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Compound Development of $Fe_{80}Cr_{20}$ Metallic Interconnect by Ball Milling and Ultrasonic Technique: Its Effect on Mass Gradation

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ABSTRACT

The investigation of metallic material for interconnect fuel cell application was very challenging due to interconnect operate at high temperature up to 1100^{0}C. However, the main problem of metallic material is mass change at high temperature is inevitable. Therefore, this research aims to analyse the compound of $Fe_{80}Cr_{20}$ and it effect of mass gradation. The methods were conducted by ultrasonic technique (UT) by frequency of 35 kHz, various ultrasonic time of 3, 3.5, 4, 4.5 and 5 h. Milling technique was conducted by milling time of 60 h with nitrogen gas condition. The analysis was conducted by Scanning Electron Microscope for compound investigation and Thermo Gravimetric Analyzer (TGA) for mass gradation analysis. The result shows that there are several compounds that developed after ball milling and ultrasonic technique such as $Fe_{80}Cr_{20}$, FeO_2 and Cr_2O_3. The UT samples shows the lowest mass gain of 31.917 mg is located at UT 4.5 h, while raw material has largest mass gain as compared to the other samples (treated samples). Meanwhile for milled and combination of milled and UT samples shows the lowest mass gain has observed at milled and UT 4.5 h with the value of 17.014 mg and 12.7 mg, respectively.

Keywords: Interconnect; Fuel Cell; Mass Change; Compound; Metallic Material

INTRODUCTION

Solid Oxide Fuel Cell (SOFC) is a promising technology for electricity generation that used to convert the chemical energy of the fuel gas to electrical energy directly (Zhang 2006; Dafit Feriyanto and Supaat Zakaria 2020). The system can reach an efficiency approximately 80% where efficiency of internal combustion engine is no more 40% when coupled with heat recovering system for cogeneration. Besides, SOFC can replace several fuels and widely implemented to power supplies from small scale distribution power supplies to large scale thermal power generation (Choudhury and Sarma 2011). The structure of SOFC is well stacked and it consists of interconnect, anode, electrolyte, cathode, and materials as shown in Figure 1 (Quadakkers 2003).

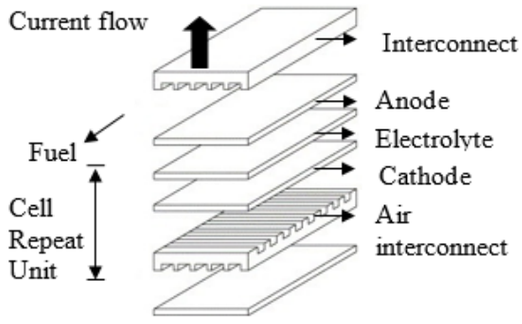

FIGURE 1. Configuration for planar design SOFC (Singhal 2000 and Quadakkers 2003)

The cell interconnection is exposed both to cathode and anode, so it must be stable chemically in both environments at temperature of 1000^{0}C. However, since the operating temperature of this material is of high range, the material should have fire resistance properties, oxidant gases, and proper coefficient of electronic conductivity (Zhang 2006; Gorte et al. 2003 and Zhong et al. 2004). These three criteria are most important for interconnect material such as high electrical conductivity, good stability in both condition reducing, oxidizing and matching for coefficient of thermal expansion (CTE) with other SOFC components (Benjamin 2004). It may be achieved by several methods that treated in interconnect material such as mechanical alloying and ultrasonic process. Those technique to compound development, reducing crystallite size and fine surface structure that promote to obtain high thermal stability.

The actual mechanical alloying process started by mixing powder with the right proportion and filling the grinding jar with powder mix and steel balls. The powder is milled with the desired milling time until a steady state condition when the composition of every powder particle was same as the proportion of the elements in starting powder mix. These processes produce nanocrystalline structures in pure metals, intermetallic compounds, and alloys systems. It was shown that nanometer size grains have been acquired. This process decreases the grain size with the increasing milling time which is inversely with melting temperature (Murty et al. 1998). It is further shown that, the most dynamic equilibrium is achieved in mechano-syntesized for more than 24 hours (Fnidiki et al. 2005). Many researchers have explored this technique in order to improve properties such as according to Redjdal et al. (2013), the FeCr alloy is used as raw material. It is treated by using mechanical milling with milling time of 4 hours and they obtained lower porosity shown by treated sample as compared to untreated sample.

