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EFFECT OF ULTRASONIC TREATMENT ON THE CONSOLIDATION OF ODS STEEL $FeCrY_2O_3$ PROCESSING WITH CAPSULATED SINTERING PROCESS

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ABSTRACT

EFFECT OF ULTRASONIC TREATMENT ON THE CONSOLIDATION OF ODS STEEL $FeCrY_2O_3$ PROCESSING WITH CAPSULATED SINTERING PROCESS. A method on the synthesis of ODS (Oxide Dispersion Strengthened) steel for advanced nuclear material was performed by ultrasonic treatment to improve the consolidation process. The raw material of Fe, Cr and Y_2O_3 powder with the composition of Fe-15 wt% Cr, 0.5 wt% and Ytria (Y_2O_3) as disperzoid were processed by the powder metallurgy method with the main process of pre-alloying, iso-compaction and sintering process. The pre-alloying process was carried out by mixing the alloying elements using ultrasonically treatment at frquency of 20 kHz with variation of 40, 50 and 60 % amplitude. Iso-compaction process was done using the load of 800 psi to obtain a pellet-shaped sample, then continued by the sintering process for consolidation. The sintering process was done in two stages, 1-step sintering and 2-steps sintering, using the heating furnace by putting the sample in a quartz capsule to prevent oxidation attack. Scanning Electron Microscopy-Energy Dispersive X-ray Spectroscopy (SEM-EDS), X-ray Diffraction (XRD), and Micro-hardness tests were carried out to analyze the microstructure and phase formation in relation to the consolidation process. The highest hardness occurred in the addition of treatment with an amplitude of 60% which produces a micro structure with the most fine grain. For 1-step sintering process, the highest hardness of 134.51 VHN obtained at 40 % amplitude. The hardness of the alloy depends on the size of the grain boundary associated with the difficulty of the dislocation movement.

Keywords: ODS steel, Ultrasonic, Ytria, Powder metallurgy, Sintering, Microhardness

ABSTRAK

EFFECT OF ULTRASONIC TREATMENT ON THE CONSOLIDATION OF ODS STEEL $FeCrY_2O_3$ PROCESSING WITH CAPSULATED SINTERING PROCESS. Metode untuk mensintesis baja ODS (Oxide Dispersion Strengthened) sebagai material maju nuklir telah dilakukan melalui perlakukan ultrasonik guna meningkatkan proses konsolidasi. Bahan dasar berupa serbuk dari Fe, Cr dan Y_2O_3 dengan komposisi masing-masing dalam persen berat Fe-15 wt% Cr, dan 0.5 wt% Ytria (Y_2O_3) sebagai bahan terdispersi diproses dengan metode Metalurgi Serbuk dengan proses utama yaitu proses pemaduan awal, proses kompaksi dan proses sintering. Proses pemaduan awal dilakukan dengan mencampur elemen-elemen pemadu menggunakan proses milling yang disertai dengan perlakuan ultrasonik dengan variasi amplitudo masing-masing 40, 50, dan 60 %. Proses kompaksi dilakukan pada beban sebesar 800 psi guna mendapatkan sampel bentuk pelet, lalu diteruskan dengan proses sintering untuk konsolidasi. Proses sintering dilakukan dalam dua jenis proses yaitu sintering satu tahap dan sintering dua tahap menggunakan tungku pemanas dengan menempatkan sampel didalam kapsul kuarsa guna menghindari oksidasi. Pengujian dengan Scanning Electron Microscopy-Energy Dispersive X-ray Spectroscopy (SEM-EDS), X-ray Diffraction (XRD), dan Microhardness dilakukan guna menanalisa struktur mikro dan pembentukan fasa dalam kaitannya dengan proses konsolidasi. Kekerasan tertinggi terjadi dalam perlakuan ultrasonik dengan amplitdo 60% yang mana menghasilkan struktur mikro paling halus. Untuk proses sintering 1-tahap, kekerasan paling tinggi sebesar 134.51 VHN diperoleh pada amplitusdo 40 %. Kekerasan paduan bergantung pada ukuran butir yang diasosiasikan dengan kesulitan pergerakan dislokasi.

