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Homogeneous Forming Technology of Composite Materials and its Application to Dispersion Nuclear Fuel

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Abstract

Powder metallurgy processing technique of metal matrix composites is reviewed and its application to process homogeneous dispersion nuclear fuel is considered. The homogeneous mixing of reinforcement with matrix powders is very important step to process metal matrix composites. The reinforcement can be ceramic particles, whiskers or chopped fibers having high strength and high modulus. The blended powders are consolidated into billets and followed by various deformation processing, such as extrusion, forging, rolling or spinning into final usable shapes. Dispersion nuclear fuel is a class of metal matrix composite consisted of dispersed U-compound fuel particles and metallic matrix. Dispersion nuclear fuel is fabricated by powder metallurgy process such as hot pressing followed by hot extrusion, which is similar to that of SiC/Al metal matrix composite. The fabrication of homogeneous dispersion nuclear fuel is very difficult mainly due to the inhomogeneous mixing characteristics of the powders from quite different densities between uranium alloy powders and aluminum powders. In order to develop homogeneous dispersion nuclear fuel, it is important to investigate the effect of powder characteristics and mixing techniques on homogeneity of dispersion nuclear fuel. A new quantitative analysis technique of homogeneity is needed to be developed for more accurate analysis of homogeneity in dispersion nuclear fuel.

1. Introduction

Metal matrix composites(MMCs) reinforced with high strength and high modulus ceramic reinforcements, have received considerable emphasis due to the potential applications as structural materials which can take advantage of their excellent specific strength and specific modulus[1]. It is known that powder metallurgy processing of metal matrix composites results in better mechanical properties with homogeneous microstructure compared to casting process[2-3]. The powder metallurgy process of metal matrix composites consists of mixing, consolidation and secondary processing and the process parameters at each stage need to be controlled to improve the homogenization and densification of metal matrix composites[4].

Mixing of powders is defined as the thorough blending of reinforcements and matrix alloy powders into same nominal composition with the uniform distribution of reinforcements[5]. A billet is formed by either hot pressing in vacuum or hot isostatic pressing. The microstructures of composites are sensitively determined by the consolidation conditions and are also closely related to the mechanical properties[6]. Secondary processing such as extrusion, rolling, forging and superplastic forming is also an important fabrication step of metal matrix composites. Specifically, in order to achieve maximum composite strengthening effect by the addition of whiskers, the whiskers need to be aligned parallel to one direction[7]. The hot extrusion process is the most common deformation process applied to align the whiskers in metal matrix composites[8-9].

Dispersion nuclear fuels consisted of U-compound fuel particles dispersed in metallic matrix are distinguished from the metallic uranium fuel used in early reactors and the UO_2(ceramic pellet) fuel used in most power reactors today[10-11]. The basic idea of a dispersion fuel is to fabricate by powder metallurgy process such as hot extrusion or hot rolling, which is similar to that of SiC/Al metal matrix composite[11]. In order to get high performance dispersion nuclear fuel, the uranium alloys powder need to be homogeneously distributed in a aluminum matrix. However, the fabrication of homogeneous dispersion nuclear fuel is very difficult mainly due to the inhomogeneous mixing characteristics of the powders from quite different densities between uranium alloy powders and aluminum powders.

In this paper, the fabrication processes of composite materials and dispersion nuclear fuels are compared. The improvement on homogeneity of dispersed uranium fuel particles in aluminum matrix is investigated to develop a high density fuel with improved characteristics such as homogeneity, fabricability, compatibility with aluminum, and stable irradiation behavior.

2. Fabrication Process of Metal Matrix Composites

The conventional fabrication process of metal matrix composites includes mixing of reinforcement with matrix powders, consolidation of blended powders and secondary processing of consolidated billet. Fig. 1 shows the conventional processing routes of SiC/Al metal matrix composite via powder metallurgy process.