Ultrasonic bath with high vibration has potential to refine the surface morphology of intermetallic compounds, breaking agglomerates and aggregates of $Fe_{80}Cr_{20}$ alloy powder (Puga et al. 2011). Ultrasonic vibration could be enhanced the other properties of FeCr alloy powder such as corrosion resistance, low oxidation kinetic, and electrochemical performance (Qingmei et al. 2007).

The iron-chromium system has been used for a long time as the basis of researches for high-strength, corrosion-resistant applications (Deni et al. 2011 and Redjdal et al. 2013). Chromium is the α-phase stabilizer when added to the iron powder since chromium has the similar crystal structure of body centre cubic (BCC). The Fe-Cr phase diagram is shown in Figure 2 (ASM Handbook 1992). At low temperatures, chromium and iron do not form a complete solid solution due to the presence of the sigma phase (σ-phase) which has a tetragonal structure and generally it is hard and brittle. The equilibrium phase diagram predicts the formation of σ-phase for alloys containing greater than 15 wt% Cr at temperatures between 475^{0}C and 821^{0}C. However, heat-treatments at high temperatures and long holding time or slow cooling rates are required for the formation of this phase. For a Fe 27 wt% Cr alloy, it was shown that σ-phase would precipitate out of the σ-phase after holding at 565^{0}C for 131 days as shown in Figure 2 (Smith 1993). A Fe-20 wt% Cr alloy, held in temperatures of 600^{0}C and above as shown in Figure 2.8, it would be within the α-phase field and would never form σ-phase after extended the high temperature exposure (Benjamin 2004).

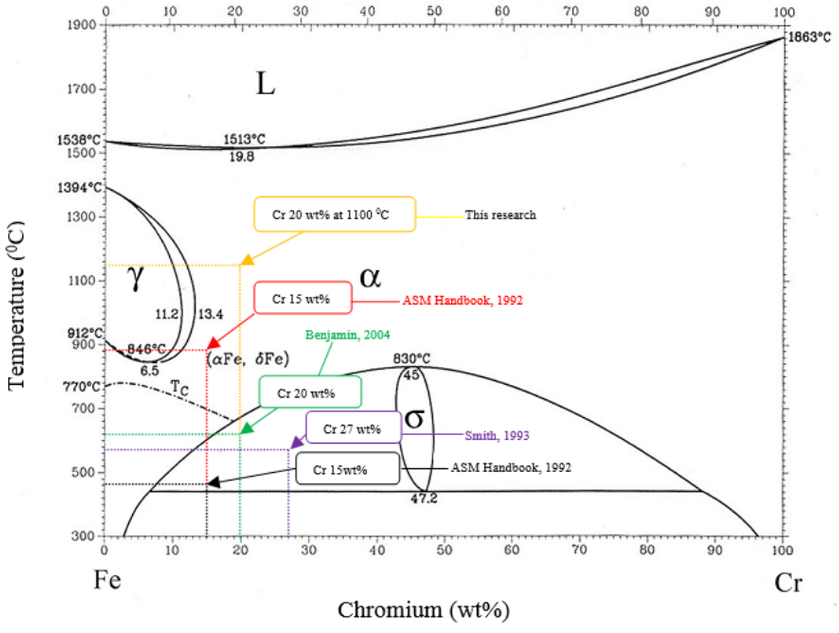

FIGURE 2. Phase diagram of Fe-Cr system (ASM Handbook 1992)

From Figure 2, it can be seen that this research study used a composition of 80 wt% Fe and 20 wt% Cr in 1100^{0}C temperature operation for thermal analysis. That area was designed because the high ductility and strength -phase will be obtained (Dafit Feriyanto et al. 2020 and 2021). Traditionally, Fe based alloy with chromium addition have been used in room and medium temperature (500^{0}C). The chromium addition on the alloy promotes the formation of a dense, adherent layer of Cr_2O_3 on the surface. The formation of a dense chromium layer is beneficial for continuing the interconnect oxidation (Jaing et al. 2002).

METHODOLOGY

$Fe_{80}Cr_{20}$ alloy powder was developed by using high energy ball milling. The preparation stages is started by the ratio of the ball (hardened stainless steel ball) and the powder mass adjusted at 13:1. The samples is inserted to jar and conditioned by the glove box machine where the pressure tube for glove box was set at 2000 psi with 20 psi of nitrogen pressure. Milling time was conducted for 60 h with the rotation speed of the planetary ball mill was 300 rpm 2 rpm, 30 minutes cycle time, 10 minutes pause time. After high energy ball milling process, an ultrasonic bath technique was conducted with frequency of 35kHz and various holding time of 3, 3.5, 4, 4.5 and 5 h. The mechanical alloying using high energy ball milling and ultrasonic are done separately. Therefore, the new method in this research is combination technique of ball milling and ultrasonic treatment in order to obtain the better properties as compared to the properties when produced by ball milling and ultrasonic technique separately.

