

1

O

f

1

RADIATION EFFECTS IN BERYLLIUM
USED FOR PLASMA PROTECTION

D. S. Gelles
M. Dalle Donne^(a)
G. A. Sernyaev^(b)
H. Kawamura^(c)

September - October 1993

Presented at the
6th International Conference
on Fusion Reactor Material
September 27 - October 1, 1993
Stressa, Italy

Work supported by
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

- (a) Karlsruhe 1, Germany
(b) SF NIKIET, Russia Federation
(c) JAERI, Tokai-mura, Japan

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

VED
DEC 30 1993

OSTI

RADIATION EFFECTS IN BERYLLIUM USED FOR PLASMA PROTECTION

D. S. Gelles
Pacific Northwest Laboratory*
Richland, WA 99352, USA

G. A. Sernyaev
SF NIKIET
Zarechnyi
Russia Federation

M. Dalle Donne
Kernforschungszentrum
Karlsruhe
Postfach 36 40
D-7500 Karlsruhe 1, Germany

H. Kawamura
Blanket Irradiation and Analysis
Laboratory, JMTR,
Oarai Research Establishment, JAERI
Oarai-machi, Nigashi Ibaraki-gun
Japan, 311-13

Abstract

This paper reviews the literature on beryllium, emphasizing the effects of irradiation on essential properties. Swelling and embrittlement experiments as a function of irradiation temperature and dose, and as a function of neutron spectrum are described, and the results are quantified, where possible. Effects of impurity content are also reported, from which optimum composition specifications can be defined. Microstructural information has also been obtained to elucidate the processes controlling the property changes.

The available information indicates that beryllium divertors can be expected to embrittle quickly and may need frequent replacement.

Introduction

Beryllium is presently a leading candidate material for fusion reactor first wall coating and divertor applications. This is largely a result of improved performance in the Joint European Torus (JET) after evaporated beryllium, and then beryllium tiles as a plasma facing material were installed in the limiter area. [1] In comparison with graphite that had been used previously, beryllium reduced the plasma radiation, increased the density limit, reduced the incidence of disruptions, and enhanced the deuteron pumping. Based in part on its good thermal and physical properties, but also a result of operating experience in JET, beryllium has recently been identified as the material of choice for the International Thermonuclear Experimental Reactor (ITER) both as a plasma facing material and as a divertor material. In that application, it would provide the interface material between plasma and confinement structure in the first fusion reactor to generate significant quantities of 14 MeV neutrons.

Beryllium is also the candidate material of choice as the neutron multiplier in a solid breeder blanket design. In that capacity, it would provide additional neutrons for tritium production in a blanket containing lithium ceramics. However, design optimization studies have shown that the blanket must contain at least 70% beryllium to maximize tritium production. If beryllium is used as a plasma facing material, a divertor material, and blanket neutron multiplier in a fusion reactor design, it is apparent that

*Operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830. 2

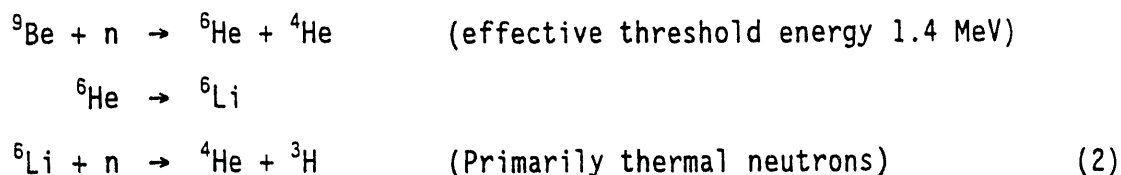
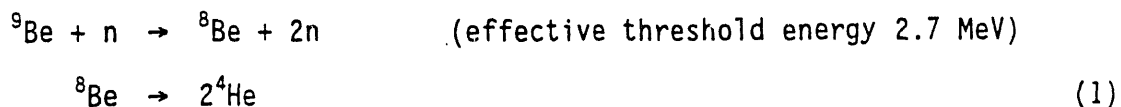
the fusion reactor will incorporate beryllium as a major fraction of the materials making up the reactor design. Operating such a fusion reactor may therefore be very dependent on the response of beryllium to 14 MeV neutron damage.

However, beryllium is degraded by radiation damage, both as a result of displacement damage and of transmutation. Displacement damage leads to point defect clustering, irradiation hardening, and embrittlement. Transmutation produces helium (and lithium), resulting in high levels of gas-driven swelling and embrittlement at high temperatures. It is not yet clear what limitations this damage will place on fusion reactor applications. Results of experiments seem to be contradictory.

The purpose of this review is to present data concerning effects of irradiation on essential properties. Swelling and embrittlement experiments as a function of irradiation temperature and dose and as a function of the neutron spectrum are described, and the results are quantified where possible. Effects of impurity content are also reported, and the optimum composition specifications can be defined. Microstructural information has also been obtained to explain the processes controlling the property changes. Previously published reviews on radiation damage effects in beryllium may be noted. [2-5]

Neutronic considerations

A major contributor to radiation damage in beryllium, in addition to displacement damage, is the production of helium and tritium from (n,2n) and (n, α) reactions. Each beryllium atom can transmute to two helium atoms depending on neutron energy according to the following reactions:



As a consequence, under high energy neutron bombardment, helium is generated in much greater quantities in beryllium than in any other metal. It has been estimated that for a 14 MeV neutron wall loading of 1 MW/m², 4.73 dpa/yr will be produced at a helium generation rate of 617 appm-He/dpa or 2920 appm-He/yr. [3]

Point Defect Accumulation and Swelling

Radiation damage in beryllium can best be characterized by distinguishing between point defect accumulation, point defect coalescence, and gas driven swelling.

Gol'tsev and coworkers [6] have determined that at low temperatures where mobility of point defects is high enough to efficiently recombine and accumulate at point defect sinks, but gas atoms are practically immobile, swelling in beryllium can be described by the expression:

$$\Delta V/V_0 = 8.2 \times 10^{-25} \phi t \quad (3)$$

where ϕt is fast fluence in n/cm^2 , $E > 0.85$ MeV. This is based on irradiations at $60^\circ C$, but can be used over the range of temperatures where helium remains in supersaturated solid solution.

From this expression, it can be noted that at a fluence of 10^{22} n/cm^2 , a swelling level of 0.8% is predicted. Sernyaev [7] has published swelling data for various grades of pure beryllium irradiated at between 450 and $500^\circ C$ to doses between 5.7×10^{21} and 1.02×10^{22} n/cm^2 , $E > 0.85$ MeV. Swelling levels of between 0.4 and 0.8% were recorded, the higher values corresponding to higher fluence levels. Sannen and De Raedt [8] have published swelling data for vacuum hot pressed beryllium irradiated at 40 to $50^\circ C$ to fluences of 0.8, 2.8, and 3.9×10^{22} n/cm^2 , $E > 1$ MeV. Swelling values of 0.65, 1.57, and 2.27% were obtained, and a somewhat lower correlation factor of $0.58 \pm 0.3 \times 10^{-25}$ was recommended for expression (3). Koonen [9] has published swelling data for vacuum hot-pressed beryllium irradiated at 40 to $50^\circ C$ to fluences as high as 8×10^{22} n/cm^2 , $E > 1$ MeV with a maximum of 2.2% diametral swelling (corresponding to 6.6% volumetric swelling if swelling were isotropic.) Finally, Burmistrov and coworkers [10] published measurements on the beryllium core moderator block of an MIR reactor irradiated to a fluence of 3.2×10^{22} n/cm^2 at $40^\circ C$, noting that the swelling did not exceed 0.3%, but that this was in agreement with expression (3). Therefore, the correlation factor of 8.2×10^{-25} may not be valid for all materials.

For example, based on the review by Dalle Donne and co-workers, [11] it can be shown that product form can control swelling response at low irradiation temperatures. Figure 1 provides a plot of volumetric swelling at temperatures below $100^\circ C$ as a function of helium content, which is proportional to fluence. Significant differences are found between the swelling measurements of Sernyaev [12] and those obtained in western experiments. [13-15] The Sernyaev data points all fall above the trend line defining western measurements. The difference is attributed to modern beryllium production and processing. The Sernyaev results are based on beryllium irradiated in the 1960s, whereas the western data in Figure 1 are more recent and show response on modern beryllium production technology. The earlier Russian beryllium is very anisotropic, with large grains and relatively high amounts of impurities (BeO and others) whereas modern beryllium made by powder processing techniques is fine grained and more isotropic. This therefore emphasizes an effect of microstructure on low temperature swelling response.