2-1. Mixing of Reinforcements with Matrix Powders

A mixing step normally follows the selection of the reinforcement and matrix of composite materials. The mixing step ultimately controls the final properties of metal matrix composites, which are intended to achieve[12]. The properties of consolidated composite materials are sensitively dependent upon the homogeneity of the mixture. Generally, dry mixing, wet mixing and ball milling are used for mixing of metal matrix composites. Fig. 2 shows three kinds of mixing techniques used in powder metallurgy process.

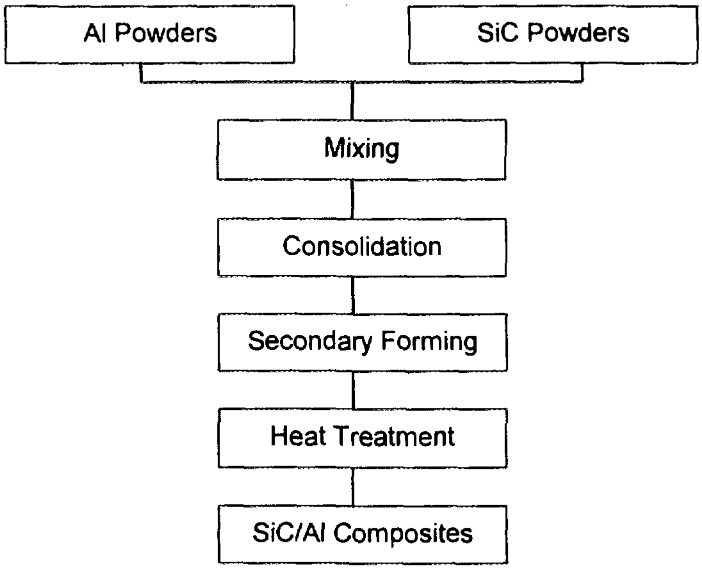
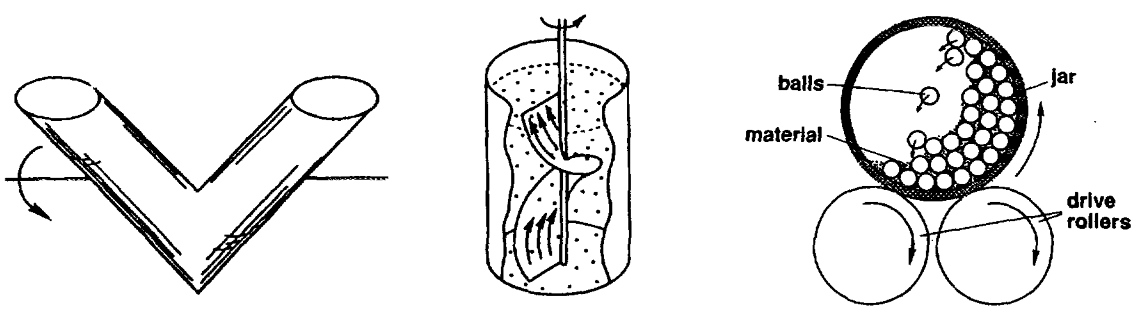


Fig. 1. Fabrication process steps of SiC/Al composites via powder metallurgy process

Dry mixing is easily employed for mixing of metal matrix composite by using rotating cylinder mixer, double cone mixer or V-mixer. The important consideration in dry mixing is that the powder must not fall freely through the air during any stage of mixing because this will always cause segregation[13]. A desirable rotational speed for homogeneous mixing is one which balances gravitational and centrifugal forces[5].

Wet mixing is preferable when the reinforcements like whisker or short fiber are easy to be agglomerated and damaged during dry mixing or ball mill[14]. The method for deagglomeration during wet mixing may involves ultrasonic agitation of alcohol/fiber suspensions[15]. It is more effective to use surface active agents to induce repulsive forces between whiskers or short fibers[16].

