and carbon dioxide contamination of the product microspheres. A small countervailing effect during plasma firing was noted in the tendency of lithium hydroxide to decompose in the plasma to lithium oxide, thereby releasing water vapor. No corresponding tendency was noted for decomposition of lithium carbonate. Thus, microsphere samples fabricated during the experimental program ranged in purity only from 22 percent to 65 percent lithium oxide.

Major conclusions concerning the fabrication of lab-scale quantities of lithium oxide microspheres which resulted from this study include:

- the basic fabrication sequence for the production of lab-scale quantities of lithium oxide microspheres seems viable
- the starting material must be a very pure lithium oxide or lithium peroxide
- as many operations preceding plasma torch firing of the pregranulate lithia as possible must be performed in a glove box or other inert, very dry carbon dioxide-free environment.

The development of a possible commercial-scale fabrication process for the manufacture of lithium oxide microspheres would appear to hinge on the following:

- The sieving, crushing, and/or ball milling of sintered or pelletized starting materials would probably be too inefficient for commercial production. Alternate large-scale pregranulation methods such as fluidized bed granulation using an acetone-soluble non-organic binder and a very dry fluidizing gas would probably be more economical.
- The plasma torch firing step seems to be very inefficient in terms of microsphere production versus total energy usage, probably due to the fact that most of the energy goes to forming and maintaining a hot plasma which vents to the outside atmosphere. Some method to more efficiently use the plasma energy for sphere forming would be desirable.

DESCRIPTION OF COOLANT LOOP AND MAJOR LOOP COMPONENTS

It was found that the practical use of Li_2O microspheres for this fusion reactor purpose requires a number of special features as listed below:

- microspheres sized to minimize system wear and attrition and provide free-flowing characteristics
- microsphere flow by gravity through reactor blanket and steam generator as a moving bed whose density approaches that of a settled bed
- a special design of an Archimedes spiral lift to circulate the bed material to provide acceptable particle attrition and system wear characteristics
- a special steam generator with double-walled croloy tubes and tube sheet buffer seals to minimize possibility of steam leaks into the moving bed
- adequate heat transfer between steam generator tubes and the moving bed through use of a suitable tube arrangement, control of voids, and maintenance of free-flowing bed properties.

Principal loop parameters are listed below in Table 2.

TABLE 2. NSS LOOP PARAMETERS

Blanket MW (th)	1200
Steam Press - psi/pascal	$1100/7.6 \times 10^6$
Feedwater Temperature - °C	232
Li_2O Temp Lv Reactor - °C	500
Li_2O Temp Ent. Reactor - °C	350
Li_2O Flow/Loop - kg/hr	2.99×10^6
Total Li_2O Lift - m	36.0
Total Lift Power - MWE	7.9
Est. Gross MW (e) ($\eta = .34$)	408
No. Coolant Loops	6

Conservative Li_2O operating temperatures and PWR steam conditions were selected to avoid use of high alloy steels in loop components and allow reasonable reductions in microsphere sintering temperature below that expected for the pure material. Location of the steam generator(s) below the reactor blanket serves to minimize that portion of the loop containing Li_2O at maximum temperatures and therefore minimizes any tendency for particle sintering. The arrangement also permits operating lift equipment at lowest possible temperatures. This minimizes problems such as cooling of bearings and drives. Arrangement simplicity results from avoidance of an intermediate coolant loop, a saturated steam cycle allowing use of an uncomplicated highly reliable steam generator and the assumption that heat storage equipment (such as required by pulsed reactor operation) would not be part of the Li_2O loop. However, hot and cool loop volumes to accommodate a typical 20-second down time between burn periods would appear to be a relatively simple addition.

Special features to permit reliable operation in a direct cycle are listed below:

- double-walled (duplex) croloy tubing to avoid thru-wall crack propagation between tube wall inspections
- shell-side helium-filled compartments adjacent to tube sheets to facilitate early detection of leaks through tube-to-tube sheet weld defects or failures and minimize amount of escaped water or steam available for contact with the flowing Li_2O microsphere bed.
- sinuous-shaped croloy tubes to impart longitudinal tube flexibility needed to avoid large loads on tube-to-tube sheet welds and ensure nucleate boiling throughout and relieve differential expansion between adjacent tubes
- short tubes with smooth inner and outer surfaces to permit rapid, effective eddy current inspection of tube walls from either end. Visual inspection of inner tube ID is also facilitated.

PARTICLE BED TRANSPORT AND HEAT TRANSFER

There are a number of considerations involved with the concept of using a gas-traversed moving bed of powder as a coolant medium, which relate to simple physical phenomena, such as attrition, errosion, conveyability and heat transfer, common to many existing industrial processes. Though none has ever proven insurmountable in commercial application, and though each has undergone considerable quantitative investigation relative to specific application, there are no entirely generalized correlations available for predicting the behavior of any randomly chosen powder in any random arrangement of process equipment. Some degree of experimental investigation on a pilot scale is therefore required in nearly all instances. By the same token, past experiences with a variety of materials in different equipment and subjected to specifically designed analytical testing certainly mitigate what might otherwise be considered technological risks by directionally indicating the relative effects of particle shape, velocity, size, loading, elasticity, and similar characteristics on attrition, erosion, conveyability, and heat transfer.