An effect of higher fluence is demonstrable from the Koonen measurements [9] where beryllium was irradiated at 40 to $50^\circ C$ in the BR2 Research Reactor. Non-linear response can be identified for fluences above 6.4×10^{22} n/cm^2 , $E > 1$ MeV so that swelling increases either in a bilinear or quadratic fashion from that dose. Based on these measurements, safety issues have been satisfied to allow irradiation of beryllium as BR2 in-core structural components to this fluence of 6.4×10^{22} n/cm^2 , $E > 1$ MeV. This

decision attests to the use of modern hot-pressed beryllium for use in high fluence irradiation applications.

Beeston and Miller [16] provide a somewhat different expression for swelling as a function of fluence applicable to the temperature range 400 to 600°C:

$$\Delta V/V_0 = 1.83 \times 10^{-58} (\phi t)^2 T^4 \quad (4)$$

where ϕt is fast fluence in n/cm^2 , $E > 1$ MeV, and T is the temperature in K. This expression was derived from density change measurements made on a beryllium cylinder that was irradiated in a flux gradient in a fast breeder reactor for 4 years and cut into pieces, providing 26 samples with fluences between 0.7 and 1.3×10^{22} n/cm^2 , $E > 1$ MeV, at irradiation temperatures between 427 and $482^\circ C$. From this expression, it can be noted that at $500^\circ C$ and a fluence of 10^{22} n/cm^2 , a swelling level of 0.65% is predicted, in reasonable agreement with the data from Sernyaev [7]. However, for an irradiation temperature of $50^\circ C$ and a fluence of 10^{22} n/cm^2 , expression (4) predicts 0.02% , and therefore, this equation cannot predict the results of Sannen and De Raedt [8] or Koonen. [9]

At temperatures where helium and tritium mobility becomes large enough to allow consolidation into large bubbles, the swelling accumulation during neutron irradiation is larger by over an order of magnitude, and the swelling dependence on fluence and temperature is different. The dependence can be described by the expression from Sernyaev [7]

$$\Delta V/V_0 \sim M \cdot T \cdot \exp[-Q/(4kT)] (\phi t)^{3/2} \quad (5)$$

where M is a structure-sensitive factor and T , Q , k , and ϕt have their usual meaning. Sernyaev has analyzed available data and found that Q is 2.1 ± 0.1 eV/at and M varies from 0.31 to 1.65×10^{-34} $K^{-1} (n/cm^2)^{-3/2}$. The dependence of M on materials parameters was found to correlate best with a processing procedure such that hot pressing produced significantly higher values for M than did extrusion. However, given a specific processing procedure, M was found to increase with decreasing oxygen content and with increasing grain size. Therefore, to minimize high temperature swelling, extruded materials with oxygen levels on the order of 3% and grain diameters between 10 and $20 \mu m$ would be recommended.

Examples of swelling in beryllium irradiated at high temperatures are given in Figure 2. Figure 2 (a) shows the data of Sernyaev [7] from which the structure-sensitive factor analysis described above is based and Figure 2 (b) gives results from Burmistrov. [10] Swelling as high as 18% can be noted in hot-pressed $56 \mu m$ grain size beryllium irradiated to 8.9×10^{21} n/cm^2 , $E > 0.85$ MeV. In both plots, it is apparent that swelling in beryllium is sensitive to manufacturing and processing variables.

To understand the consequences on swelling of helium present in beryllium, a number of experiments have been performed where specimens irradiated at low temperatures were subsequently heated to higher temperatures, and the resultant increases in swelling were measured. [16-22] Often, these experiments used 1 hour annealing increments, but many followed the same condition at temperature for many hours. The swelling produced depended somewhat on the neutron fluence, because higher doses produced higher

helium levels. Doses were poorly quantified, but ranged from 5×10^{20} to 10^{22} n/cm². The general response of annealing experiments can be summarized as follows. Effects of annealing on swelling were only observed for temperatures of 600°C and above, but at 600 and 700°C, all annealing experiments produced swelling levels of 1% or less. [17,21] One hour anneals at 800°C produced swelling levels as high as 14% [17], but generally the values were negligible [18-20,22] or between 3 and 5.5%. [21] One hour anneals at 900°C raise the swelling somewhat, giving values ranging from 0.5 to 17%, and 1000°C anneals raised the maximum values to about 32%, [17] but still higher annealing temperatures of 1100 and 1200°C produced swelling only on the order of 40%. [17,20] The data have been analyzed to show Arrhenius behavior (with an activation energy of 40 kcal/mole, about the value for self-diffusion in beryllium) for 1 hour anneals over the temperature range 600 to 900°C, but a saturation in behavior at higher temperatures occurred, corresponding to about 30% (after heating to about 1000°C for 1 hour.) [17]

When longer annealing times were used, [19,20,22] swelling sometimes increased to much higher levels, approaching saturation behavior as noted above. For example, at 800°C where 1 hour anneals produced negligible swelling, anneals for 20 hours [20] and 1000 hours [22] produced swelling on the order of 10%, and at 900°C, anneals for 700 hours produced 10% swelling [22] and for 10 hours produced 25% swelling. [20] The swelling as a function of annealing time has been shown to give linear behavior on log-log plots, and an activation energy of 22 kcal/mole was calculated for the controlling process. [22] However, two examples of rapid swelling after apparent saturation are reported [20], probably similar in process to the tritium burst-release identified in more recent experiments where most of the tritium was released in a sudden burst. [8,23]

More recently, Sernyaev [23] has studied the initiation and growth of helium gas bubbles in single crystal beryllium irradiated at 60°C to fluences of 2.6 and 5.1×10^{21} n/cm², $E > 0.8$ MeV, and then annealed at temperatures of 100 to 1100°C with 1 hour hold times. Techniques employed included small angle x-ray scattering (SAS), differential microcalorimetry (DMC), and transmission electron microscopy (TEM), and it was possible to demonstrate that bubble nucleation occurred at temperatures from 350 to 600°C and that bubbles grew above 600°C. The SAS measurement also allowed estimates of mean bubble diameter as a function of annealing time, with the interesting result that the specimen irradiated to lower fluence developed larger bubbles at temperatures above 775°C with bubbles approaching 12.5 nm in diameter.

Mechanical properties - hardening and embrittlement

Beryllium metal has the hexagonal close packed (HCP) crystal structure with a c/a ratio of 1.5677 at temperatures below 1254°C. As with other HCP and body centered cubic crystal structures, beryllium displays a thermally activated deformation response at low temperatures, leading to increasing yield strength with decreasing temperature, and a tendency for twinning and brittle fracture at low temperatures. Slip has been observed on basal, prism, and pyramidal planes in beryllium, but due to the thermal activation process that controls at low temperatures, slip can become difficult, and embrittlement results if the temperature is low enough. However, beryllium differs from other metals in that the elastic moduli are very high, the

interatomic distance is very short, the Poisson's ratio is very low, and the Debye temperature is very high. [24] All are indications of strong interatomic forces, and therefore beryllium is more sensitive than most metals to embrittlement at low temperatures.

However, manufacturers have successfully produced beryllium with reasonable room temperature mechanical properties. In general, two approaches are responsible: higher purity and refined grain size. This has generally necessitated the use of powder metallurgy fabrication techniques. However, there are few producers of beryllium in the world.