Ball milling is used for homogeneous mixing of strongly agglomerated powders[17]. The impact between balls breaks apart the stable agglomerates. The typical mill used for these purposes has been the tumbler ball mill which is a cylindrical container rotated about its axis in which balls impact upon the powder charge[13]. The balls may roll down the surface of the chamber in a series of parallel layers or they may fall freely and impact the powder and balls beneath them.



(a) (b) (c)

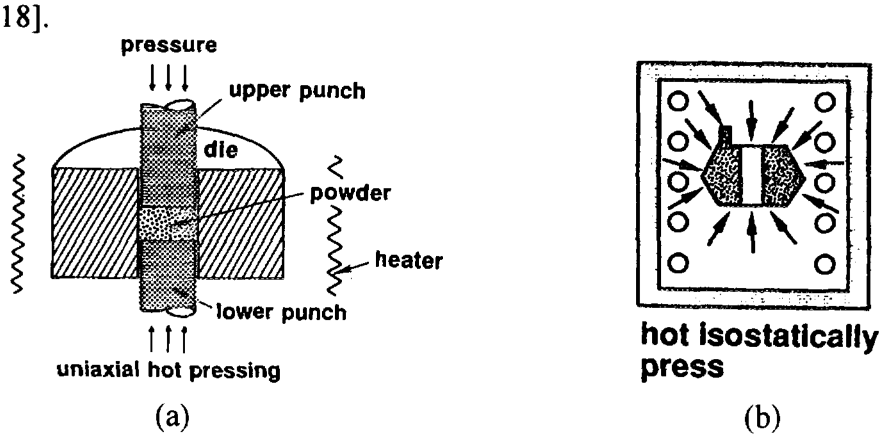
Fig. 2. The mixing methods in powder metallurgy process: (a) dry mixing, (b) wet mixing and (c) ball mill.

2-2. Consolidation of Blended Powders

Final billet fabrication involves cold compaction, outgassing and hot isostatic or vacuum hot pressing as shown in Fig.3[6]. While the principal function of the cold compaction step is to provide a compact have some green strength, it is essential that the cold compaction densities be controlled to insure an open interconnection pore structure. The latter allows exit of the most common secondary forming process has been extrusion[19]. Consolidated billets, having relative density above about 95%, can be fabricated into a wide variety of shapes with almost various gaseous products that are liberated during subsequent heating and outgassing. Reinforcement/matrix blend outgassing involves the removal, through the combined action of heat, vacuum, and inert gas flushing. For example, outgassing of SiC reinforced aluminum metal matrix composites involves removal of adsorbed water from both SiC and aluminum, as well as chemically bound water from the aluminum alloy[12].

Hot pressing is the usual consolidation step by simultaneous application of pressure and temperature[6]. The application of pressure at the sintering temperature accelerates the kinetics of densification by increasing the contact stress between particles and rearranging particle positions to increase density. Because of the increased contact stress at particle to particle interfaces, the energy available for densification is many times higher than for conventional sintering[13]. This result in less porosity, less grain growth and thus higher strength. It can also result in a decrease in the amount of sintering aid required for densification. Hot isostatic pressing benefits from reduced friction between die wall and powder, which is characteristic of rigid die compaction[6]. The uniformity of stress transmitted through the powder mass and the density distribution in the green compacts produced in rigid dies are much less uniform than in isostatic compaction.

The effects of temperature and pressure during hot pressing on the tensile properties and microstructures of SiCw/2124Al composites revealed that the tensile strength of composites increased with increasing vacuum hot pressing temperature, since the aspect ratio of whiskers and density of the composite were improved due to the softening of 2124Al matrix with increasing volume fraction of liquid phase[18]. The vacuum hot pressing pressure was needed to be higher than 70MPa to achieve high densification above 99% of SiCw/Al composite. The vacuum hot pressing pressure above 70MPa was not helpful to enhance the tensile strength of SiCw/Al composite due to the decrease in aspect ratio of SiC whiskers from damage during consolidation[18].