There are two analysis that performed in this research which are compound analysis and thermal stability analysis. Compound analysis achieved by SEM/EDS that use 1000 times magnification for raw material and Ultrasonic Technique (UT) samples while 2000 times for Milled 60 h sample and combination technique samples. Thermal analysis of $Fe_{80}Cr_{20}$ alloy was performed by using Thermo Gravimetric Analysis (TGA). The samples were measured by using microbalance with accuracy of 0.1 mg then inserted into a TGA machine using temperature of 1100 ^{0}C, heating rate and cooling rate of 10 ^{0}C per minute.

RESULTS AND DISCUSSION

TGA ANALYSIS

TGA phenomena may cause physical mass changes such as gas absorption, gas desorption phase transition and for chemical such as decomposition, break down reaction, gas reaction, and chemisorptions (Darwin et al. 2010). Subsequently, to obtain the -phases which have high ductile and strength properties it needs 20 wt% of Cr from the FeCr alloys. That composition is promoted in order to not obtain the -phases which have hard and brittle properties. The comparison data is shown in Figure 3, the sample oxidized is in temperature of 1100^{0}C. The temperature of 1100^{0}C was selected because to know the thermal stability of the ferritic steel ($Fe_{80}Cr_{20}$) as interconnect material at high temperature which is usually applied to ceramic materials.

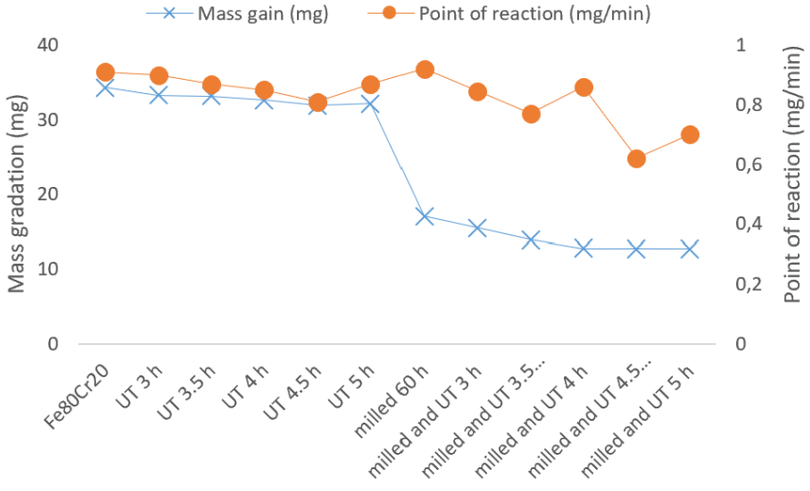

FIGURE 3. TGA data of the treated and untreated samples

Crystallite size of the sample designation has high influence on the mass gain. Even though in higher temperature operation, milled 60 h sample and combination technique sample have smaller mass gain than raw material and UT samples. This is due to the reducing crystallite size after each treatment. Therefore, crystallite size is decreased with the mass gain decreased as well. For the point of reaction and mass change derivative, it is relatively decreased by decreasing the crystallite size. This can be founded at milled and UT 4.5 h with the values of 0.62 mg/min and 0.0855 mg/^{0}C, respectively.

It can be concluded that the crystallite size decreased as mass gain decreased. It is supported with the previous research that growth rate of crystallite size after consolidation process by using hot press (Deni 2011). They found that the growth rate of crystallite size from 5.82 nm to 38.51 nm after consolidation process located at $Fe_{80}Cr_{20}$ with milling time of 60 h (growth rate of 85%) and also growth rate of $Fe_{80}Cr_{20}$ with a milling time of 40 h from 6.38 nm to 53.32 nm (growth rate of 88%) (Hendi 2010).

EDS ANALYSIS

The EDS analysis was conducted on all samples to measure the value of iron (Fe) and chromium (Cr) in individual particles. The EDS analysis of raw material and represent each parameter of ultrasonic treatment samples, ball milling sample and combination technique samples. Figure 4 to 7 show the spectrum of the chemical composition of treated and untreated samples which were produced from EDS analysis of the raw material ($Fe_{80}Cr_{20}$), UT 4.5 h, milled 60 h, and combination technique sample (milled and UT 4.5) sample, respectively. The most appropriate of concentration of the iron and chromium were observed of 79.95 wt% and 20.05 wt% respectively in particle surface region which located at milled and UT 4.5 h sample.