Irradiation adversely affects the mechanical properties of beryllium due to two processes. Point defects produced by radiation damage can cluster to form obstacles to dislocation motion, and helium can migrate to point defect sinks to form bubbles which are also obstacles to dislocation and grain boundary motion. These bubbles can provide nucleation sites for cracking. Restrictions to dislocation motion are manifested as hardening, strength increases, ductility decreases, fracture toughness decreases, and embrittlement. A large number of studies on beryllium have included experiments to study these phenomena. [17-20,22,25-42] These include results on strength and ductility changes due to irradiation, [18-20,22,25-38] bend test response, [17,38] hardness, [17,18,20,25,27,36,39] fracture toughness, [40-42] and stress rupture. [36,39] However, most of those studies were performed about 30 years ago when an international effort was made to develop beryllium as a fuel cladding material for fission reactors. Unfortunately, the dosimetry is crude, and it is very difficult to quantitatively compare results of the different studies. The database on irradiation effects to mechanical properties has been reviewed previously. [36,43]

Studies on strength and ductility changes due to irradiation have shown that, as with swelling studies, irradiation temperature affects response. At low irradiation temperatures, strength increase due to irradiation is most evident, whereas at high irradiation temperatures on the order of 650°C, irradiation embrittlement can occur without significant strength increase. To minimize the possibility of zero ductility response, a number of yield strength measurement techniques have been applied including uniaxial tensile strength, compressive strength, shear strength, splitting tensile strength, flexural strength and three point bend tests.

However, the general trend of the response following low-temperature irradiation is for strength to begin to increase at fluences on the order of 10^{19} n/cm² ($E > 1$ MeV), to saturate as ductility approaches zero, and then to decrease with further fluence. An example is shown in Figure 3 (a) providing comparison of available data for increase in tensile yield strength as a function of fluence. The original plot, provided by Hickman, [43] predicted linear behavior based on the "□" symbols, but more recent data do not support a linear description. Figure 3 (b), originally provided by Beeston and coworkers, [32] shows compressive fracture or yield strength as a function of fluence for specimens irradiated at 120°C and emphasizes the tendency for reduced strength beyond fluences where saturation occurs. Note that saturation in compressive tests can be expected to occur at a higher fluence because compression allows testing to higher levels of strength before failure.

Hardness measurements further demonstrate linear response in strength with fluence at low temperatures. Figure 4 (a) shows diamond pyramid hardness for beryllium irradiated in the temperature range 35 to 100°C, originally compiled by Hickman, [43] but now including further data, [29] plotted logarithmically as a function of fluence. Hardness values begin to increase at fluences on the order of 2×10^{19} n/cm², similar to the yield strength response. The plot is linear with only a hint of saturation.

However, response following irradiation at higher temperatures gives reduced hardening. Figure 4 (b) has been prepared to show this effect. The trend line from Figure 4 (a) is reproduced, and available data points showing hardness increase are plotted and labelled with the irradiation temperature. The tendency is for 280 to 400°C data points to lie close to the trend line, but as irradiation temperature is increased, hardness increase is reduced. Irradiation at 700°C [39] produces softening, and those data points have not been included.

Strength changes following irradiation at 280°C and above follow these trends, but the dependence of strength increase as a function of fluence may be different. Walters [34] has noted that yield strength as a function of dose is linear when plotted as $(\phi t)^{1/2}$ for specimens irradiated at 350°C. Similar slopes are produced by the data from other experiments following irradiation from 280 to 500°C. This response is shown in Figure 5.

At still higher temperatures, such as 650°C and above, the strength behavior is more complicated. About half the experiments indicate a strength increase, [25,26,30] and the rest show a decrease. [18,27,33,39] An explanation is provided in Figure 6, which shows an example of compressive yield strength at room temperature in beryllium specimens irradiated at 650°C. [38] The strength is found to increase to a maximum at fluences on the order of 6×10^{20} n/cm², $E > 0.85$ MeV, and then to decrease to levels significantly lower than the unirradiated strength. This response would explain why, in Figure 4 (b), the hardness values for several lower fluence, high temperature irradiation conditions appear near or on the trend line.

Concurrent with irradiation induced strengthening, reduction in ductility and embrittlement occurs. This was generally because ultimate tensile strength reduced with fluence, and therefore, the allowable plastic deformation was reduced. A number of examples are found in the literature where irradiation resulted in negligible ductility or complete embrittlement during bend testing [16] and tensile testing. [20,21,29,37,38] This included irradiations at temperatures of 100°C or below to fluences as low as 4×10^{21} n/cm², [17,20,29], irradiations at 280 to 400°C to fluences of 10^{21} n/cm², [20,37,38] and irradiation at 650°C to 10^{21} n/cm². [38] However, it is likely that in all cases, the sources of beryllium used older production techniques. No results on hot pressed powder products are reported showing completely brittle response.

Three reports have addressed the issue of fracture toughness degradation due to irradiation. [40-42] However, the first two consider irradiation at liquid nitrogen temperature and are therefore not fusion relevant. Moderate reductions in fracture toughness were found in those cases following low fluence irradiation (8×10^{18} [40] and 7×10^{17} n/cm² [41]). They mainly serve to demonstrate the thermal activation nature of beryllium; testing at lower temperature gives lower fracture toughness values. A report by Beeston is

more relevant. [42] The material studied was nuclear-grade hot-pressed beryllium, including a porous product intended to expand understanding of porous material response, and irradiation was to fluences of 3.5 to 5.0×10^{21} n/cm^2 , $E > 1$ MeV at $66^\circ C$. The fracture toughness of solid beryllium was found to be reduced from 12.0 MPa $m^{1/2}$ to 5 MPa due to irradiation, and the fracture toughness of porous beryllium with a slightly higher unirradiated value of 13.1 MPa $m^{1/2}$ was reduced to 4.2 MPa.

Microstructural investigations

A number of studies have included efforts to examine microstructural features to better understand irradiation phenomena in beryllium. These have included fractographic examinations to reveal bubbles on grain boundaries, transmission electron microscopy to reveal bubble distributions, and transmission electron microscopy to reveal dislocation loops and black spot damage. Early workers relied on either replica techniques, scanning microscopy, or even optical microscopy to show fracture initiation sites. [17,20,25,26,28,30,33,39,44] Helium bubble investigations considered smaller bubble sizes by using transmission electron microscopy, [10,16,20,23,37,45-48] and often employed annealing experiments to better understand bubble development. [17,19,20,22,23,32,34,44-48] Several studies have noted dislocation loop development as well. [19,23,32,34,37,49-51]

These microstructural investigations confirmed the different regimes of response as a function of temperature noted previously. At irradiation temperatures of $400^\circ C$ and below, damage consisted of black spot or loop damage. [19,23,32,34,37,45,47,49,50] Burgers vector analysis of the loop structure in irradiated beryllium, when attempted, gave different results. Following neutron irradiation at $350^\circ C$ to 2×10^{20} fast n/cm^2 , dislocation loops 20 to 70 nm in diameter were generally found on $\{11\bar{2}0\}$ planes, but additional small numbers were also observed on $\{10\bar{1}0\}$, $\{1101\}$, and $\{11\bar{2}2\}$ planes, and the Burgers vector responsible in these latter cases was expected to be $\frac{1}{3}\langle 11\bar{2}3 \rangle$. [49] Following neutron irradiation to 4×10^{21} n/cm^2 , $E > 1$ MeV, at $400^\circ C$, a low density of $\frac{2}{3}\langle 11\bar{2}0 \rangle$ dislocation tangles and a high density of $c\langle 0002 \rangle$ loops, approximately 20 nm in diameter, were found. [37] Finally, following 1 MeV electron irradiation at $100^\circ C$ to a dose of 4 dpa, loops appeared to be close to end-on orientation near $\{11\bar{2}0\}$ planes, and therefore, a Burgers vector of $\frac{2}{3}\langle 11\bar{2}0 \rangle$ was indicated. This probably means that the common Burgers vector for loops in irradiated beryllium is $\frac{2}{3}\langle 11\bar{2}0 \rangle$, but loops with a \bar{c} component do form at a slower rate.

At higher temperatures, the microstructural product of irradiation is mainly bubbles. Helium bubbles are observed at grain boundaries following irradiation in the temperature range 325 to $400^\circ C$, [16,37] and are reported on dislocations within grains following irradiation at 450 to 550 . [20,37,46,47] However, following irradiation at temperatures of $600^\circ C$ and above, all investigators report bubbles, generally on grain boundaries. [10,19,25,26,28,30,33,39,44,46,47] Many of the fractographic studies revealed that large bubbles located on grain boundaries were faceted. This was also noted for smaller cavities, [37,46,47] but in general, bubbles were considered to be spherical.

