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Processing of Boron Nitride Nanotubes Reinforced Aluminum Matrix Composite

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Abstract

Aluminum and its alloys are one of the most favored metal-based materials for engineering applications that require lightweight materials. On the other hand, composites are getting more preferable for different kinds of applications recently. Boron nitride nanotubes (BNNTs) are one of the excellent reinforcement materials for aluminum and its alloys. To enhance mechanical properties of aluminum, BNNTs can be added with different processes. BNNT reinforced aluminum matrix composites also demonstrate extraordinary radiation shielding properties. This study consists of BNNT reinforced aluminum matrix composite production performed by casting method. Since wetting of BNNT in liquid aluminum is an obstacle for casting, various casting techniques were performed to distribute homogeneously in liquid aluminum. Different methods were investigated in an aim to incorporate BNNT into liquid method as reinforcement. It was found that UTS was increased by 20% and elongation at fracture was increased by 170% when BNNT was preheated at 800°C for 30 minutes.

Keywords: Aluminum, Casting, BNNT, Composite, Tensile

1. Introduction

Aluminum is one of the important engineering metals thanks to its improved properties. Since aluminum has lower density compared to other metals that are suitable for industry, it is mostly used in lightweight applications. The “composite” term requires one reinforcement material in a continuous phase called “matrix”. Aluminum matrix composite (AMC) is material which is based on aluminum matrix exhibiting enhanced mechanical properties. Metal matrix composites such as AMC contain at least one metal and one reinforcement material such as oxide, fiber, particle, or compound. Since the aluminum used as the main structure in AMC materials, they exhibit high specific strength, high stiffness and good wear resistance. These are the important features of AMC structures which makes them suitable for advanced aviation and space applications. Recent researches show that AMCs which are reinforced with Al_2O_3 and SiC are the most common applications. All of these ceramics which are used as reinforcement improve different features of material. In addition to these reinforcements, B4C is also used to obtain different enhanced properties for AMCs [1]. Ceramics such as SiC and Al_2O_3 have higher hardness than aluminum however because of the higher density of these ceramic materials, they have higher weight than aluminum. This difference brings important restrictions for industrial applications. Since AMC materials have ceramic reinforcement in aluminum structure, they exhibit high strength values while having lightweight properties. So, AMC materials have combinations of advanced properties of both ceramics and aluminum [1].

There are different properties which are improved by reinforcement of aluminum with materials such as ceramic, compound or oxide. AMC structures exhibit high strength, high hardness and high stiffness. Also, these composites show better performance even at elevated temperatures. In addition to well-known improved properties, reinforcement of AMCs with ceramic particles provides advanced coefficient of thermal expansion and wear resistance [2]. Also, AMC structures exhibit good electrical and thermal conductivity which provides enhanced properties at elevated temperatures [3].

Production techniques that are used in composites have a great effect for determining the final properties of composites. For metal matrix composites (MMCs), there are several techniques providing high production quality. However, manufacturing of aluminum matrix composites in large-scale is possible with 3 main different techniques:

- Solid State Processes
- Liquid State Processes
- Deposition (Liquid - Solid) Processes

Distribution of reinforcements in composites should be homogeneous to obtain desired properties. Therefore, obtaining the most suitable particle size and shape is crucial for applying proper manufacturing techniques. Inhomogeneities of reinforcements in composites is one of the most challenging topics for metal matrix composites [4].

Boron nitride nanotubes (BNNTs) are the materials that promise to be an alternative to carbon nanotubes (CNTs) in many areas, especially in space technology. The main reason that allows BNNT to be used as an alternative to CNT is that the BNNT structure consisting of a co-axial hexagonal boron nitride (*h*-BN) network [5] is analogous to the CNT structure. Other similar properties of these two materials are due to the peculiar properties, structure and polymorphism of BNNT [6]. In spite of the limited studies on BNNTs compared with CNTs due to the challenging production of a high amount of BNNTs, in recent years, the interest in theoretical experiments of BNNT is increasing.