Kata kunci: Baja ODS, Ultrasonik, Ytria, Powder metallurgy, Sintering, Microhardness

INTRODUCTION

ODS steel is an advanced material selected as a candidate for nuclear material, especially used for fuel cladding material of advanced nuclear power plant [1]. The use of ODS steels as a candidate for cladding material, due to its superiority in strength and creep resistance and has good nuclear properties at high temperatures [2]. A ferritic ODS steel type in Fe-Cr alloy matrix with a composition of about 15% Cr dispersed with Ytrium-oxide exhibits good character material that maintains its mechanical properties up to 1000 ^{o}C [3-5]**.**

Increasing the strength and creep resistance of ODS steel at high temperature is mainly caused by the dispersion of the Ytria (Y_2O_3) which functioned as a pinning to inhibit the dislocation movement [6]. Ytria is a rare the earth oxide which is widely used as a disperse oxide because it is non-toxic [7]**,** has a high melting point value and the structural is stable at 2410 ^{o}C [8]. Homogeneous distribution of Ytria in the ferritic ODS steel of Fe-Cr matrix is take an important role in increasing the mechanical properties of the ODS steel [9].

Therefore, the ODS steel is currently produced by based on the powder metallurgy method which successfully disperse the oxide homogeniously. This method can beperformed by the processing step of mechanical alloying, isostatic compaction and continued by the sintering process for consolidation [10-12]. The alloying process in the synthesis of ODS steel can also increase the grain boundary that also increase the strength.

An important challenge in this method is the process of consolidation quality relating to cases of homogeneity of oxide dispersion and also the alloying process [13-15]. Improvement of the alloying and sintering process were now performed to obtain good consolidation. The use of ultrasonic in the pre-alloying process showed a significant effect on the consolidation of the ODS alloys[16,17]. However, to obtain optimal consolidation process it is necessary to conduct some experiments for the selection of power, time and amplitude.

This experiment was focused on consolidation of ultrasonically varied amplitude microalloyed ODS steel of Fe-Cr powders. The consolidation process was carried out in the heating furnace with sample in a capsule to prevent the oxidation in high temperature through 1-step sintering process and 2-steps sintering process. Effect of ultrasonic treatment with variation of amplitude will effect on cavitation size and then will effect on microalloyed particle size. The first step sintering of 2- steps sintering so that the densification is active in the final sintering step. Then, the characteristics of hardness of alloys are dominated by the matrix (Fe-Cr).

MATERIALS AND METHODS

The experiment was done at Center for Science and Technology of Advanced Materials Laboratorium facilities-BATAN, 71 building of Puspiptek area. The experiment was carried out with the operation step as illustrated in the block diagram in Figure 1.

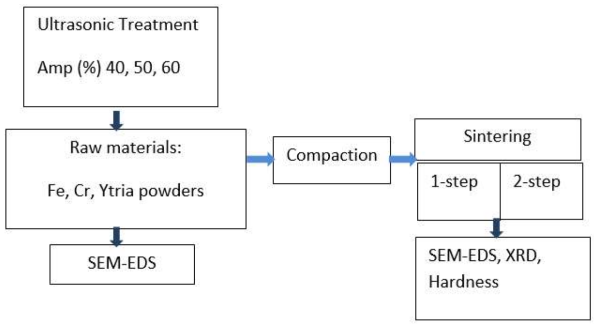

Figure 1. Block Diagram of ODS steel processing.

The raw materials are Fe, Cr, and Y_2O_3 powders from Aldrich with 99,9% purity. Those materials were mixed and alloyed with the composition of Fe 84.5, Cr 15 and Y_2O_3 0.5 wt %. The mixing and pre alloying were performed by the ball milling process with ultrasonic treatment. The ultrasonic of horn type in toluene solution at frequency of 20 kHz for 50 hours was used to improve the pre-alloying process and the homogeneity of Ytria distribution [14,15,18]. Three different amplitude modes of 40%, 50% and 60% were applied on the process, therefore called samples of A, B, and C respectively. The microalloyed powders used in this experiment was found from reference [18] result. Each of mixture was then compacted in pellet shaped with 8000 psi load-pressure using isostatic pressure. Consolidation of the sample was carried out by sintering process in the heating induction furnace. To prevent the sample from the oxidation attack during the sintering process the sample was put in a quartz tube capsule. To evaluate the relation between the sintering process parameters and the consolidated of sample, the sintering process was done in 1-step sintering process was carried out at 1300^{O}C for holding time of 2 hours, and the 2-steps sintering process was carried out at 1000 ^{o}C with holding time for 1 hour and continued to second step 1300^{O}C with holding time for 2 hours. The description of samples were described in Table 1.