A major obstacle to the use of solids/gas mixtures for heat transfer and heat transport in closed loops has been availability of a practical pumping method to circulate the material. The problem is somewhat simplified in this application by use of gravity to produce bed flow through system heat exchange components and piping. The pumping device is then required only to lift the flowing bed from a lower to a higher elevation so that it can resume circulation through the system.

In spite of this simplification, it was found that none of the various types of particle lift equipment available today seemed capable of meeting the requirements of this application.

A search for other possible methods suggested that the Archimedes spiral lift such as used in ancient times for irrigation pumping may have application here if it could be adapted to lift particles instead of liquids.

Archimedes-type lift tubes are enclosed in pressure tight casings having seal welded flanges at each end to permit inspection and maintenance of rotating parts and bearings. Key design parameters are listed below:

Annulus	OD-m	ID-m	Sliding Speed at OD-m/sec
1	3.04	2.43	4.57
2	0.86	1.90	3.58
3	1.85	1.48	2.41

Each annulus was considered 65 percent full over a 90 degree arc and

Lift Tube Inclination = 40°
Screw Pitch Angle = 15° (each annulus)
Li_2O Flow Rate/Loop = 9.30 CFS

Li_2O feed to each stage is provided by a rotating feeder cup attached to the lower end of the lift tube assembly. Li_2O particles are supplied to this cup by means of nozzles. Ducts to each nozzle penetrate the casing. These penetrations utilize welded construction to achieve pressure tight conditions.

Potential advantages of this method of particle lift are enumerated below:

a. low particle attrition similar to that obtainable with mass lift or bucket elevators.

b. continuous flow as compared with batch-type operation of mass lift method

c. reduced Li_2O inventory through elimination of pressurizing and receiving tanks of mass lift

d. reduced system complexity through elimination of tanks, shut-off valves, gas compressors, cyclones, gas filters, elevator buckets, etc.

e. unique ability to operate at atmospheric pressure or even in a high vacuum

f. can make use of mechanical drive technology provided by screw lift elevators and high temperature tube kilns.

Saturated steam is generated from the primary loop through the transfer of sensible heat from the moving bed of lithium oxide microspheres to secondary water in a croloy longitudinal tube heat exchanger. Though mentioned qualitatively in the literature many years earlier and explored experimentally in fringe areas, it was not until about 1960 that experimental techniques and reasonable theoretical models showed quite convincingly that the transfer of heat between a moving bed of solid particles and a solid surface depends principally on the movement of the particles past the surface. Harakas and Beatty [7] measured heat transfer coefficients for a moving bed of fine particles with various interstitial gases. Heat transfer was from electrically heated surfaces at zero angle of incidence to a particle bed moving at various velocities relative to the surface. The data obtained was correlated with a simple heat transfer model which assumed a purely conductive transfer from the heated surface into a semi-infinite material moving past the surface. Heat transfer rates were found to correlate with the size of the particle, the thermal conductivity of the interstitial gas, the velocity of the bed past the heated surface, and the length of the heated surface. Brinn, et al [8] experimentally studied heat transfer from jacketed cylinders, heated by condensing steam, to granular solids in rod-like flow vertically through the cylinder. Air was used as interstitial gas, and two kinds of sands were used for the moving bed. Heat transfer coefficients were correlated with a modified Graety number and the effects of cylinder diameter, heated surface length, and temperature driving force were identical with those predicted by equations for rod-like flow of fluids through pipes. Botteril [9] investigated heat transfer coefficients generated in a stirred packed bed under differing degrees of vaccuum and treated his data in terms of convective tranfer from hot surface gas to solids as a function of particle residence time. Dunsky [10] presented the results of similar experiments which show excellent agreement with the previous data. Thornton [11] measured heat transfer from heated moving beds of glass microspheres with air and helium as interstitial gases flowing over spiral tubing containing flowing water and correlated the data as Nusselt vs bed Reynolds number. Comparisons of the data of Harakas and Beatty, Betteril, and

Thornton expressed in terms of Nusselt vs Reynolds number (defined for interstitial gas properties) indicate that heat transfer coefficients for a moving bed of microspheres flowing over steam generator tubing are significantly greater than those for fluidized beds. Properly defined moving bed flow parameters for the lithium oxide moving bed could result in heat transfer coefficients on the order of 500-1000 BTU/hr-ft^2-°F or 30-60 W/m^2-hr°K.

PARTICLE ATTRITION/EROSION

A number of theories have been advanced for relating particle strength, or physical properties, to fracture, but none have as yet yielded to a calculational model for predicting fracture rate. Since the mechanism of particle attrition can vary, and even appears to be related in some instances to the size distribution of the particles [12,15], it is not surprising to find an overall correlation lacking. Nevertheless, several obvious effects have been explored to a reasonable degree.