The excellent mechanical property of BNNTs, which is a high stiffness with Young's Modulus of 1.2 TPa, and their density of 1.3-1.4 (g/cm^3) [7] make them proper lightweight reinforcements for metallic, polymeric and ceramic composites [5]. According to Bettinger et al. [8], even though the modulus of elasticity of single layered BNNT is slightly lower than that of CNT, BNNT stands out due to its yield resistance and thermal stability [8]. Another property that makes BNNT surpass CNT is its high oxidation resistance. CNT can be easily oxidized in air at 400 °C [9] and burn completely under an effective oxygen source at 700 °C due to low oxidation resistance caused by its large surface area and defects formed at the tip of tubes [10]. Chen et al. [11] reported that BNNTs, on the other hand, remain stable up to 700 °C in any structure, while they resist up to 900 °C in a cylindrical structure. In another experimental study, it has been observed that BNNT remains stable up to 850 °C without any deterioration in its morphology. Besides all these, the most extraordinary properties of BNNT, which also distinguishes it from CNT, comes from its ionic bonding nature. The dipole moment between H_2 and nanotube structure induced by ionic B-N bonds, provides a unique hydrogen storage capacity [5]. This property is quite important in radiation shielding applications since the charge-to-mass ratio of hydrogen, which is the highest of any elements, offers the best shielding feature [7]. Neutron radiation is the main type of radiation that is dangerous for spacecraft. Elements of B and N are extremely suitable for shielding from neutron radiation due to their larger neutron absorption cross-sections than that of C and other elements [7]. Moreover, due to their ionic bonding nature, BNNTs are wide band gap materials having a bandgap of 5.5 eV [12-13] which makes them electrical insulators, and the electrical properties of BNNT are not dependent on their diameter of tube, chirality and morphology as they are for CNTs [12].

In the production of aluminum matrix CNT composite, the reaction occurs between CNT and aluminum at the interfaces and forming the Al_4C_3 (aluminum carbides) structure has led to very low mechanical properties [14]. As a result of the research made to solve this serious problem, the issue of using BNNTs, which are claimed to be used as reinforcement in composite structures due to their properties, has come to the fore.

One of the most important properties determining the strength of the bonding formed at the interface is the orientation relationship between Al and the reaction product. AlN provides a stable and strong interface bond by forming a low-energy and coherent surface with Al, allowing BNNT to bond to the matrix. This relationship is the opposite of Al and AlB2. Since the planes harmony between these two is much less, a partial coherent structure is provided. For this reason, AlB2 provides a much lower bonding strength than AlN [15]. In BNNT reinforced Al composites, thanks to the diffusion event mentioned above, more coherent AlN formation causes a stronger interface bonding. The interface between Al and BNNT is schematically given in Figure 1.

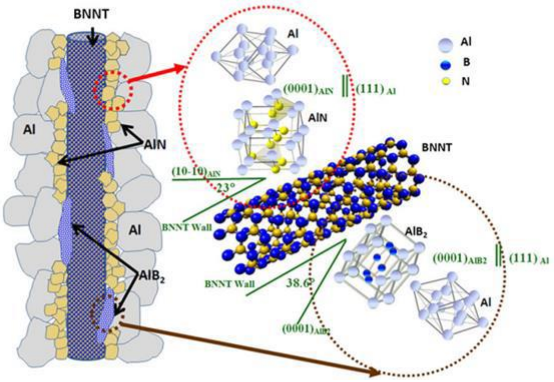

Fig. 1. Schematic representation of Al-BNNT interface [16]

In the light of all the above information, as can be seen in the figure, the AlN coherent structure, which is mostly formed at the interface, provides a strong interface bond. In the light of the researches and comments made, it was determined that BNNT can be used as reinforcement in Al metal matrix.

Cong et al. [17] investigated the effect of BNNT volume fraction and size on mechanical behavior in his studies. In this study, he simulated the data to be obtained when applying tensile stress to the composite using Molecular Dynamics simulation. As a result of his studies at standard ambient conditions and at 0.0005 /ps strain rate, the samples with BNNT added in different size and volume fractions were exposed to tensile loading. He obtained different stress-strain curves for each sample. But the interesting point is that in all of these samples, 2 different peaks were observed in certain strain values. As explained in the schematized visual in Figure 2.15, it has been observed that the Al matrix at a certain strain value breaks under tensile loading but BNNT does not rupture and provides a secondary strength to the composite. The second stress reduction occurs when the BNNT reinforcement is also broken.