Many studies included microstructural examinations following high temperature annealing treatments. [17,19,20,22,23,32,34,44-48] The results reported can be summarized with three observations: bubbles formed on dislocations as a result of anneals in the temperature range 300 to 900°C, [20,39,45,46,48] dislocation loops were annihilated following annealing in the temperature range 500 to 600°C, [19,34,47,48] and only bubbles were present following anneals at 600°C and above. [17,20,22,32,34,48] Also of note is an observation of Shiozawa and co-workers [46,47] that bubbles attached to dislocation networks did not change size and distribution below 900°C. Bubble growth under annealing conditions was presumed to occur only under dislocation and grain boundary sweeping conditions. However, this observation may only apply to annealing experiments; under neutron irradiation, helium atom migration was feasible.

Composition optimization

Three examples of composition optimization have been provided above. Beryllium fabrication is optimized by purification and refined grain size. Analysis of swelling response at high temperatures by Sernyaev [7] showed that swelling is increased (when the M parameter is increased) with decreasing oxygen content and with increasing grain size. Extruded materials with oxygen levels on the order of 3% and grain diameters between 10 and 20 μm would be recommended to minimize swelling at high temperatures. Finally, comparison of swelling response at low temperatures [11] showed that modern fine-grained isotropic materials provide greater swelling resistance than older, coarse grained, anisotropic material with high levels of impurity. All three optimizations indicated that modern fine-grained beryllium will be more serviceable for plasma protection than the materials on which early experiments were based.

Health issues

Use of beryllium is associated with a hazard to health. It has been reputed to be one of the most toxic elements known. However, the toxicity of beryllium metal and its oxide is usually manifested as a lung disease, resulting from inhaling the powder. [52,53] Now known by the name berylliosis, the disease is understood to arise from inhaling powder particles of sufficient fineness (<5 μm) to reach certain lung membranes where they can cause allergic reaction. Large amounts of particles can overwhelm the lungs and lead to respiratory failure. This response may occur relatively soon after excessive exposure, or after a latent period of several years, and the severity of response is variable from one person to another. In fact, other health risks from beryllium, such as cancer, are now discounted. [53]

Therefore, the use of beryllium, per se, in a fusion device does not constitute a hazard to health. Only inhaling significant quantities of fine powders of beryllium leads to berylliosis. It can be argued that the presence of tritium in a fusion device constitutes a much greater safety issue.

Discussion

The preceding description of neutron damage to beryllium indicates that beryllium components must be designed with care. Response is different as a function of irradiation temperature, and damage at low temperatures can lead to gross property changes following heating to higher temperatures. The basic mechanisms controlling behavior are displacement damage and gas production by transmutation. Response can best be divided into four regimes of temperature: (1) low temperature (<RT) response where point defects are created, but mobility is so low that coalescence is rare, (2) somewhat higher temperatures (RT to 300°C), where point defects are mobile, but gas atoms are practically immobile, (3) still higher temperatures (300 to 600°C), where gas atoms become mobile, and (4) very high temperatures where gas pressure driven swelling becomes dominant (>600°C). The lowest temperature regime has been ignored in this report.

Microstructural evolution in these temperature regimes is as follows. At temperatures from room temperature to 300°C where point defects are mobile but gas atoms are not, point defects coalesce into dislocation loops, and the loops grow and evolve into a complex dislocation network with basal and non-basal Burgers vectors represented. Gas atoms become trapped in the microstructure. Therefore, at these temperatures, beryllium swells at a modest rate, and hardens, which leads to embrittlement. Growth (change in shape under irradiation) can also be anticipated. At temperatures from 300 to 600°C where gas atoms become mobile, gas bubbles form, most visibly on grain boundaries, but they are also probably present on dislocations. The accumulation of helium at bubbles is probably by a mechanism similar to solute segregation, in which impurity atoms are dragged to point defect sinks by interaction with moving point defects. Swelling and mechanical property degradation occur at somewhat different rates in this temperature range than at lower temperatures. At temperatures over 600°C where gas bubbles can come into thermal equilibrium, bubbles grow in response to internal pressurization, leading to enhanced swelling and the creation of very large bubbles on grain boundaries. These large bubbles provide crack nucleation sites resulting in embrittlement without significant increases in strength.

Mechanical property degradation can be severe. Examples are provided showing complete ductility loss in beryllium following irradiation to doses on the order of 10^{21} n/cm². However, it is apparent that large variations in response are possible, depending on fabrication procedures and the resultant product form. Results on more modern materials prepared by powder processing and hot pressing show acceptable ductility and fracture toughness, or lower swelling, and even porous beryllium made in this way gives acceptable ductility and toughness. [31,42] Therefore, it appears that the disappointing results obtained during beryllium fuel cladding development efforts can be mitigated by improved fabrication procedures.

Given the significant property degradation of beryllium under neutron irradiation, it may be necessary to design blanket and first wall areas so that beryllium components are not required to provide structural support. For example, beryllium can be encapsulated in the blanket and can be replaced by plasma spraying procedures on the first wall. The divertor design may pose greater problems.

However, it must be emphasized that available database information is extremely limited with regard to material produced by modern methods. Also, all irradiation tests have been performed using fission reactors

where the major transmutation response is defined by equation (2). As a consequence, effects of lithium and tritium production are overemphasized, and the effects of helium production are somewhat reduced in comparison with the fusion condition where large numbers of neutrons have energies above 2.7 MeV, and therefore equation (1) applies. A more complete database is required, using prototypic materials irradiated at high temperatures appropriate for divertor applications. Also, high energy neutron irradiation is needed.

Conclusions

This review of neutron irradiation effects to beryllium demonstrates that beryllium is degraded by radiation damage both as a result of displacement damage and of transmutation. Swelling and embrittlement are significant with complex temperature dependencies. Degradation in the properties of beryllium can be expected to be of concern for divertor applications, may lead to thermal conductivity changes at high swelling levels in blanket applications, and may contribute to flaking of beryllium first wall coatings. However, more modern fabrication techniques provide materials with greater radiation resistance, and there is cause for hope that modern materials will have sufficient radiation resistance to allow components to operate to fluences on the order of 6×10^{22} n/cm², $E > 1$ MeV or higher.

A much more complete testing program, including high energy neutron irradiation, is required to qualify these newer materials for fusion applications and to determine lifetime limits.

References

- [1] P. R. Thomas and the JET Team, presented at the 9th International Conference on Plasma Surface Interactions and Controlled Fusion Devices, held in Bournemouth, UK, 21-25 May 1990, reproduced in JET-P (90) 41,1,
- [2] M. F. Smith and A. W. Mullendore, J. Nucl. Mater., 122 & 123 (1984) 855.
- [3] W. G. Wolfer and T. J. McCarville, Fusion Tech., 8 (1985) 1157.
- [4] J. B. Mitchell, J. Fusion Energy, 5 (1986) 327.
- [5] K. L. Wilson, R. A. Causey, W. L. Hsu, B. E. Mills, M. F. Smith, and J. B. Whitley, J. Vac. Sci. Technol., A, 8 (1990) 1750.
- [6] V. P. Gol'tsev, G. A. Sernyaev, and Z. I. Chechetkina, Radiatsionnoe Materialovedenie Berilliya, Minsk: Nauka i Tekhnika (1977) 38.
- [7] G. A. Sernyaev, "Swelling of Beryllium in a Mode of High-Temperature Neutron Irradiation," Voprosy Atomnoi Nauki i Tekniki, No 2 (56), (1991) 16, PNL-TR-488.