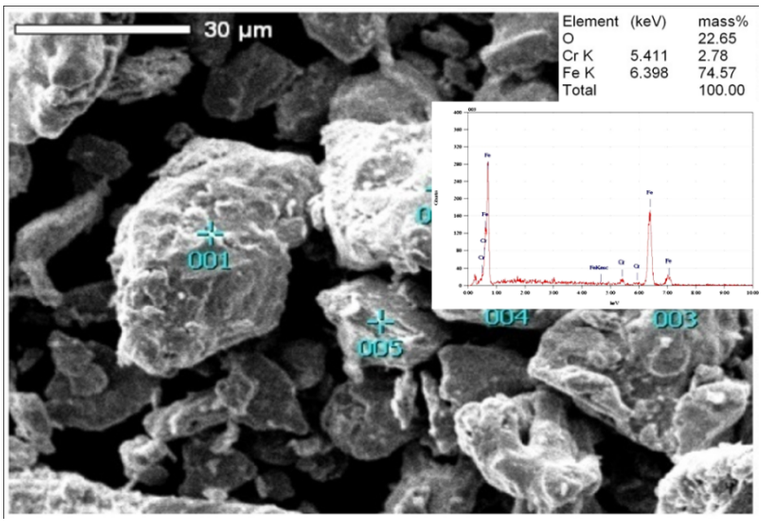

FIGURE 4. EDS result of $Fe_{80}Cr_{20}$ alloy powder

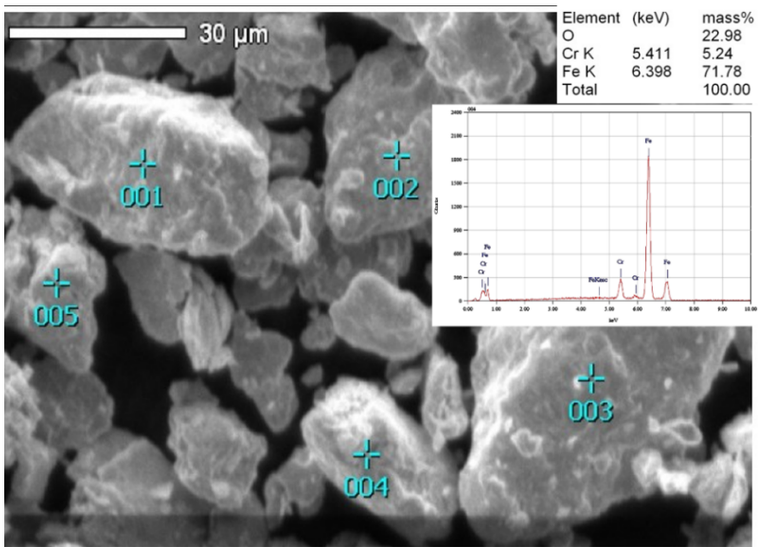

FIGURE 5. EDS result of UT 4.5 h

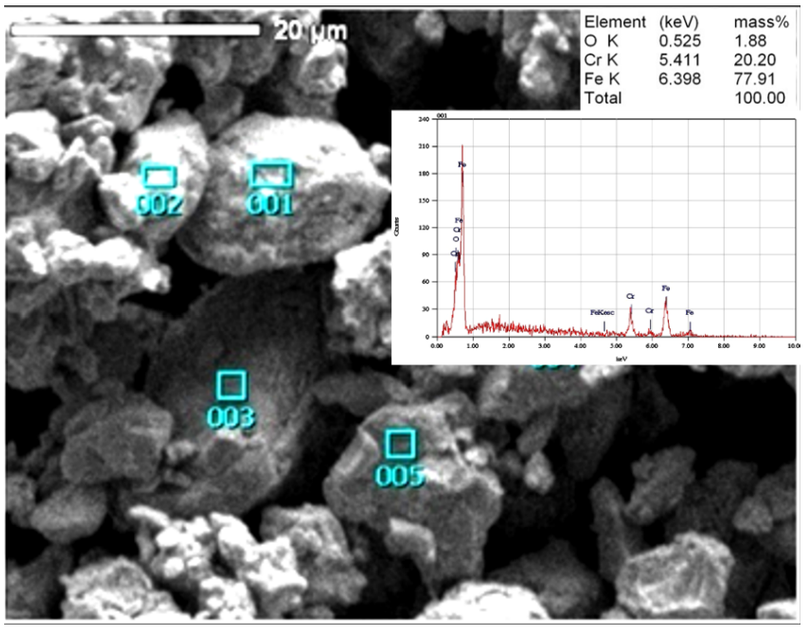

FIGURE 6. EDS result of Milled 60 h

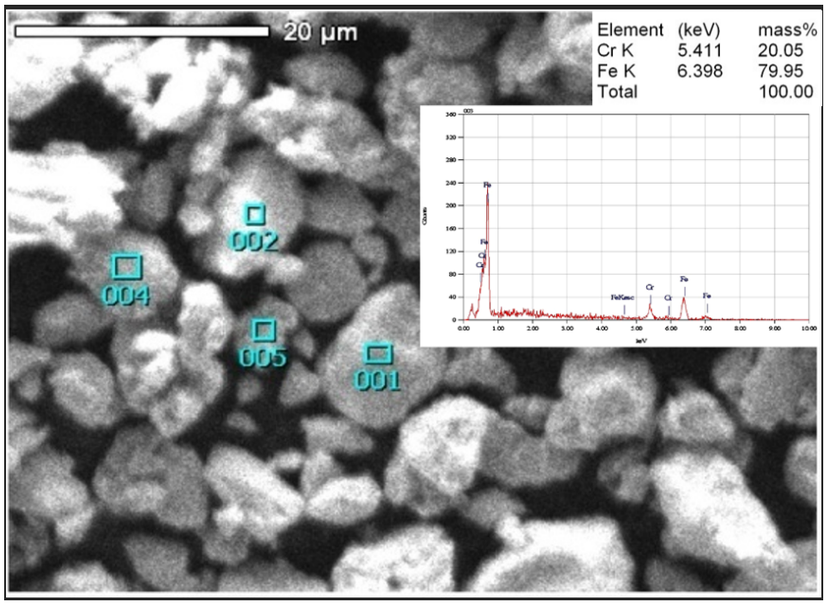

FIGURE 7. EDS result of milled and UT 4.5 h

Figure 4. the EDS analysis of raw material treated by manual mixture. Therefore, the value of the chemical composition of Fe and Cr in individual powder is not appropriate for the work composition (80 wt% Fe and 20 wt% Cr). High concentration of the oxygen is caused by many voids on the pure Fe powder. Figure 5 shows the improving homogenous and surface morphology samples by using ultrasonic treatment with mainly observed of Fe, Cr and O. Its composition value is 71.78 wt% Fe, 5.24 wt% Cr and 22.98 wt% O. The concentration of the oxygen on this samples can be reduced using ultrasonic treatment, the collision is occurred on each particle. Therefore, some voids can be reduced. The concentration of the Fe and Cr powder in individual particle is not appropriate because the ultrasonic treatment is focused to break agglomerates and improve the homogeneity. Figure 6 shows that the material was mixed by using ball milling machine, the iron is 77.91 wt% and chromium is 20.20 wt% and oxygen is 1.88 wt%. Concentration of oxygen is significantly reduced as compared to UT sample due to during ball milling process, the ball slugging the powders which lead to develop the alloy powder with minimum oxygen cavities. Figure 7 