Many studies have been conducted in which the production of BNNT reinforced Al composite was tested with spark plasma sintering and the resulting mechanical properties and strengthening mechanisms were examined. One of the most enlightening researches on the subject has been done by Lahiri [16]. Within the scope of this study, powders containing 2% and 5% BNNT as volume fraction and pure Al powder were prepared. According to the values given, a 54% increase in Yield strength was observed in the structure with the addition of 5% BNNT compared to pure Al. In addition, the BNNT bridge structure and the "sword in sheathe" structure observed on the fracture surface in the sample with 5% BNNT addition clearly show the strengthening mechanism in composites with BNNT reinforcement Al matrix [16]. While an increase of 21% is observed in the hardness of pure Al due to strain hardening, this increase is observed as 59% in the composite structure [16].

In this study, the production of boron nitride nanotube reinforced aluminum matrix composites by casting method was tried. Different methods were investigated, and mechanical properties were evaluated.

2. Experimental work

Aluminum A356 alloy was used as the matrix material in the composite production. The powder used as reinforcement contained 52 wt% boron nitride with 99.8% purity and 48 wt% CNT with 97% purity. The ratio of the reinforcer was targeted to be 0.1 wt% in all casting trials.

In order to characterize the effect of BNNT reinforced A356 alloy, a series of castings were made into a sand mould that produced 10 cylindrical bars. The bars were 8.5 mm diameter with 160 mm height. The melt was prepared in ICS Induction Furnace A50 SiC crucible at 750ºC. Degassing was carried out with Pyrotek ceramic lance with nitrogen gas purges for 12 minutes.

After the castings were complete, all samples were subjected to X-ray for porosity analysis using YXLON MU-2000. Then, T6 heat treatment was applied to all samples: 6 hours of solution heat treatment at 540 °C, quenching at 80 °C and 4 hours of artificial aging at 160 °C. The tensile test samples were machined to ASTM E8 standard and tests were carried out on ZWICK 250 kN Tensile Testing Machine. Reduced pressure test (RPT) samples were collected to ensure that the melt cleanliness was high enough. Bifilm index [18–20] measurements were made for each casting.

Fig. 2. Sand moulds for producing 10 tensile test bars

For the casting trials, different ways of introducing BNNT into the liquid aluminum was investigated. A summary of test setup is given in Table 1. In the first casting sets (Method 1), base A356 was cast with no reinforcement addition as a reference. In Method 2, powders were placed in the pouring basin in an aim to be carried out by the flowing liquid metal. In Method 3, powders were placed in the runner. In Method 4, powders were added to the liquid metal during pouring towards the down sprue with a vibration plate. In Method 5, powders were added to the liquid metal in the crucible. In Method 6, the powders were added through the degassing shaft together with carrier gas (nitrogen) during degassing. In Method 5 and 6, BNNT was preheated to 800ºC for 30 minutes and then they were added to melt.

Table 1.
Methods used for casting trials

Method	Condition
(1)	Base alloy – A356, no addition of BNNT
(2)	BNNT was placed at the pouring basin
(3)	BNNT was placed at the runner
(4)	BNNT was added to the sprue by vibrating plate during filling
(5)	BNNT was preheated at 800ºC for 30 minutes and added to melt in crucible prior to casting
(6)	BNNT was preheated at 800ºC for 30 minutes and added to melt through degassing lance during degassing operation

3. Results and Discussion

The melt quality was measured by using reduced pressure test. Cross-section of RPT samples are given in Fig 3.