- [8] L. Sannen and Ch. De Raedt, "The Effects of Neutron Irradiation on Beryllium," presented at 17th Symposium on Fusion Technology, Rome Sept. 14-18, 1992.
- [9] E. Koonen, in Proceedings of the International Symposium on Research Reactor Safety, Operations and Modification, AECL-9926, Vol. 3 (1990) 737.
- [10] V. N. Burmistrov, Yu. D. Goncharenko, V. A. Kazakov, O. Yu. Shvedov, V. A. Gorokhov, and I. B. Kupriyanov, "Certain Aspects of the Radiation Stability of Beryllium with Application to Synthesis Reactor Conditions from Results of Tests in a Fission Reactor," presented at the Second International Conference on Effects of Irradiation on Materials for Fusion Reactors, held September 21-24, 1992 in St. Petersburg, Russia, PNL-TR-487.
- [11] M. Dalle Donne, F. Scaffidi-Argentina, C. Ferrero and C. Ronchi, "Modelling of Swelling and Tritium Release in Irradiated Beryllium," proceeding of ICFRM-6.
- [12] G. A. Sernyaev, "Some Physical Properties of Beryllium as a Material for Neutron Multiplier," in Modular Variant of ITER Ceramic Blanket (ITER, Garching, Germany, July 1990).
- [13] J. M. Beeston, "Properties of Irradiated Beryllium: Statistical Evaluation, EG&G Idaho Report TREE-1063 (1976).
- [14] E. Koonen, "Study of Irradiation Effects and Swelling of Irradiated Beryllium," CEN/SCK Report, Reactor Safety Analysis BR2 Department (1989). (See also [9])
- [15] J. M. Beeston, G. R. Longhurst, L. G. Miller, and R. A. Causey, "Gas Retention in Irradiated Beryllium," EGG-FSP-9125 (1990).
- [16] J. M. Beeston, L. G. Miller, E. L. Wood, Jr., and R. W. Moir, J. Nucl. Mater., 122 & 123 (1984) 802.
- [17] J. B. Rich, G. B. Redding, and R. S. Barnes, J. Nucl. Mater., 1 (1959) 96.
- [18] B. S. Hickman, in The Metallurgy of Beryllium, Inst. of Metals Monograph No. 28 (Institute of Metals, London, 1961) 410.
- [19] J. B. Rich and G. P. Walters, in The Metallurgy of Beryllium, Inst. of Metals Monograph No. 28 (Institute of Metals, London, 1961) 362.
- [20] J. B. Rich, G. P. Walters, and R. S. Barnes, J. Nucl. Mater., 4 (1961) 287.
- [21] C. E. Ells and E. C. W. Perryman, J. Nucl. Mater., 1 (1959) 73.
- [22] S. Morozumi, S. Goto, and M. Kinno, J. Nucl. Mater., 68 (1977) 82.

- [23] G. A. Sernyaev, "The Formation of Helium Bubbles and Energy Phenomena in Beryllium," Voprosy Atomnoi Nauki i Tekniki, No 2 (56), (1991) 82, PNL-TR-490.
- [24] D. McLean in Conference Internationale sur la Metallurgie du Beryllium (Presses Universitaires de France, Grenoble, 1965) 3.
- [25] B. S. Hickman, G. Bannister, J. H. Chute, J. G. McCracken, R. Smith, and J. C. Bell, "Irradiation of Beryllium at Elevated Temperatures," TRG Report 540, UKAEA, 1963.
- [26] B. S. Hickman and G. Bannister, "Irradiation of Beryllium at Elevated Temperatures," Part II, AAEC/E-115, 1963, also issued as TRG Report 532.
- [27] B. S. Hickman and G. T. Stevens, "The Effect of Neutron Irradiation on the Mechanical Properties of Irradiation of Beryllium at Elevated Temperatures," Part II, AAEC/E-115, 1963, also issued as TRG Report 532.
- [28] G.T. Stevens and B. S. Hickman, "Effect of Irradiation on the Mechanical Properties of Beryllium Metal," AAEC/E-133, 1965.
- [29] M. H. Bartz, in Proceedings of the 2nd United Nations International Conference on the Peaceful Uses of Atomic Energy, Vol. 5 (United Nations, Geneva, 1958) 466.
- [30] J. M. Beeston, in Effects of Radiation on Structural Materials, ASTM STP 426, (ASTM, Philadelphia PA, 1967) 135.
- [31] J. M. Beeston, G. R. Longhurst, R. S. Wallace, and A. P. Abeln, J. Nucl. Mater., 195 (1992) 102.
- [32] J. M. Beeston, M. R. Martin, C. R. Brinkman, G. E. Korth, and W. C. Francis, in Symposium on Materials Performance in Operating Nuclear Systems, M. S. Wechsler and W. H. Smith, Eds., CONF-730801 (1973) 59.
- [33] E. D. Hyam and G. Sumner, in Radiation Damage in Solids, I (IAEA, Vienna, 1962) 323.
- [34] G. P. Walters, J. Less Common Metals, 11 (1966) 77. (Earlier version in Conference on the Physical Metallurgy of Beryllium, Gatlinburg, 1963, CONF-170, 138.)
- [35] E. H. Smith, J. L. Liebenthal, B. V. Winkel, J. M. Beeston, and W. C. Francis, in Symposium on Materials Performance in Operating Nuclear Systems, M. S. Wechsler and W. H. Smith, Eds., CONF-730801 (1973) 41.
- [36] J. M. Beeston, Nucl. Engineering and Design, 14 (1970) 445.
- [37] D. S. Gelles and H. L. Heinisch, J. Nucl. Mater., 191-194 (1992) 194.

- [38] V. Barabash, "Brief Review of Be Study for Plasma Facing Component in RF for ITER Reactor," presented at the US/RF Exchange Meeting, September 17-19, 1992, in St. Petersburg, Russia.
- [39] J. R. Weir, in The Metallurgy of Beryllium, Inst. of Metals Monograph No. 28 (Institute of Metals, London, 1961) 395.
- [40] D. L. Harrod, T. F. Hengstenberg and M. J. Manjoine, J. Materials, 4 (1969) 618.
- [41] R. L. Kesterson, Trans. Amer. Nucl. Soc., 14 (1971) 607.
- [42] J. M. Beeston, in Effects of Radiation on Structural Materials, ASTM STP 683, (ASTM, Philadelphia PA, 1979) 309.
- [43] B. S. Hickman, in Studies in Radiation Effects, Series A, Vol. 1, G. J. Dienes, Ed. (Gordon & Breach Science Publishers, Inc., N.Y., 1966) 72.
- [44] R. Sumerling and E. D. Hyam, in The Metallurgy of Beryllium, Inst. of Metals Monograph No. 28 (Institute of Metals, London, 1961) 381.
- [45] R. S. Barnes in The Metallurgy of Beryllium, Inst. of Metals Monograph No. 28 (Institute of Metals, London, 1961) 372.
- [46] Y. Mishima, S. Ishino, and S. Shiozawa, in Beryllium 1977, (The Royal Society, London, 1977) 25/1.
- [47] S. Shiozawa, thesis, "The Electron Microscopic Studies on the Irradiated and Deformed Hexagonal-Close-Packed Metals, University of Tokyo, 1977.
- [48] R. Nagasaki, S. Ohashi, S. Kawasaki, Y. Karita and N. Tsuno, J. Nucl. Sci. Tech., 8 (1981) 546.
- [49] G. P. Walters, C. M. Van Der Walt, and M. J. Makin, J. Nucl. Mater., 11 (1964) 335. (Earlier version in Conference on the Physical Metallurgy of Beryllium, Gatlinburg, 1963, CONF-170, 126.)
- [50] G. J. C. Carpenter and R. G. Fleck, in Beryllium 1977, (The Royal Society, London, 1977) 26/1.
- [51] R. G. Fleck, in Effects of Radiation on Materials, ASTM STP 782, (ASTM, Philadelphia PA, 1982) 735.
- [52] W. Jones Williams, in Beryllium 1977, (The Royal Society, London, 1977) 47/1.
- [53] O. P. Preuss, Fusion Tech., 8 (1985) 1137.

Figure Captions

Figure 1. Swelling in beryllium irradiated at temperatures below 100°C as a function of helium content.