Table 1. Sample treatment on ODS steel processing.

Sample	Ultrasonically Treatment at amplitude (%)	Sintering process
A	40	1 and 2-Steps Sintering
B	50	1 and 2-Steps Sintering
C	60	1 and 2-Steps Sintering

Effect of the amplitude mode in the ultrasonic treatment to the microstructure of sample during the milling process was evaluated by the Scanning Electron Microscopy Energy Dispersive Spectroscopy (SEM-EDS, type SEM JEOL JSM-6510LA equipped with EDS) test. Analysis of the consolidation behavior for each sample was then performed based on the micro structure and properties of the sample after sintering process. Characterization of sample was carried out by analyze the morphology observed by SEM-EDS test. The XRD test of SHIMADZU XRD with Cu-Kα source was also performed to evaluate the phase formation of the Fe-Cr alloy. Mechanical properties of the consolidated samples were studied using the Vickers microhardness (HV) measurement.

RESULTS AND DISCUSSION

Morphology of samples with ultrasonic pre-microalloyed Fe-Cr-Y$_2$O$_3$powders observed by SEM-EDS were depicted in Figure 2 (microalloyed powders used from as. The elemental analysis of ultrasonically pre-microalloyed as depicted in Figure 2 are listed in Table 2.

Table 2. Elemental composition contained in ultrasonically pre-microalloyed Fe-Cr-Y$_2$O$_3$ alloy using SEM-EDS.

Amplitudo	Fe content	Cr content	Y_2O_3 content
40	87.98	10.09	1.93
50	90.50	9.50	0
60	93.83	6.17	0

As depicted in Figure 2, the powders consists of microalloyed Fe-Cr-Y$_2$O$_3$, Fe-Cr, Fe and Cr particles. This micro alloyed was confirmed by elemental contents as indicated in Table 2. Effect of ultrasonic treatment can be observed in the morphology of sample with initial