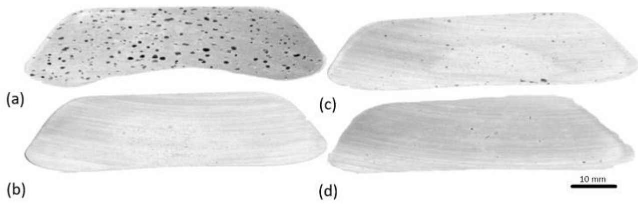

Fig. 3. Cross section of RPT samples

Before degassing, the bifilm index was measured as 188.3 mm (Fig 3a). After degassing, the bifilm index was measured to be 12.5, 47.1 and 27.9 mm (Fig 3 b, c and d, respectively) which lies in the category of good melt quality [18-20]. Therefore, it was determined that the tensile test results were not affected by the melt cleanliness level. Additionally, YXLON MU-2000 X-ray machine was used to check the porosity levels in the cast cylinders. As seen in Fig 4, no porosity was observed in the cast bars.

Fig. 4. X-ray images of tensile test bars

For the Methods 5 and 6 (Table 1), the powders were heated to 800°C and held for 30 minutes in an aim to remove CNT and also increase the wettability of powders. XRD analysis was carried out and the result is given in Fig 5. The change from wide to narrow and sharp peaks in XRD diagram indicated that CNT was removed from BNNT. It is important to note that the color of powders was changed from grey to white after 30 minutes of holding at 800°C.

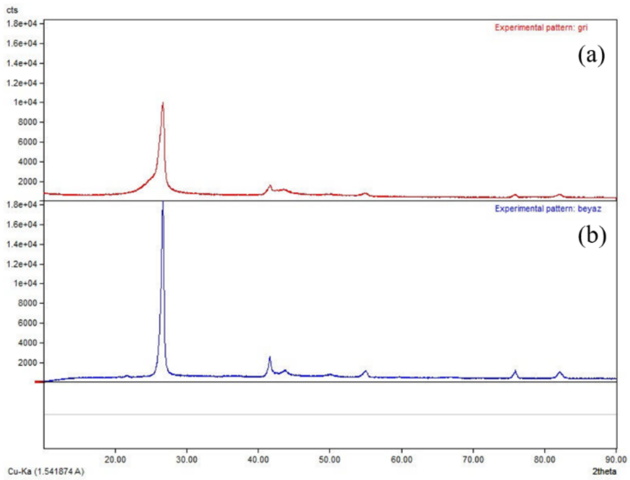

Fig. 5. XRD analysis of a) untreated powders with a composition of 52%BN and 48% CNT, b) Heat exposed powders

Tensile test results are summarised in Figs 6-9 as bar charts for different methods studied in this work. The yield strength appears to be close to each other (Fig 6) with Method 6 having the lowest value of 205.8 MPa and Method 4 giving the highest as 240.6 MPa in which the BNNT was added with vibration towards the sprue during filling. Compare to the base alloy with no reinforcement, the increase in yield strength is approximately 5% for most of the methods.

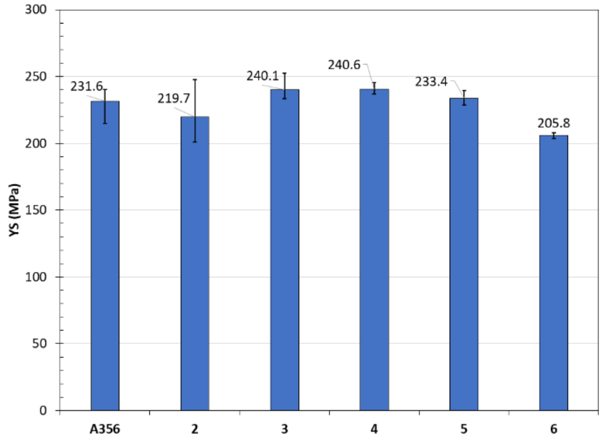

Fig. 6. Yield strength change of composites with regard to method they were produced

In the tensile test, it was found that the average yield strength of base A356 alloy was 231.6 MPa. The yield strength of the BNNT reinforced castings were found to be very close to the base alloy. There was only a 3.8% increase in the yield strength that corresponded to 9 MPa of increase overall.

Among the methods investigated in this work, the standard deviation of yield strength values was quite low in Methods 4,5 and 6. It can be said that in these methods, homogeneous mixing and distribution of the powder was achieved. Similar observation was made for UTS values as well.

