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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue, Argonne, Illinois 60439

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Status of the Database for Solid Breeder Materials*

by

Michael C. Billone
Fusion Power Program
Energy Technology and Technology Development Divisions

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R. F. Mattas
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D. K. Sze

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Status of the Database for Solid Breeder Materials*

M. C. Billone

Fusion Power Program
Energy Technology and Technology Development Divisions
Argonne National Laboratory
Argonne, IL 60439 (USA)

Abstract

The databases for solid breeder ceramics (Li_2O , Li_4SiO_4 , Li_2ZrO_3 and LiAlO_2) and beryllium multiplier material were critically reviewed and evaluated as part of the ITER/CDA design effort (1988-1990). The results have been documented in a detailed technical report (ANL/FPP/TM-263, 1993) which includes progress made in expanding the solid breeder and beryllium databases up through September 1993. Emphasis was placed on the physical, thermal, mechanical, chemical-stability/compatibility, tritium retention/release and radiation stability properties which are needed to assess the performance of these materials in a fusion reactor environment. Materials properties correlations were selected for use in design analysis, and ranges of input parameters (e.g., temperature, porosity, etc.) were established. The need for updating the ceramic breeder database was discussed at the Third Ceramic Breeder Blanket Interactions (CBBI-3) workshop at UCLA in June 1994. Progress made in expanding the ceramic breeder database and plans for updating the database are discussed.

1 - Introduction

The solid breeder database was critically reviewed during the ITER/CDA phase. Correlations were developed for the physical, thermal, mechanical, chemical/compatibility, tritium retention/release and radiation stability of the ITER/CDA ceramic breeder candidates (Li_2O , Li_4SiO_4 , Li_2ZrO_3 and LiAlO_2), as well as for Be multiplier material. The results of this effort are documented in Ref. 1, which includes progress made in developing the database up through September 1993. Since that time, the databases for Li_4SiO_4 and Li_2ZrO_3 have increased significantly and some progress has been made in establishing a database for the low-activation substitute for Li_2ZrO_3 , namely Li_2TiO_3 . Plans for updating the database were discussed at the Third Ceramic Breeder Blanket Interactions (CBBI-3) workshop at UCLA in June 1994. Two experts were selected by consensus for each material. M. C. Billone (ANL) volunteered to coordinate the effort and be in charge of publishing the final document.

The experts chosen at CBBI-3 to participate in updating the database are K. Noda (JAERI) and O. D. Slagle (PNNL) for Li_2O , N. Roux (CEA) and P. Gierszewski (CFFTP) for Li_2ZrO_3 , J. Miller (AECL/CRL) and C. E. Johnson (ANL) for Li_2TiO_3 , S. Casadio (ENEA/CRE) and B. Rasneur (CEA) for LiAlO_2 , H. Werle (FzK) and R. Penzhorn (FzK) for Li_4SiO_4 and H. Kawamura (JAERI/ORE) and V. Barabash (ITER/JCT-Garching) for Be.

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In the case of Li_2O , the database was reasonably complete at the time that Ref. 1 was issued. Additional information has since been provided by K. Noda and D. Yamaki on the compatibility of Li_2O /stainless-steel and $\text{Li}_2\text{O}/\text{Be}$ and by O. D. Slagle on the results of the BEATRIX irradiations. Drs. Roux and Gierszewski have provided thermal and tritium retention/release properties for Li_2ZrO_3 as well as for Li_2TiO_3 . S. Casadio has provided both a 1991 ENEA program status reports on the properties and of LiAlO_2 and Li_2ZrO_3 and a 1992-1994 ENEA program status report on the properties and performance of LiAlO_2 . Indirect contributions have also been made by J. Miller, C. E. Johnson, H. Werle, R. Penzhorn, H. Kawamura and V. Barabash through their publications.