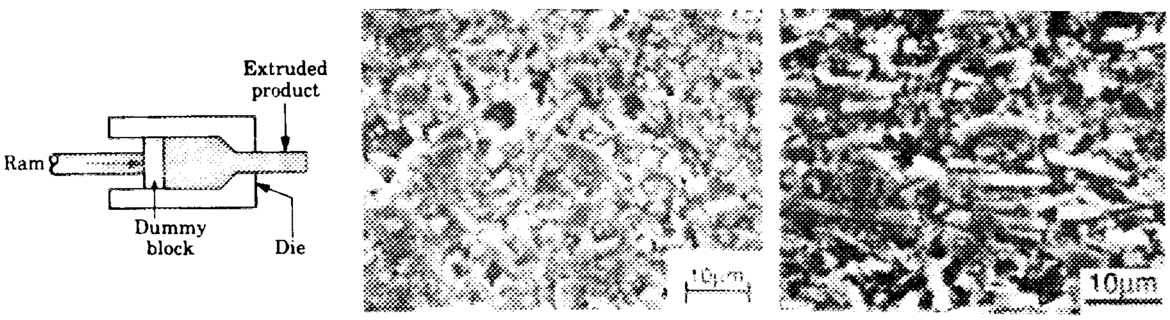
(a) (b)

Fig. 3. The schematic showing of consolidation method of blended powders in powder metallurgy process: (a) hot pressing and (b) hot isostatic pressing.

2-3. Secondary Forming Processing of Consolidated Billet

Secondary forming processing such as extrusion, rolling, forging and swaging is also an important fabrication step of metal matrix composites[6]. The density and homogeneity of metal matrix composites could be enhanced by the secondary forming processing[16]. The full relative density above 99% by secondary forming process. The secondary process conditions, such as temperature, deformation rates, and flow conditions, are uniquely identified with each composite system.

Specifically, in order to achieve maximum composite strengthening effect by the addition of whiskers, the whiskers need to be aligned parallel to one direction. The hot extrusion process is the most common deformation process applied to align the whiskers in metal matrix composites as shown in Fig.4(a). In SiCw/21214Al metal matrix composites, the extrusion improved the alignment of whiskers along extrusion direction but the aspect ratio of whiskers were decreased due to increased damage on whiskers during extrusion as shown in Fig. 4[20]. Higher extrusion temperature results increased tensile strength of SiC_w/2124Al composite due to increased relative density of composites and increased aspect ratio of whiskers. But, if extrusion temperature is too high, surface cracking occurred known as fir-tree cracking[20].



(a) (b) (c)

Fig. 4. (a) The extrusion process for consolidated billet. The scanning electron micrographs of SiCw/Al metal matrix composites (b) before extrusion and (c) after extrusion, showing the effect of extrusion on the aspect ratio and misorientation of whiskers in SiCw/2124Al metal matrix composites.

3. Fabrication Process of Dispersion Nuclear Fuels

Dispersion nuclear fuels consisted of U-compound fuel particles dispersed in metallic matrix are distinguished from the metallic uranium fuel used in early reactors of the UO_2(ceramic pellet) fuel used in most power reactors today[10-11]. The primary requirement for a good composite structure is a homogeneous distribution of the reinforcement in the matrix. Therefore, the basic idea of a dispersion fuel is to isolate the fuel particles by powder metallurgy processes such as hot extrusion or hot rolling, which is similar to that of SiC/Al metal matrix composite.

The RERTR program has begun an aggressive effort to develop dispersion fuels for research and test reactors with uranium densities of 8 to 9gU/cm^3 at the same time required homogeneity and formability criteria should be considered an unqualified success[11]. High-density uranium compounds are listed in Table 1. Many of these compounds offer no real density advantage over U_3Si_2 and have less desirable fabrication and performance characteristics as well. Of the higher-density compounds, U_3Si has approximately a 30% higher uranium density but the density of U_6X compounds would yield the factor of 1.5 needed to achieve 9gU/cm^3 uranium loading. Fig. 5 shows the conventional fabrication process of dispersion nuclear fuel via powder metallurgy process, which is similar to that of SiC/Al metal matrix composite.