In the computational research made by Rohmann et al. [21], it has been reported that the tensile strength value of BNNT reinforced aluminum matrix composite produced by powder metallurgy can only reach 300 MPa with the addition of 3 to 5 wt% powder [17]. Ultimate tensile test results (Fig 7) are varying between 250-350 MPa. Method 5 reveals the highest UTS value of 334.9 MPa while Method 3 has the highest scatter. The increase of UTS value compare to base alloy was around 16% in Method 5, followed by 14.3% increase in Method 4 with a value of 329.8 MPa. It is concluded that higher UTS can be achieved by casting method compared to powder metallurgy with even less amount of reinforcer addition.

For the elongation at fracture values (Fig 8), there is a significant difference between the different methods. While Methods 1-3 are in the range of 2% elongation, Methods 4-6 are showing an average of approximately 5% elongation at fracture. The casting method that provides the highest increase of 175% in elongation is Method 5 where the pre-heated powder was added to the crucible and mixed with molten alloy prior to casting.

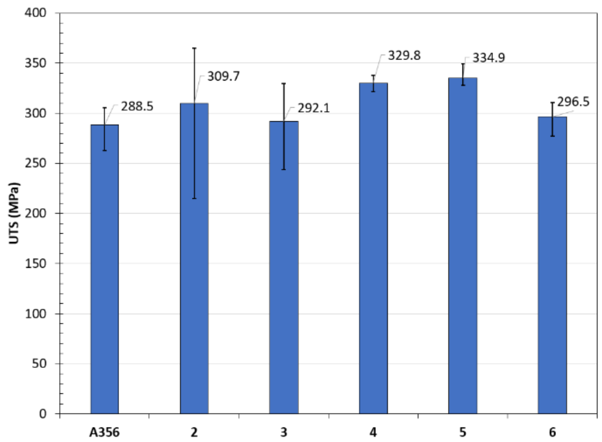

Fig. 7. Ultimate tensile strength change of composites with regard to method they were produced

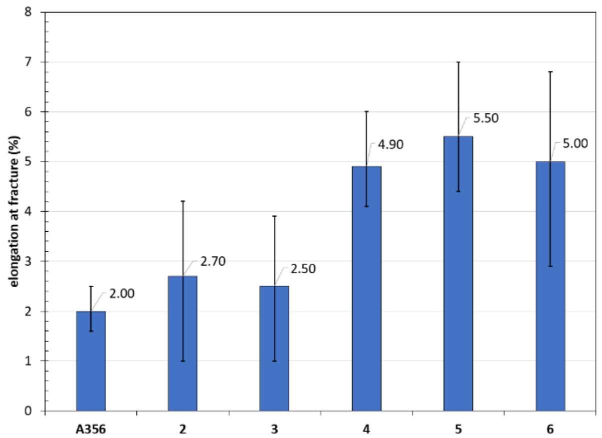

Fig. 8. Elongation at fracture change of composites with regard to method they were produced

A similar finding was observed for the toughness values (Fig 9). Methods 4-6 reveal a significant increase in toughness. The highest toughness was achieved with Method 5.

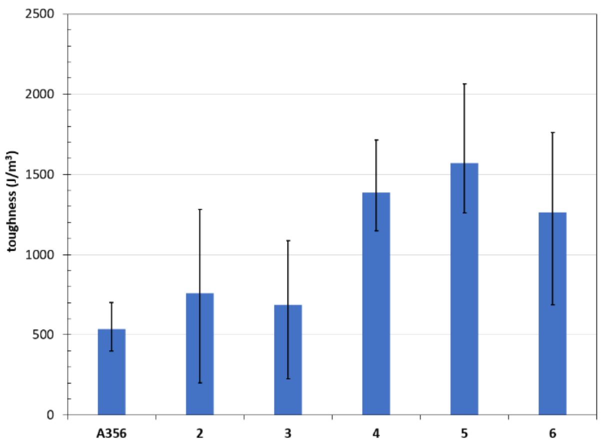

Fig. 9. Toughness change of composites with regard to method they were produced

From the data collected from tensile tests, statistical analysis was carried out. Weibull analysis was used, and survivability plots were drawn. The results can be seen in Figs 10-12. For the interpretation of survivability plots, there are two important points. First one is the location of the data. As the line is placed towards the right-hand side of the plot, it indicates that the representative values of the data are high. The second point is the steepness of the slope where the higher the steepness, more reliable and reproducible the result would be. From this perspective, according to Fig 10, Method 2 has the least producible results showing high scatter for yield stress. Although Method 6 reveals the highest reproducible results, Methods 3 and 4 have the highest yield strength value.