2 - Peer Review Process

Developing the solid breeder database involves a great deal more than merely collecting data. In general, the larger the database for a particular property, the greater the scatter and the greater the need for engineering judgment. The case of the thermal conductivity of Li_2ZrO_3 is a good example. P. Gierszewski (Ref. 2) has reviewed the database for the thermal conductivity of Li_2ZrO_3 . The data set includes a range of grain sizes (~ 1 to $\sim 80 \mu\text{m}$), of densities (66 to 87% of theoretical), and a range of atmospheres (vacuum and He). Most of the data decrease with increasing temperature up to $\sim 500^\circ\text{C}$ and then either increase or level off. While thermal-conductivity models predict an increase in thermal conductivity with density, the data do not clearly show this trend. The following are some ideas by Gierszewski for rationalizing the data: eliminate large-grain-size samples which exhibited cracking; eliminate samples which exhibited phase impurity; and group the remaining data into two sets, 79-81% dense and 83-87% dense. Figure 1 shows the results of the filtered data sets. Up to about 600°C , the behavior of the two data sets is classical in that thermal conductivity decreases with increasing temperature and porosity. However, the higher-density data set levels off with increasing temperature while the lower-density data set appears to increase with temperature, such that the two data sets cross at $\sim 700^\circ\text{C}$. Figure 2 shows the results of best-fit correlations to the data in Fig. 1 along with the ITER (Ref. 1) correlations evaluated at 80 and 85% dense. Notice that the ITER correlation for 80% dense material and the new correlation for $80 \pm 1\%$ dense material are in good agreement up to $\sim 800^\circ\text{C}$. However, the ITER correlation for 85% dense material deviates significantly from the best-fit correlation for $85 \pm 2\%$ dense material, and hence the data, in the density range of 83-87% dense. Although the best-fit correlations were adjusted to cross at as high a temperature as possible, the two curves do cross at $\sim 1000^\circ\text{C}$.

To resolve the issue of the best representation of Li_2ZrO_3 thermal conductivity at densities $> 81\%$, either a better interpretation of the existing database is needed or new data at high density (e.g., 87%) are needed. Not included in the Fig. 1 data sets are the CEA thermal conductivity (really thermal diffusivity) data for Li_2ZrO_3 . Rasneur, Mougin and Roux (Ref. 3) refer to thermal conductivity data for fine-grain-size Li_2ZrO_3 sintered to densities of 70-85%. However, the data are not published. The degree to which there is still uncertainty in the thermal conductivity of $> 80\%$ dense Li_2ZrO_3 depends to some degree on how well the CEA data agree with other existing sets of data.

3 - Proposal for Updating the Solid Breeder Database

Based on a discussion during and after the presentation of this paper, the experts' list was modified. G. W. Hollenberg of PNNL replaces the retired O. D. Slagle for the second Li_2O expert. C. Alvani of ENEA/CRE replaces B. Rasneur of CEA as the second LiAlO_2 expert. There was also some discussion on the need to include Be in the database updating. Beryllium databases have been prepared for non-blanket applications such as plasma-facing component. Several members of the solid breeder community participated in this updating of the Be database. It was decided that only blanket performance parameters which are not adequately covered in other Be databases should be included in the proposed CBBI update. These include: dependence of material properties on porosity, thermal and thermal-mechanical behavior of Be pebble beds, and compatibility between Be and solid breeders in pebble or sintered-product form. M. C. Billone of ANL agreed to summarize the Be section, as well as to coordinate the updating of the solid breeder database. Another point agreed on by the group is that Li_4SiO_4 with TeO_2 should be considered as a "new material" for the purposes of generating a database.

Reference materials for each solid breeder ceramic were then discussed. The following references are recommended.

3.1 - Li_2O

3.1.1 - Compatibility

K. Noda and D. Yamaki, Summary of Li_2O /stainless-steel and Li_2O /Be compatibility, Sept. 1, 1995.

W. Dienst, D. Schild and H. Werle, "Tritium Release of Li_4SiO_4 , Li_2O and Beryllium and Chemical Compatibility of Beryllium with Li_4SiO_4 , Li_2O and Steel (SIBELIUS Irradiation)," KfK 5109, December 1992.

3.1.2 - Tritium Retention/Release and Irradiation Effects

W. Dienst, D. Schild and H. Werle, "Tritium Release of Li_4SiO_4 , Li_2O and Beryllium and Chemical Compatibility of Beryllium with Li_4SiO_4 , Li_2O and Steel (SIBELIUS Irradiation)," KfK 5109, December 1992.

9- C. E. Johnson, L. A. Neimark, and D. L. Bowers, "Characteristics of Lithium Ceramics Irradiated in the FUBR-1B/BEATRIX I Experiment, " CBBI-4 Proceedings, Kyoto, Oct. 11, 1995, pp 51-69.