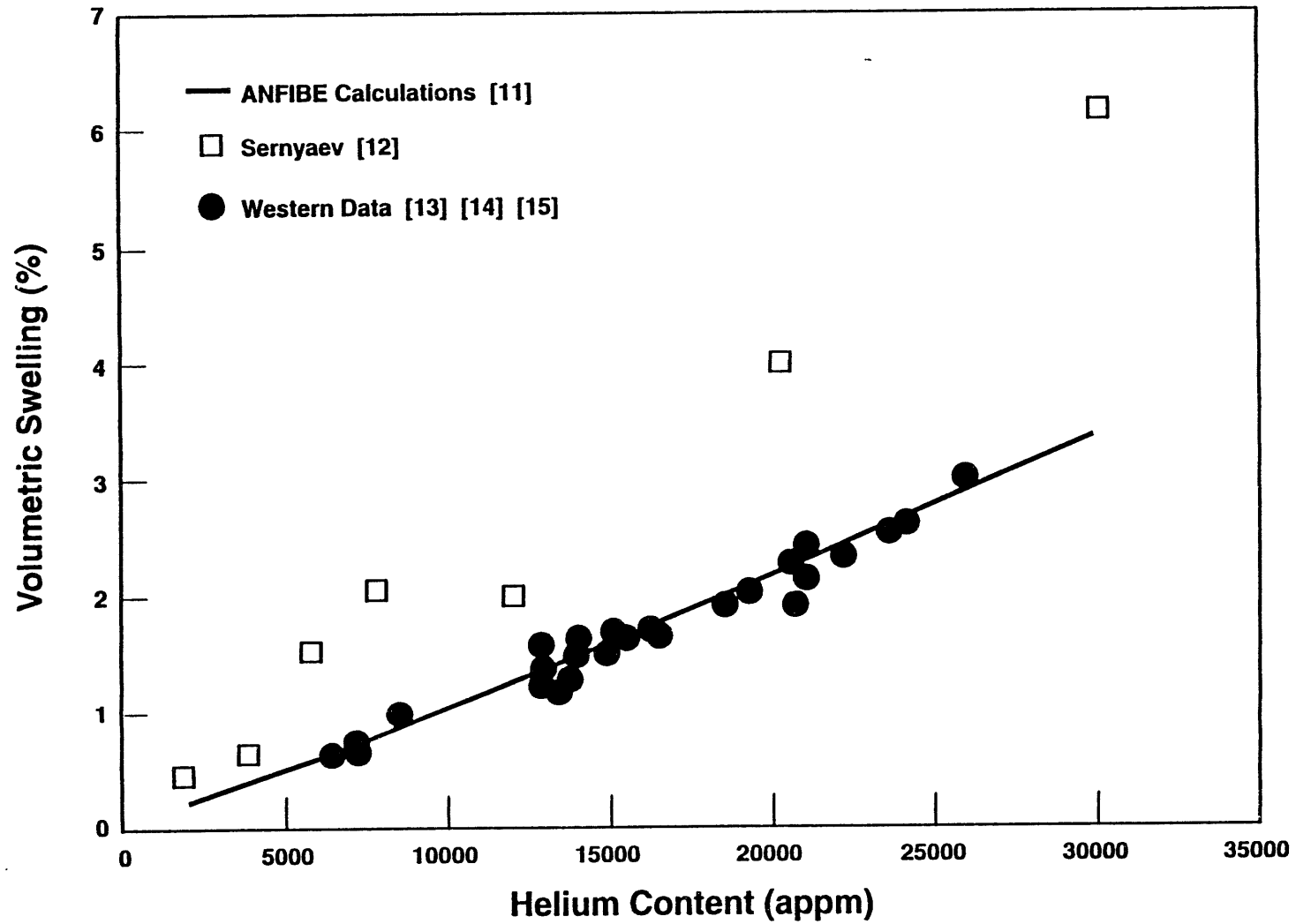
Figure 2. Swelling in Beryllium as a function of irradiation temperature (a) from [7] and (b) from [10]. Notation in (a) is as follows: (1) is single crystal; (2, 6, 7) are hot pressed, grain size (g.s.) 56 μm ; (3,5) are hot pressed, g.s. 600 μm ; (4) is thermally extruded, g.s. 400 μm . Also, (1-3) are at 6×10^{20} , (4,5) are at 5.7×10^{21} , (6) is at 8.9×10^{21} and (7) is at 1.02×10^{22} n/cm^2 , $E > 0.85$ MeV.; notation in (b) is as follows: (1) g.s. 30 μm , (2) g.s. 20 μm , and (3) g.s. 8 to 13 μm and fluences for (1,2) 5.7×10^{21} , and for (3) 3.5×10^{21} n/cm^2 ($E > 0.1$ MeV).

Figure 3. Tensile strength increase (a) or compressive fracture or yield strength (b) as a function of Fluence ($E > 1$ MeV) for beryllium specimens irradiated at 75 to 125°C.

Figure 4. Increase in hardness as a function of fluence for beryllium specimens (a) irradiated at 35 to 100°C and (b) irradiated at 280 to 650°C.

Figure 5. Increase in yield strength as a function of fluence to the 1/3 power for intermediate irradiation temperatures as per Walters. [34] All measurements are in tension and for fluences of $E > 1$ MeV except where noted.

Figure 6. Compressive strength and Elongation as a function of fluence ($E > 0.85$ MeV) for beryllium specimens irradiated at 650°C. [38]



39309001.2

Figure 1. Swelling in beryllium irradiated at temperatures below 100°C as a function of helium content.