There is ample evidence among circulating solids processes, whether moving beds, fluid beds, or even the motion of sands at ocean shores or over desert dunes, that movement of the particles leads to a rounding of edges and that material loss in circulating non-spherical particles far exceeds that when circulating spheres of the same composition. Initially, losses from fluid bed units always decrease with time, and bed samples exhibit greater sphericity than the particles originally charged. Spheres permit the greatest uniformity of stress distribution throughout their volume with minimal concentration over lesser cross sectional areas which would increase fracture potential. The impact of a perfect sphere at any point would presumably be resisted by the shear across any diameter thereby presenting the maximum possible resisting surface shear area per particle mass.

There exist in the literature a reasonably extensive amount of data on the erosive action of particle-laden streams impinging on various surfaces. [14,15,16] Though the data appear at times to be quantitatively at variance, there is general agreement that: a) round particles erode surfaces at about half the rate attributable to angular particles; b) erosion varies approximately with the cube of the particle velocity; c) erosion rate decreases rapidly when particle size becomes smallr than 0.03" (.08 cm); d) erosion rate increases only gradually when particle size becomes larger than about 0.05" (.13cm); e) erosion rate is a maximum at particle-to-surface impingement angles of 30° to 50° from the plane of the surface; f) erosion rate increases with particle loading until the load becomes great enough to shield the eroding surface.

By analogy, these effects reflect on attrition of the impinging particles. Angular particles are more erosive but also attrit at a greater rate by losing their angular corners upon impinging a surface or another particle. Small particles are known to attrit at a lower rate than larger particles presumably due to their lower mass; as particle size becomes excessively large, the rate cannot continue to increase because of the smaller ratio of contact area to particle mass. The effect of particle velocity on attrition remains unresolved but also appears to be exponential. The shielding effect of high particle loadings on surface erosion does not reduce attrition. There is evidence that in such instances attrition is enhanced because particles then impinge upon particles.

REFERENCES

1. D. K. Sze, I. N. Sviatoslavsky, C. C. Wang, J. Wrezel, E. T. Cheng, and C. W. Maynard
A BWTR (Boiling Water Tokamak Reactor) Blanket Concept Study
Proc. Third Topical Meeting on the Technology of Controlled Nuclear Fusion
May 1978 Santa Fe, N.M.

2. D. K. Sze, D. C. Schluderberg, I. N. Sviatoslavsky
Gravity Circulated Solid Blanket Design for a Tokamak Fusion Reactor
Proc. Second Topical Meeting on the Technology of Controlled Nuclear Fusion
September 1976 Richland, Washington

3. S. I. Abdel-Khalik, R. W. Conn, W. G. Wolfer, E. N. Larsen, I. N. Sviatoslavsky
A Novel Blanket Design for ICTR's
Proc. Third Topical Meeting on the Technology of Controlled Nuclear Fusion
May 1978 Santa Fe, N.M.

4. E. M. Larsen, R. G. Clemmer, D. K. Sze
Tritium Recovery and Handling in a Gas-Carried Li_2O CTR Blanket
Trans. American Nuclear Society June 1976

5. E. M. Larsen, S. I. Abdel-Khalik, M. S. Ortman
Tritium Pathways and Handling in a Laser Fusion Reactor
Nuclear Technology V. 41 November 1978

6. E. T. Chang, T. Y. ng, D. K. Sze
Neutronics Studies of the Gas-Carried Li_2O Cooling/Breeding Fusion Reactor Blanket and Shield
Trans. American Nuclear Society June 1976

7. N. K. Harakas, K. O. Beatty
Moving Bed Heat Transfer: Effect of Interstitial Gas with Fine Particles
Chem. Eng. Progr. Symposium Series 59, No. 41 122-8, 1963

8. M. S. Brinn, S. J. Fiedman, F. A. Gluckert, R. L. Pigford
Heat Transfer to Granular Materials: Settled Beds Moving Downward through Vertical Tubes
Ind. Eng. Chem. 40, 1050 (1948)

9. J.S.M. Botteril
British Chemical Engineering 58, 174, 1951

10. V. D. Dunsky, S. S. Zabrodsky, A. I. Tamarin
On Mechanism of Heat Transfer between a Surface and a Bed of Moving Particles
Proc. Third International Heat Transfer Conference V.4 August 1966

11. T. A. Thornton, D. C. Schluderberg
Moving Bed Heat Transfer for Advanced Power Reactor Applications
Proc. Miami International Conference on Alternative Energy Sources
December 1977 Miami, Florida

12. I. Evans
Third Congress of the European Federation of Chemical Engineers
pp D66-71, June 25, 1962

13. Int. Chem. Eng. 13, No 2 pp 221-225 April 1973

14. K. Wellinger
Metallkunde 40, No. 10 361-364 (1949)

15. K. Wellinger, H. Vetz
Zeitschrift Ver. Deut. Ing. 96, No. 2 November 1954

16. K. Wellinger, H. Vetz
Zeitshrift Ver. Deut. Ing. 95, No. 26 November 1953