G. W. Hollenberg, R. G. Clemmer, O. D. Slagle and M. E. Vaughn, "Nondestructive Examination of Lithium Ceramics Irradiated to Extended Burnup Levels, " CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 70-92.

O. D. Slagle, G. W. Hollenberg et al., "BEATRIX-II, Phase I: Data Summary Report," PNL-10279, January 1995.

O. D. Slagle, G. W. Hollenberg, et al. "BEATRIX-II, Phase II: Data Summary Report" PNNL-11148, May 1996.

H. Kwast, M. Stijkel, R. Muis and R. Conrad, "EXOTIC: Development of Ceramic Tritium Breeding Materials for Fusion Reactor Blankets: The Behaviour of Tritium in: Lithium Aluminate, Lithium Oxide, Lithium Silicates, Lithium Zirconates," ECN-C-95-123, December 1995.

3.2 - Li_2ZrO_3

3.2.1 - Properties and Performance Compilations

S. Casadio et al., "Status Report 1991 on E.N.E.A. Contribution to the Development of Lithium Aluminate and Zirconate Ceramic Breeder Materials for BIT DEMO Relevant Blankets," ENEA/RTI/INN 92 (6), 1992.

3.2.2 - Thermal and Elastic Properties

P. Gierszewski, "Heat Capacity of Lithium Metazirconate," Fus. Tech. 23 (1993) 331-332.

P. Gierszewski, "Thermal Conductivity of Lithium Metazirconate," Fus. Tech. 23 (1993) 333-336.

B. Rasneur, J. Mougin and N. Roux, "Study of the Thermomechanical and Tritium Release Behaviour of Li_2ZrO_3 as Tritium Breeding Ceramic for the European BIT Blanket Concept," J. Nucl. Mater. 212-215 (1994) 923-926.

N. Roux, J. Avon, A. Floreancig, B. Rasneur, and S. Ravel, "Low-Temperature Tritium Releasing Ceramics as Potential Materials for the ITER Breeding Blanket," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 452-468. (Also ICFRM-7 paper)

3.2.3 - Interaction with Moisture and Other Gases

C. Alvani, M. R. Mancini, F. Alessandrini and S. Casadio, "Environmental Effects on Li_2ZrO_3 and Properties Evolution as a Function of the Fabrication Route," CBBI-2 Proceedings, Paris, Jan. 1994, pp 369-378.

C. Alvani, S. Casadio and M. R. Mancini, "Interaction of Li_2ZrO_3 with Various Gases (H_2 , H_2O , CO_2 and O_2) as Studied by Temperature Programmed Desorption TPD," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 274-284.

J. P. Kopasz and C. E. Johnson, "Investigations of the Interactions of $\text{H}_2\text{O}(\text{g})$ with Li_2ZrO_3 ," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 224-234.

Y. Kawamura and M. Nishikawa, "Adsorption Characteristics of Water Vapor on Li_2ZrO_3 ," J. Nucl. Mater., 218 (1994) 57-65.

C. Alvani, M. R. Mancini and S. Casadio, "Hydrogen Oxidation and Water Adsorption-Desorption on Li_2ZrO_3 Surface," these proceedings.

3.2.4 - Tritium Retention/Release and Irradiation Performance

J. M. Miller and R. A. Verrall, "Performance of a Li_2ZrO_3 Sphere-Pac Assembly in the CRITIC-II Irradiation Experiment," J. Nucl. Mater. 212-215 (1994) 897-901.

B. Rasneur, J. Mougin and N. Roux, "Study of the Thermomechanical and Tritium Release behaviour of Li_2ZrO_3 as Tritium Breeding Ceramic for the European BIT Blanket Concept," J. Nucl. Mater. 212-215 (1994) 923-926.

9- C. E. Johnson, L. A. Neimark, and D. L. Bowers, "Characteristics of Lithium Ceramics Irradiated in the FUBR-1B/BEATRIX I Experiment," CBBI-4 Proceedings, Kyoto, Oct. 11, 1995, pp 51-69.

G. W. Hollenberg, R. G. Clemmer, O. D. Slagle and M. E. Vaughn, "Nondestructive Examination of Lithium Ceramics Irradiated to Extended Burnup Levels," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 70-92.