3-1. Production of Fuel Powders

Uranium aluminide and uranium silicide are produced either by arc melting or by induction melting and casting. For production of U_3Si powder, various standard technique is used to comminute to powder for example, hammer milling, ball milling, machining of the resulting chips or use of a shatterbox[21]. Typically the particle size is limited to <150μm (in many cases to <125μm) and up to 40wt% of the powder can be <44μm in size. Recently, Kim and Kuk have devised a method for producing spherical U_3Si and U_3Si_2 powders with reproducible particle size distribution[21]. Also, U_3O_8 has been produced by several methods. The U_3O_8 powder used in the HFIR and several other U.S. research reactor is produced by the peroxide precipitation process, calcining in a low grade nitrogen atmosphere at 800°C, and calcining again in air at 1400°C[10].

Table 1. Nominal Density, Uranium Content and Melting Point of Uranium Compounds[11].

Compound	Density, g/cm^3	U-Density, gU/cm^3	Melting Point, °C
UO_2	10.9	9.7	2750
U_4O_9	11.2	9.7	(a)
UC	13.6	13.0	2400
UN	14.3	13.5	2650
UAl_2	8.1	6.6	1590
U_3Si_2	12.2	11.3	1650
U_3Si	15.4	14.8	930(b)
U_6Ni	17.6	16.9	790(c)
U_6Fe	17.7	17.0	815(c)
U_6Mn	17.8	17.0	725(c)

(a)Transfors to UO_2 at high temperature, (b)Peritectoid temperature (c)Peritectic temperature.