introduce new method to refine the surface morphology, break the agglomerates and reduce the powder size with the composition for iron powder of 79.95 wt% and chromium of 20.05 wt%. New method of developing alloy powder has proved effective by using this combination technique. Appropriate concentration of the Fe and Cr which indicated that the -phase of $Fe_{80}Cr_{20}$ alloy has achieved. The -phase is needed because of the high corrosion resistance, high ductility and high strength of -phase of $Fe_{80}Cr_{20}$ alloy (Sebayang et al. 2013 and Dafit et al. 2014). From the Figure 4.18 indicated that the material was confirmed as work composition which produced $Fe_{80}Cr_{20}$ alloy powders and the composition relatively closer.

CONCLUSION

Based on EDS analysis, the $Fe_{80}Cr_{20}$ alloy has been observed in individual particle when treated using milled by 60 h and combination technique (new method). In milled 60 h the EDS result is 1.88 wt% O, 20.20 wt% Cr and 77.91 wt% Fe while in combination technique samples is 20.05 wt% Cr and 79.95 wt% Fe. New method of developing nanocrystalline through the milling combined with ultrasonic treatment was explored. It can reduce the crystallite size up to 98 % when compared to the raw material. Increasing the thermal stability indicated by decreases mass gain up to 63%, 62% and 25% as compared to raw material, UT samples and milled 60 h sample, respectively.

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DECLARATION OF COMPETING INTEREST

None

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