J. G. van der Laan, H. Kwast, R. Conrad and R. May, "Irradiation Behavior of Ceramic Breeder Materials at High Lithium Burnup," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 128-141.

H. Kwast, M. Stijkel, R. Muis and R. Conrad, "EXOTIC: Development of Ceramic Tritium Breeding Materials for Fusion Reactor Blankets: The Behaviour of Tritium in: Lithium Aluminate, Lithium Oxide, Lithium Silicates, Lithium Zirconates," ECN-C-95-123, December 1995.

J. G. van der Laan, Marcel Stijkel and R. Conrad, "Post Irradiation Examination of EXOTIC-7: Irradiation to High Lithium Burnups," these proceedings.

O. D. Slagle, G. W. Hollenberg, et al., "BEATRIX-II, Phase II: Data Summary Report" PNNL-11148, May 1996.

3.3 - Li_2TiO_3

3.3.1 - Properties

N. Roux, J. Avon, A. Floreancig, B. Rasneur, and S. Ravel, "Low-Temperature Tritium Releasing Ceramics as Potential Materials for the ITER Breeding Blanket," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 452-468. (Also ICFRM-7 paper)

P. Gierszewski, "Review of Lithium Metatitanate," CFFTP G-9561, Oct. 1995.

J. W. Davis and A. A. Haasz, "Thermal Diffusivity/Conductivity of AECL Li_2TiO_3 ," CFFTP G-9567, Nov. 1995.

3.3.2 - Tritium Retention/Release and Irradiation Effects

J. P. Kopasz, J. M. Miller and C. E. Johnson, "Tritium Release from Lithium Titanate, A Low Activation Tritium Breeding Material," CBBI-2 Proceedings, Paris, January 1994, pp 81-95.

N. Roux, J. Avon, A. Floreancig, B. Rasneur, and S. Ravel, "Low-Temperature Tritium Releasing Ceramics as Potential Materials for the ITER Breeding Blanket," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 452-468. (Also ICFRM-7 paper)

3.4 - LiAlO_2

3.4.1 - Properties and Performance Compilations

S. Casadio et al., "Status Report 1991 on E.N.E.A. Contribution to the Development of Lithium Aluminate and Zirconate Ceramic Breeder Materials for BIT DEMO Relevant Blankets," ENEA/RTI/INN 92 (6), 1992.

F. Alessandrini, C. Alvani, S. Casadio, M. R. Mancini, A. Moauro, C. A. Nannetti and F. Pierdominici, "Status Report on ENEA Contribution to Development of Lithium Aluminate Ceramic Breeder Development for BIT DEMO Relevant Blankets (1992-1994 Program)," Doc. INN/NUMA/MATAV(95)2.

3.4.2 - Tritium Retention/Release and Irradiation Effects

9- C. E. Johnson, L. A. Neimark, and D. L. Bowers, "Characteristics of Lithium Ceramics Irradiated in the FUBR-1B/BEATRIX I Experiment," CBBI-4 Proceedings, Kyoto, Oct. 11, 1995, pp 51-69.

G. W. Hollenberg, R. G. Clemmer, O. D. Slagle and M. E. Vaughn, "Nondestructive Examination of Lithium Ceramics Irradiated to Extended Burnup Levels," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 70-92.

J. G. van der Laan, H. Kwast, R. Conrad and R. May, "Irradiation Behavior of Ceramic Breeder Materials at High Lithium Burnup," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 128-141.

H. Kwast, M. Stijkel, R. Muis and R. Conrad, "EXOTIC: Development of Ceramic Tritium Breeding Materials for Fusion Reactor Blankets: The Behaviour of Tritium in: Lithium Aluminate, Lithium Oxide, Lithium Silicates, Lithium Zirconates," ECN-C-95-123, December 1995.

J. G. van der Laan, Marcel Stijkel and R. Conrad, "Post Irradiation Examination of EXOTIC-7: Irradiation to High Lithium Burnups," these proceedings.

3.5 - Li_4SiO_4

3.5.1 - Compatibility

W. Dienst, D. Schild and H. Werle, "Tritium Release of Li_4SiO_4 , Li_2O and Beryllium and Chemical Compatibility of Beryllium with Li_4SiO_4 , Li_2O and Steel (SIBELIUS Irradiation)," KfK 5109, December 1992.