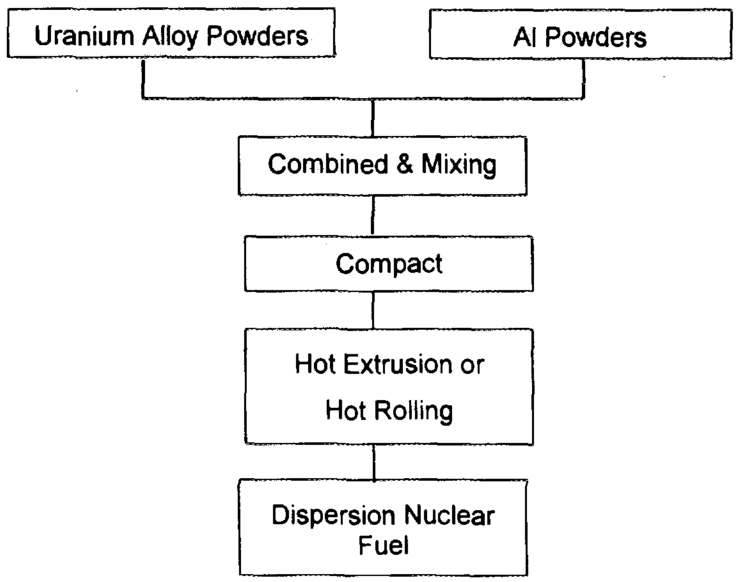


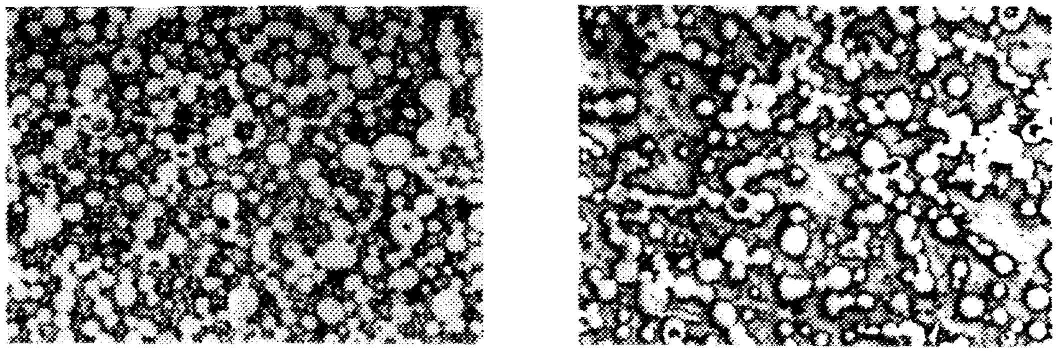
Fig. 5. Fabrication process of U/Al dispersion nuclear fuel via powder metallurgy process.

3-2 Mixing of Dispersion Nuclear Fuel

The objective of mixing is homogenization, manifesting itself in a reduction of concentration within the agitated system. There are several factors affecting the mixing characteristics of powder materials. The particle shape affects the flow characteristics of a mass of particles during mixing process[22]. Smooth, round, spherical particles tend to flow readily than rough irregularly shaped particles. Particle size is also of importance[23]. Segregation of particles according to relative sizes can occur whenever a solid mass comprised of a range of particle sizes is moved[5]. Usually, the smaller particles rise to the top of the mass and the larger particles settle to the bottom. Also, if a mass of particles of widely different densities are being blended, segregation will occur similar to that encountered when blending widely different-size particles[22]. Moisture content is another factor that influences the flow of solids. The sticky particles retard flow and are a deterrent to the mixing process especially if they stick or adhere to the walls of the mixer or cause agglomeration of particles.

Not only do the particulate solids properties influences their mixing characteristics, but also the equipment geometry[13, 22]. The size and shape of body and stirrer(if used) affect the particle flow patterns and velocities. Even the point of addition of ingredients and the surface finish of equipment parts have some effect on the particle movements within the mixer. The mixer rotational speed has a major effect on particle movement and rate of mixing[13, 22]. The sequence of addition of ingredients can also have a major effect on mixing.

Uranium alloys powder and aluminum powders were blended using drum-type blender with various fuel fractions[25]. Typical micro-graphs of dispersion nuclear fuel cross section are shown in Fig. 6(a). However, sometimes aluminum rich regions were found as shown in Fig. 6(b). This inhomogeneous microstructures is occurred during mixing the powders due to the widely different densities between U_3Si and aluminum powders. Therefore, it is important to improve the homogeneity in blended powders of U-alloy powder and Al powders.



(a) (b)

Fig. 6. (a) Typically microstructure of homogeneous by distributed U_3Si fuel particles in Al matrix and (b) microstructure of inhomogeneous by distributed U_3Si fuel particles illustrating aluminum rich area.

3-3 Consolidation and Secondary Forming of Dispersion Nuclear Fuel

After mixing the powders, compaction is widely used to consolidate the blended powders. The compaction process is important to control the green density of compacts, which influences the densification of the samples during sintering and secondary forming process[26]. In generally, cold die compaction is performed to produce a compact billet of dispersion nuclear fuel[11].

There are two types of processes for secondary forming of dispersion nuclear fuel. Extrusion process is used to produce fuel tubes or rods, for examples, using the KMMR[25]. The other is roll-bonding process for producing fuel plates[10,27]. For extrusion process, compacted billet is extruded up to the diameter for fuel rods about 400°C. For rolling-process, the compact is placed into an aluminum alloy frame, and two cover plates are welded on to produce a rolling billet. Some fabricators vacuum degas the compact before the rolling billet is assembled in order to remove moisture or lubricant remaining from the compaction process. The rolling billet is then preheated and rolled through several passes with intermediate reheating until the thickness reaches 110-120% of the final plate thickness. Rolling temperatures range from about 425°C to about 500°C, depending on the particular cladding alloy being used. We can not expect to increase the fuel volume fraction significantly, if at all, beyond 53%. Thus our only hope lies in finding a much-higher-density fuel than U_3Si_2 with however, similar characteristics such as homogeneity, fabricability, compatibility with aluminum, and stable irradiation behavior.