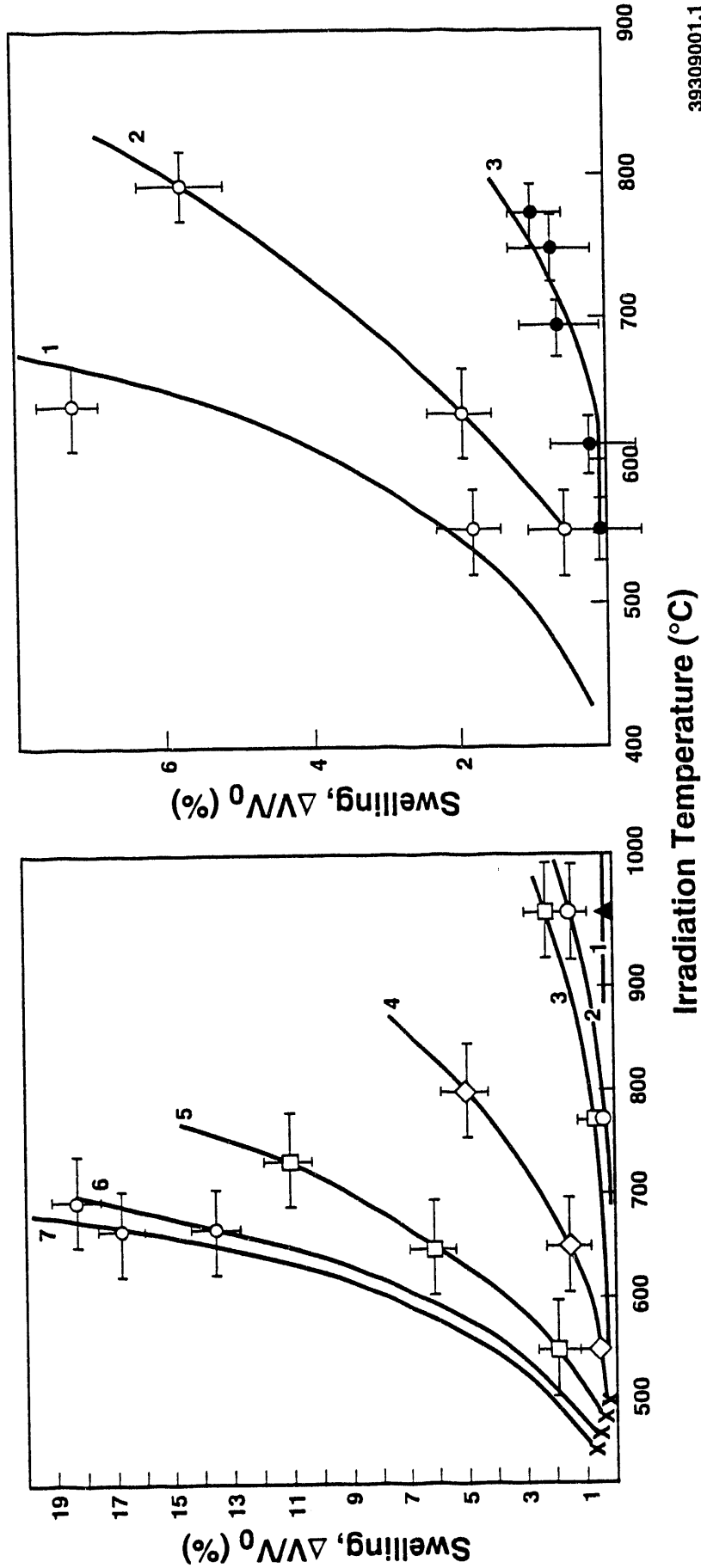
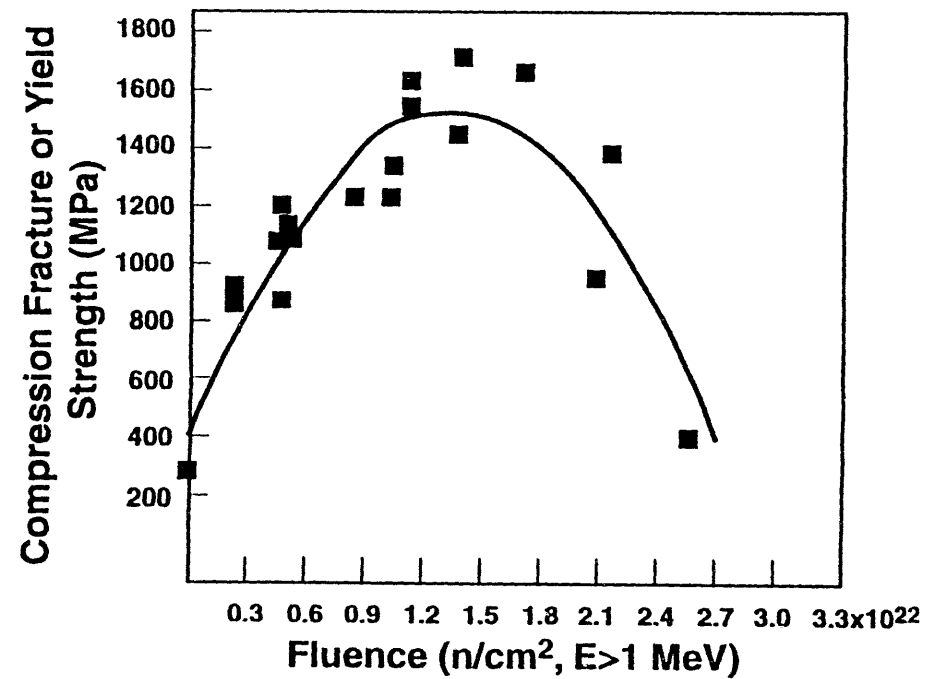
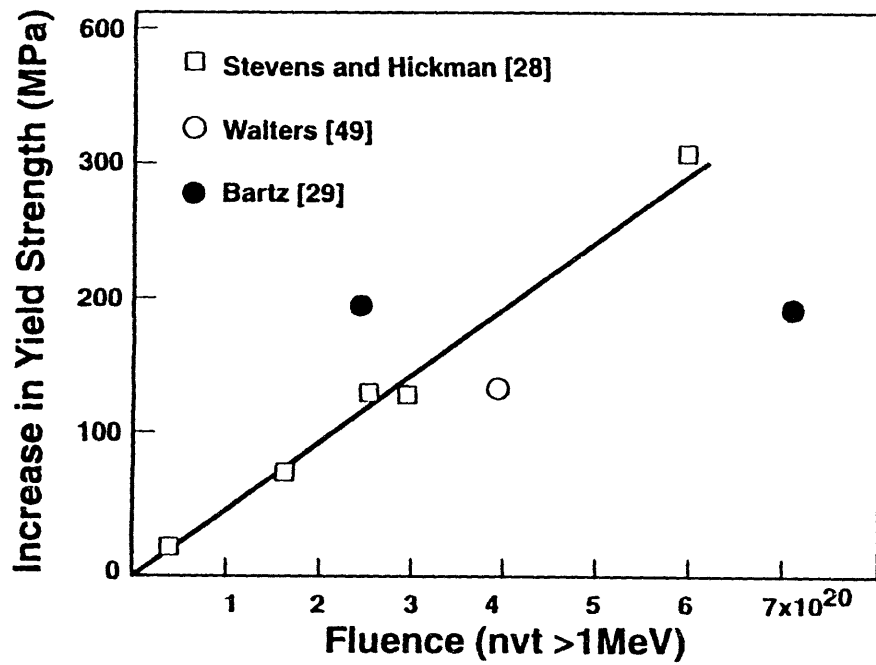
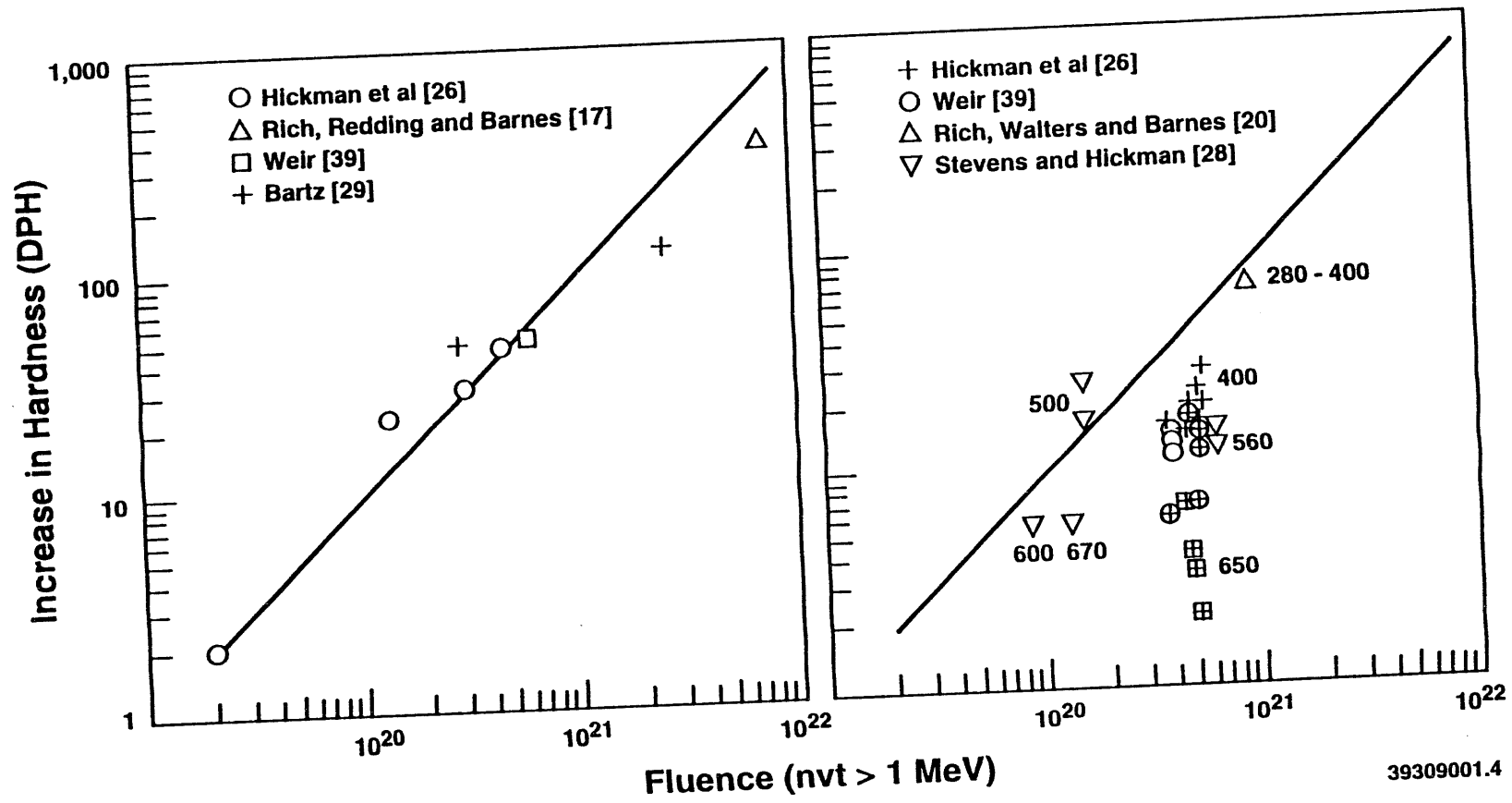


Figure 2. Swelling in Beryllium as a function of irradiation temperature (a) from [7] and (b) from [10]. Notation in (a) is as follows: (1) is single crystal; (2, 6, 7) are hot pressed, grain size (g.s.) $56 \mu\text{m}$; (3, 5) are hot pressed, g.s. $600 \mu\text{m}$; (4) is thermally extruded, g.s. $400 \mu\text{m}$. Also, (1-3) are at 6×10^{20} , (4, 5) are at 5.7×10^{21} , (6) is at 8.9×10^{21} and (7) is at 1.02×10^{22} n/cm^2 , $E > 0.85 \text{ MeV}$; notation in (b) is as follows: (1) g.s. $30 \mu\text{m}$, (2) g.s. $20 \mu\text{m}$, and (3) g.s. 8 to $13 \mu\text{m}$ and fluences for (1, 2) 5.7×10^{21} , and for (3) 3.5×10^{21} n/cm^2 ($E > 0.1 \text{ MeV}$).