3.5.2 - Interactions with Moisture

Y. Kawamura, M. Nishikawa and K. Tanaka, "Adsorption Characteristics of Water Vapor on Li_4SiO_4 ," J. Nucl. Mater., 208 (1994) 308-312.

3.5.3 - Tritium Retention/Release and Irradiation Effects

W. Dienst, D. Schild and H. Werle, "Tritium Release of Li_4SiO_4 , Li_2O and Beryllium and Chemical Compatibility of Beryllium with Li_4SiO_4 , Li_2O and Steel (SIBELIUS Irradiation)," KfK 5109, December 1992.

9- C. E. Johnson, L. A. Neimark, and D. L. Bowers, "Characteristics of Lithium Ceramics Irradiated in the FUBR-1B/BEATRIX I Experiment," CBBI-4 Proceedings, Kyoto, Oct. 11, 1995, pp 51-69.

G. W. Hollenberg, R. G. Clemmer, O. D. Slagle and M. E. Vaughn, "Nondestructive Examination of Lithium Ceramics Irradiated to Extended Burnup Levels," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 70-92.

J. G. van der Laan, H. Kwast, R. Conrad and R. May, "Irradiation Behavior of Ceramic Breeder Materials at High Lithium Burnup," CBBI-4 Proceedings, Kyoto, Oct. 9-11, 1995, pp 128-141.

H. Kwast, M. Stijkel, R. Muis and R. Conrad, "EXOTIC: Development of Ceramic Tritium Breeding Materials for Fusion Reactor Blankets: The Behaviour of Tritium in: Lithium Aluminate, Lithium Oxide, Lithium Silicates, Lithium Zirconates," ECN-C-95-123, December 1995.

J. G. van der Laan, Marcel Stijkel and R. Conrad, "Post Irradiation Examination of EXOTIC-7: Irradiation to High Lithium Burnups," these proceedings.

3.6 - Li_4SiO_4 with TeO_2

M. Dalle Donne, G. Piazza, A. Weisenburger, H. Werle, V. Geller, B. Speit, D. Sprenger, "Development Work for Lithium Orthosilicate Pebbles," to be submitted for publication

4. - Discussion

The proposed process of updating the solid breeder database consists of the two experts critically reviewing the data and correlations in ANL/FPP/TM-263 for their designated material, comparing these data and correlations to the information in the more recent publications listed above, coming to agreement regarding changes which should be made to the database/correlations and submitting their recommendations to M. C. Billone. Dr. Billone is to act as an impartial judge with regard to the recommended improvements. If the recommendations are well-documented and justifiable, then Dr. Billone submits these recommendations to the group as a whole for approval. If not, then Dr. Billone interacts with the two experts until agreement is reached. Thus, the final product is a documented database which has met with the approval of the entire CBBI database working group.

References

1. M. C. Billone, W. Dienst, T. Flament, P. Lorenzetto, K. Noda and N. Roux, "ITER Solid Breeder Blanket Materials Database," Argonne National Laboratory Report, ANL/FPP/TM-263, Nov. 1993.
2. P. Gierszewski, "Thermal Conductivity of Lithium Metazirconate," Fus. Tech. 23 (1993) 333-336.
3. B. Rasneur, J. Mougin and N. Roux, "Study of the Thermomechanical and Tritium Release Behaviour of Li_2ZrO_3 as Tritium Breeding Ceramic for the European BIT Blanket Concept," J. Nucl. Mater. 212-215 (1994) 923-926.

Fig. 1. Thermal conductivity data for unirradiated Li_2ZrO_3 as reported in Ref. 2. The numbers in parentheses indicate ranges of specimen density (in % theoretical). The data set has been filtered in that results for large grain size samples ($\geq 10 \mu\text{m}$) and for samples with phase impurity have not been included.

Fig. 2 Comparison between correlations for the thermal conductivity of Li_2ZrO_3 . The correlations labeled ITER (80%) and ITER (85%) are from Ref. 1, where the numbers in parentheses represent density in % theoretical. The correlations labeled New Correlation ($80 \pm 1\%$) and New Correlation ($85 \pm 2\%$) represent best-fit correlations to the data presented in Fig. 1.