4. Characterization of Homogeneity in Composite Materials

Several techniques have been proposed to characterize the homogeneity of composite materials processed by powder metallurgy. However, the current techniques are all limited to the qualitative comparison of homogeneity in multi-phase materials.

4-1. Characterization of Homogeneity by Composition Analysis

The homogeneity of the mixture may be evaluated from the scatter compositions of samples, which can be expressed on a weight basis[24]. For examples, the composition of a binary mixture of the material A and B containing the weight proportions W_A and W_B of both constituents is given by the relations

$$P_A = \frac{W_A}{W} \quad \text{and} \quad P_B = \frac{W_B}{W} \qquad (1)$$

where P_A and P_B are the mean concentrations of the individual constituents in the mixture and W is the total amount of the mixed material. Therefore

$$P_A + P_B = 1 \qquad (2)$$

For judging the function of the mixture a set of small samples of weight w is taken from the bulk of the mixture. For an ideal mixture the sum of the proportions by weight of all samples gives the respective weight of each component, in the total mixture;

$$X_{A_i} = \frac{w_A}{w} \quad \text{and} \quad X_{B_i} = \frac{w_B}{w} \qquad (3)$$

$$X_{Ai} + X_{Bi} = 1 \qquad (4)$$

$$\sum_{i=1}^{N} X_{A_i} + \sum_{i=1}^{N} X_{B_i} = P_A + P_B = 1 \qquad (5)$$

In a random mixture, the values X_{Ai}, X_{Bi} always differ from the values required for perfect mixing and are given by P_A and P_B. Therefore, dispersion of the weight frequencies X_{Ai} and X_{Bi} is existed. The homogeneity of mixture is then given by the ratio sample standard deviation and standard deviation of the weight frequencies of an equilibrium mixture.

4-2. Characterization of Homogeneity by Microstructure Analysis

Microstructure of materials are studied experimentally using various methods of imaging to resolve specific microstructural elements. These methods include transmission microscopy through thin slices and reflected microscopy on surfaces and polished sections. In all cases, attention is focused on images of the microstructure and these, in many cases, are found to be of considerable complexity. The characterization of microstructures usually requires an analysis of a large number of complicated images with each of them containing the microstructural elements required as well as some artifacts produced by the imagining technique[28]. The term image processing is used to describe operations that are performed on images of microstructures in order to correct them or to make more accessible to quantitative analysis.

Whatever the imaging technique, it produce a 2-D image that can be recorded either on a film or displayed on a monitor. An image contains specific patterns of points and regions, which differ in their intensity or color. From this point of view, the image is a function of two spatial coordinates, for instance (x,y), which ascribes to each point on the plane a specific value of point image intensity. Two-dimensional images of the microstructure can be viewed as sets of different color/gray level dots filling the picture. This image can be divided into a finite number of elements. These pixels forming the image are distinguished by different gray level. If the intensity of the gray tone is measured on a scale ranging form 0 to N_b, then the image analyzed can be represented as a matrix[28].

To analyze the homogeneity, the particles are characterized by a distribution function which defines the relative number of particles of size. This distribution function can be of a discreet of continuous type. Two of these characteristics of the distribution include;(a) some measure of the value about which the observations tend to center, and (b) the spread or variation of the observations about the central value. The measure described in (a) above is called the measure of central tendency and may be expressed as the median, mode, or most commonly, the arithmetic mean. The measures which describe the spread or variation of observations are the standard deviation and the variance[24,28].