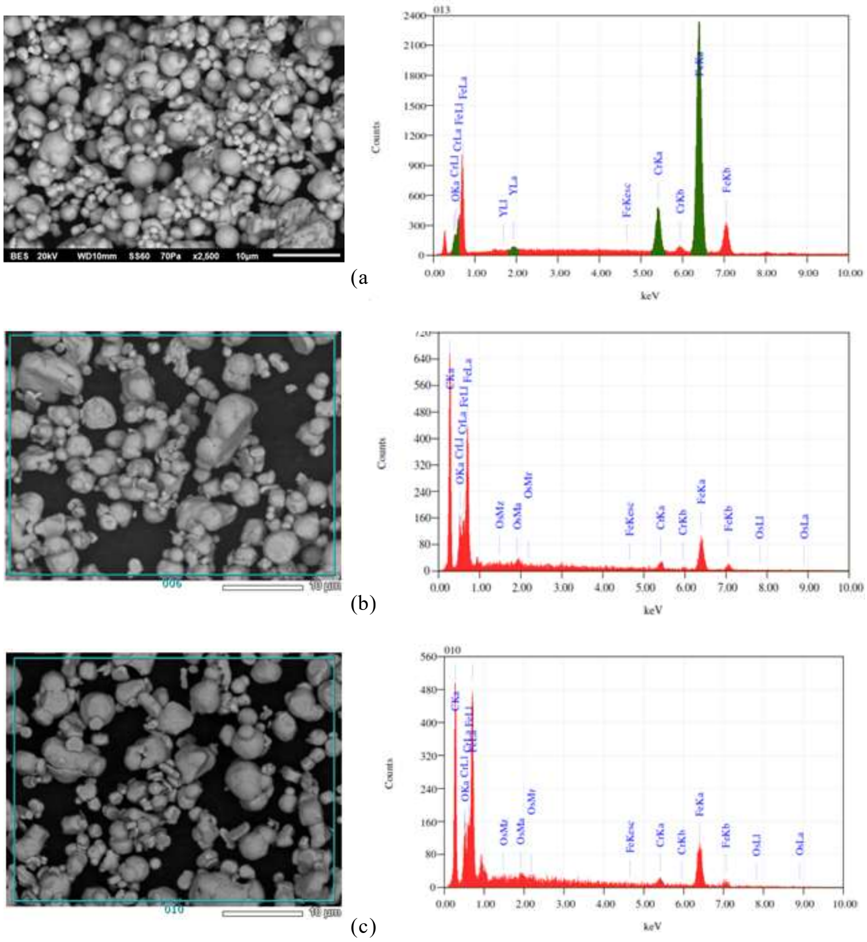

Figure 2. Microstructure ofFe-Cr-Y$_2$O$_3$ particles after ultrasonically treatment, with EDS profile of (a). sample A, (b). sample B, and (c). sample C [18].

necking of micro elements showing the pre-alloying process. These results has been found by references of [14,15,19]. This phenomenon may be explained that microalloying process using acoustic probe in toluene solution resulted cavitation explosion that can produce jetting with high pressure and very high temperature. These results can be used for reducing the particle size and even alloying metals [14,19-21]. The alloying process was then continued in the sintering process by merge two or more elements utilize heat from the furnace. The sintering process also consolidated the elements to increase the density and the mechanical properties of the alloy. In sintering process, Fe elements as the main alloying has melting point of 1538^{O}C with the optimal recrystallization on 0.7Tm [22]. The sintering process was done at temperature of 1300 C for optimal recrystallization so the powders diffused to form link between particles each other that can be reduced the voids between particles and be minimized crack initiation [23].

Morphology for 1-step sintering observed using SEM-EDS are depicted in Figure 3. The elemental analysis of 1-step sintering on ultrasonically pre-microalloyed Fe-Cr-$Y_2O_3$was listed in Table 3.

The product of 1-step sintering process resulted lamellar pattern with finer particle size. This phenomenon caused by homogenization of micro size particles during

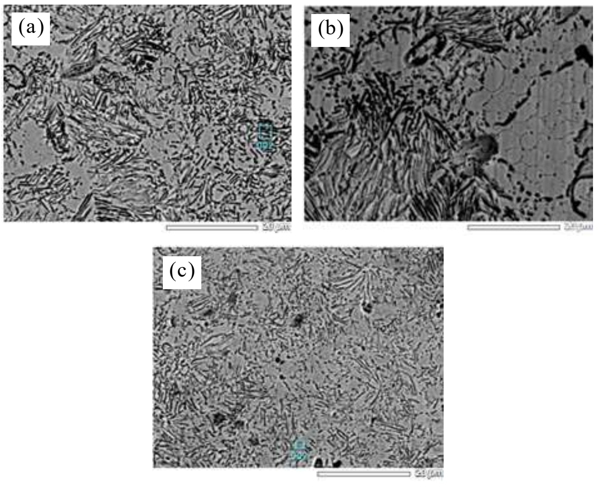

Figure 3. Microstructure of 1-step sintering Fe-Cr-Y_2O_3 carried out using SEM-EDS of ultrasonically treatment at (a). 40% of amplitude, (b). 50%, and (c). 60%.

Table 3. Elemental composition contained in 1-step sintering of ultrasonically pre-microalloyed matrix Fe-Cr-Y_2O_3 alloy using SEM-EDS.

Sampel	Amplitudo (wt %)	Fe content (wt %)	Cr content (wt %)	Y content (wt %)
A	40	82.42	15.83	1.75
B	50	82.06	16.74	1.2
C	60	86.30	13.49	0.21