39309001.3

Figure 3. Tensile strength increase (a) or compressive fracture or yield strength (b) as a function of Fluence ($E > 1 \text{ MeV}$) for beryllium specimens irradiated at 75 to 125°C.



39309001.4

Figure 4. Increase in hardness as a function of fluence for beryllium specimens (a) irradiated at 35 to 100°C and (b) irradiated at 280 to 650°C.

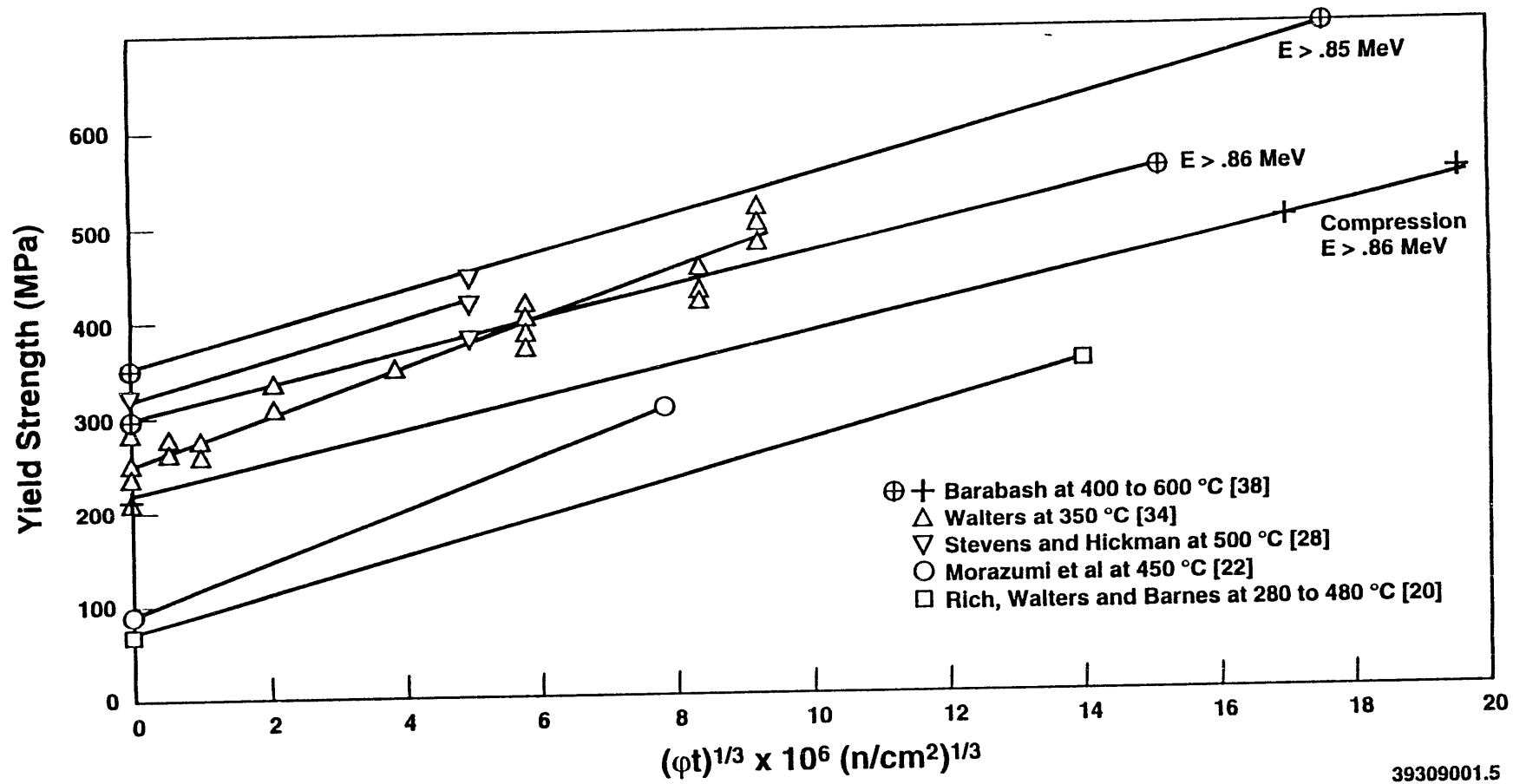
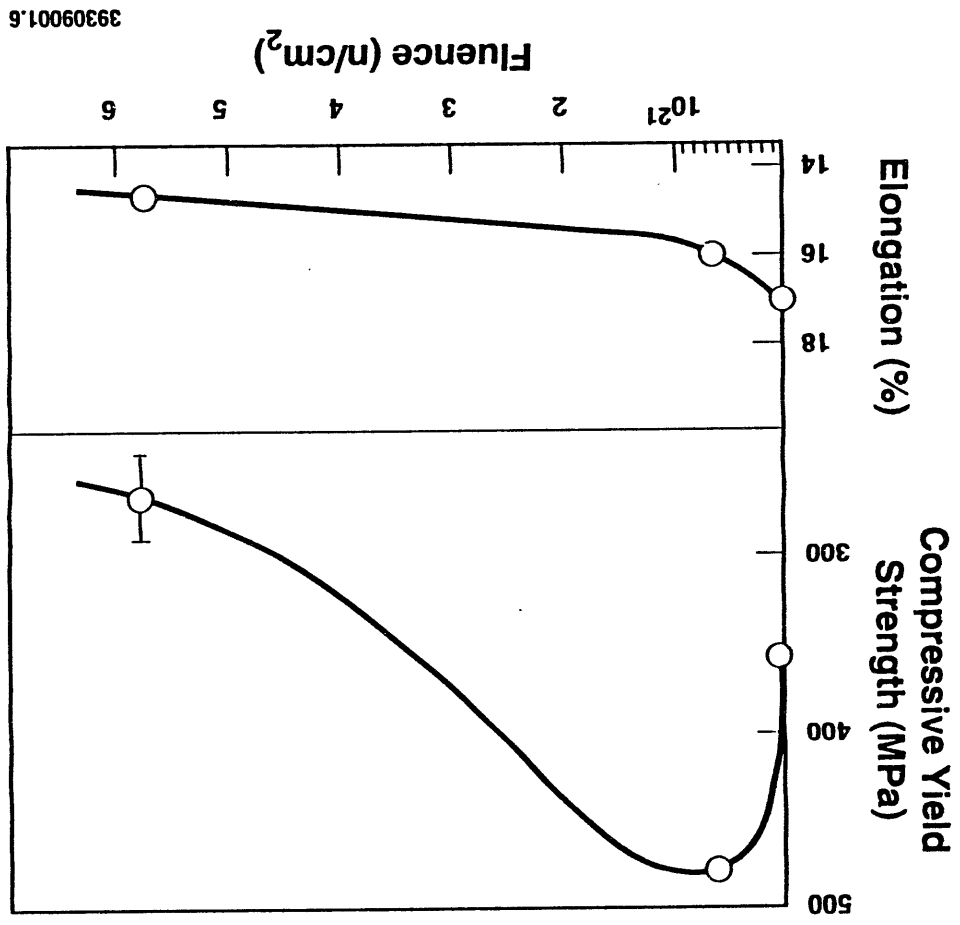


Figure 5. Increase in yield strength as a function of fluence to the 1/3 power for intermediate irradiation temperatures as per Walters. [34] All measurements are in tension and for fluences of E > 1 MeV except where noted.

Figure 6. Compressive strength and Elongation as a function of fluence ($E > 0.85$ MeV) for beryllium specimens irradiated at 650°C. [38]



39309001.6

DATE

FILMED

2 / 8 / 94

END