4-3. Characterization of Homogeneity by Radiograph Analysis

This method is suitable for testing the homogeneity of metals, either in solid or powdered form, and finely ground oxide materials that are intended for use as reference materials in X-ray emission or optical emission spectroscopy, or both[27]. This procedure, which is based on statistical methods, consisted of stepwise instructions for testing homogeneity of candidate reference materials and requires that repeated measurements on the same specimen have sufficient precision, that is, repeatability. In order to do X-ray emission spectroscopy test procedure, select optimum instrumental conditions to assure adequate count rates from each element to be tested in the specimens. It is needed to select a counting time that is long enough to minimize the random error due to counting, and measure the element of interest on the specimens. And repeat the measurements of X-ray intensity until a minimum of four sets have been made, and examine the data and discard any values which have been determined to be outlier. After this procedures, the calculation can be done to determine homogeneity of a 95% significance level for this procedure[27]. Fig. 7 shows the homogeneity data point plot by radiography attenuation.

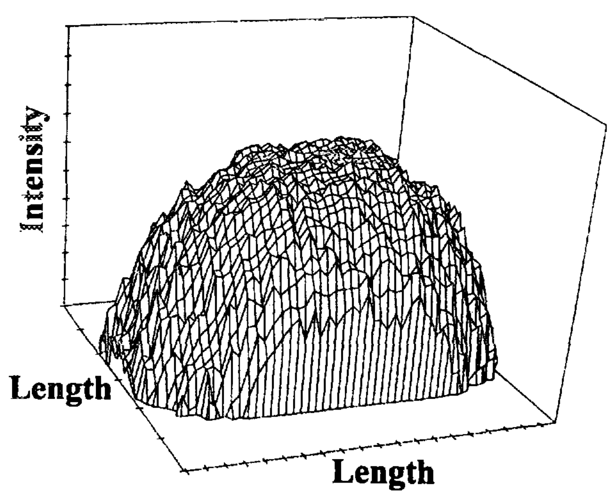


Fig.7 Homogeneity data point plot of U_3Si_2 powders[27]

γ-scanner is used to examine the homogeneity of the fuel cores. U-235 mass and active fuel length are evaluated on the basis of the 186 keV gamma emission intensity. Self- and cladding absorption are corrected on the basis of the 136, 265 and 401 keV gamma transmission intensities. We can evaluate that simultaneous acquisitions of the combined emission and transmission gamma spectrum and of the emission profile by front and/or rear scanning, on-line data processing and printing, plotting of spectrum and profile such as Fig. 7.

In order to develop high performance dispersion nuclear fuel, the improvement of inhomogeneity of U fuel particles in Al matrix is the key technology. It is required to investigate the effect of powder characteristics, such as average size, size distribution, shape and density, on homogeneity after mixing of dispersion nuclear fuels. In addition, a new characterization technique of homogeneity is needed to be developed for quantitative analysis of homogeneity in extended dispersion nuclear fuels.

5. Conclusions

Powder metallurgy processing technique for metal matrix composites is reviewed and its application to process homogeneous dispersion nuclear fuel is considered. The homogeneous mixing of reinforcement with matrix powders is very important step to process metal matrix composites and performed via dry mixing, wet mixing and ball mill process. The blended powders are consolidated into billets using hot pressing or hot isostatic pressing followed by secondary forming processing, such as extrusion, forging, rolling or swaging. Dispersion nuclear fuels is a composite materials in which U-compound fuel particles dispersed in Al matrix. The fabrication process of dispersion nuclear fuel is typical powder metallurgy process, which is identical to that of metal matrix composite. When the particles of uranium alloy powders and aluminum powders are mixed, homogeneous mixing is very difficult due to the large difference in densities between uranium alloy powders and aluminum powders. In order to develop high performance dispersion nuclear fuel, the improvement of inhomogeneity of U fuel particles in Al matrix is the key technology. It is required to investigate the effect of powder characteristics, such as average size, size distribution, shape and density and mixing process parameters, on homogeneity of dispersion nuclear fuels. In addition, a new characterization technique of homogeneity is needed to be developed for quantitative analysis of homogeneity in dispersion nuclear fuels.

6. References

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