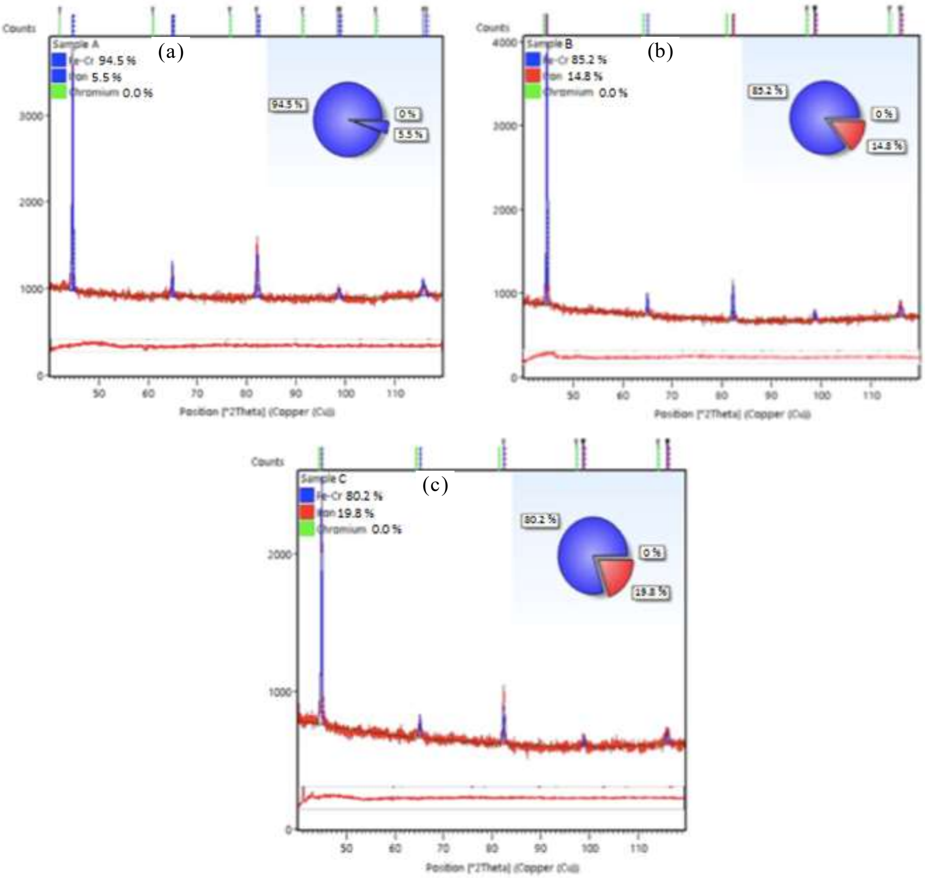

Figure 4. Rietveld Refinement of fitting of XRD peaks pattern using HighScore for (a) Sample A, (b) Sample B, and (c) Sample C.

the pre-alloying process so that the diffusion process is easier to unity [14].

Rietveld refinement of fitting of XRD peaks pattern using HighScore are depicted in Figure 4.

Rietveld refinement of fitting of peaks pattern showed that Fe-Cr phase contained biggest amount of 94.5% in sample of 1-step sintering process at amplitude of 40%, followed of 85.2 % on 50 % and 80.2% on 60% Amplitude. As in Table 2, the Fe elemental amount (in situ) of Fe in sample A is less than in sample B and less than in sample C. This indicate that the sample A has the more fully alloyed Fe-Cr parcile or partially alloyed Fe-Cr particle than Fe elemental or Cr elemental particles[14]. Then. after sintering process, sampel A has the biggest amount of Fe-Cr alloy than others. There is no peak of Y_2O_3 indicated in Figure 4, caused it has amount of less than 5 wt% as limitation of XRD apparatus itself for phase inspection.

The 2-step sintering process was carried outat 1300^{O}C with holding time for 2 hours. The micro structure observed by the SEM-EDS test was depicted in Figure 5 and the elemental composition was shown in Table 4.

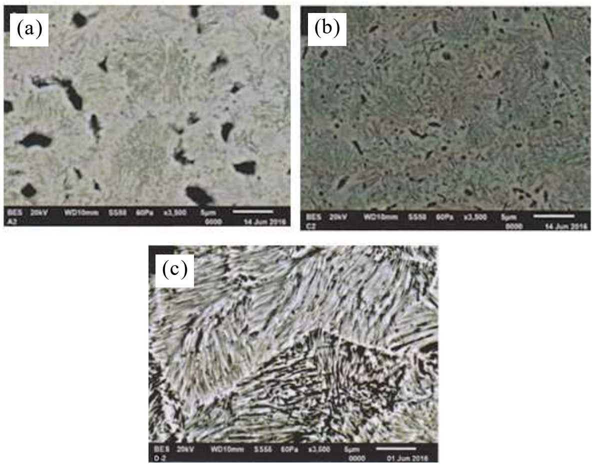

Figure 5. Microstructure of 2-steps sintering Fe-Cr-Y_2O_3 in Amplitude 40, 50, and 60.

Table 4. Elemental composition contained in matrix Fe-Cr-Y_2O_3 phase at 2-steps sintering process.

Sampel	Amplitudo (wt %)	Fe content (wt %)	Cr content (wt %)	Y content (wt %)
A	40	81.31	17.86	0.83
B	50	89.54	10.01	0.45
C	60	83.63	16.02	0.35

The second stage of the sintering process, which is a continuation of the previous sintering process, shows changes in the micro structure with finer grain sizes.SEM-EDS test for sample of ultrasonic treatment with different amplitudeafter 2-stage sintering process showed the Fe-Cr-Y_2O_3 alloy phase, with different structure and composition. Sample of ultrasonic treatment with 40% amplitude showed morphology of ferrite phase with lameral patterns. At amplitude 50 %, the Fe-Cr-Y_2O_3 alloy showed ferritic structure equipped with a flat patternwith grainboundary contrast. The structural phase in amplitude of 60% showed the Fe-Cr-Y_2O_3 alloy structures with contrast and grain boundaries in different flat patterns [24]. The greater sonication Amplitude, the greater cavitation formed[25], then the greter microalloyed particle size.

To evaluate the consolidation behavior, the hardness measurement with Vickers Hardness Number method was done. Plotting of VHN for 1-step and 2-step sintering ultrasonically pre-microalloyed listed in Table 5.

Table 5. The hardness of Fe-Cr-Y_2O_3 for sample after 1 and 2-steps sintering process.

Sampel	Hardness (VHN)	
	1-Step	2-Steps
A	134.41	160.2
B	133.28	127.78
C	133.52	163.77

Table 5 shows that the effect of amplitude is not directly proportional to the hardness of the alloy after sintering process. This phenomenon was predicted due to the random cavitation process during the ultrasonic treatment that affects the microstructure and homogenization of the micro elements. For 1-step sintering process, the highest hardness of 134.51 VHN obtained at 40 % amplitude. The hardness of the alloy depends on the size of the grain boundary associated with the difficulty of the dislocation movement [26]. As shown in Figure 3, the 1-step sintering on ultrasonic pre-microalloyed Fe-Cr-Y_2O_3 at amplitude of 40 % resulted more finer grain than the amplitude of 50% and 60% and therefore this ODS steel most tough to withstand mechanical deformation.

The 2-stage of the sintering process, shows that the hardness value was also consistent with the grain size as observed and explained in the microstructure. As Table 4, the matrix composition containing at most of elemental, therefore has teh lowest hardness. As for 2-steps sintering process, first step sintering of 2- steps sintering so that the densification is active in the final sintering step[12]. Then, the characteristics of alloys are dominated by the matrix (Fe-Cr) alloy. The greatest value obtained by the Fe-Cr-Y_2O_3 sample with 60% amplitude. In this process the diffusion occurs in grain and lattice boundaries and results in significant grain growth which increases the strength of ODS steel alloys [21].

CONCLUSION

1-step sitering process and 2-steps sintering process have been done for ultrasonically varied amplitude microalloyed powders for consolidation. Increasing of the amplitude percentage is not proportional to the value of the hardness and density of the material which shows the consolidation performance of ODS steel alloys. Treatment with an amplitude of 50% showed the highest density value due to the high cavitation formation rate so that the collision in micro particle are the most perfectand dispersed homogeneuosly. The highest hardness occurred in the addition of treatment with an amplitude of 60% which produces a micro structure with the most fine grain. For 1-step sintering process, the highest hardness of 134.51 VHN obtained at 40 % amplitude. The hardness of the alloy depends on the size of the grain boundary associated with the difficulty of the dislocation movement.

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