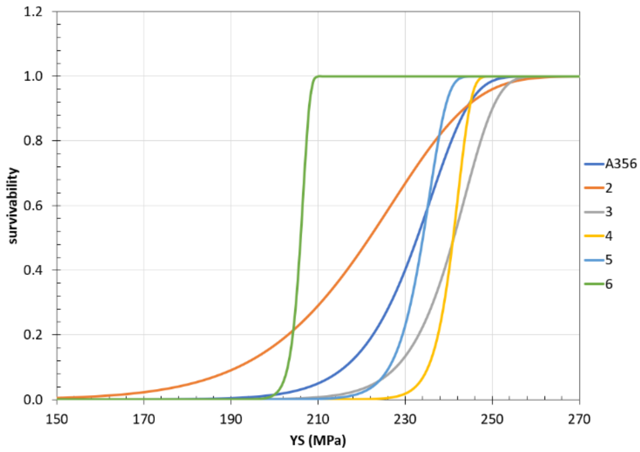

Fig. 10. Survivability plot of yield stress

Based on Figure 11, Methods 2 and 4 show the least reliable UTS values giving high scatter. For example, Method 2 has the potential to give 485 MPa, but also, it has the potential to have 135 MPa as well. The upper and lower limit values show how reproducible the parameter is. Thus, Method 5 being on the far right with the steepest line reveals the highest UTS with most reliable values ranging only between 290-330 MPa with ±20 MPa.

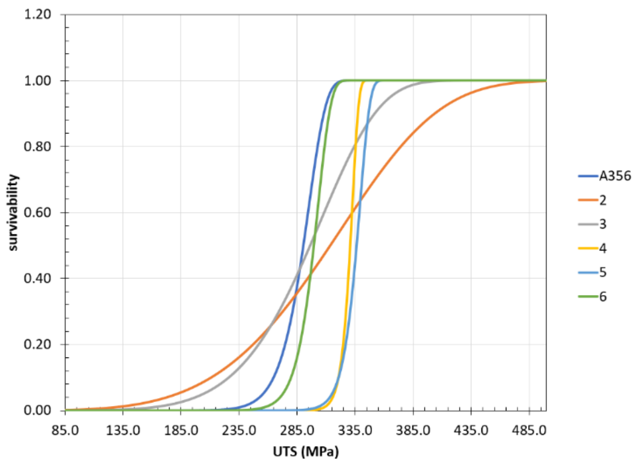

Fig. 11. Survivability plot of UTS

For the survivability plot of elongation at fracture values (Fig 12), base alloy without any reinforcement has the most reliable results with an average of 2% and a scatter of ±0.5. On the other hand, Methods 4 and 5 have the highest elongation at fracture with a scatter between 2-8 % potential value.

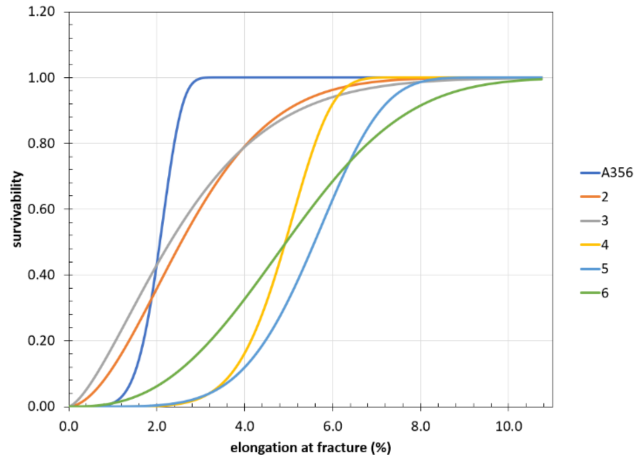

Fig. 12. Survivability plot of elongation at fracture

6. Conclusions

The main objective of this study was to observe the effects of BNNT reinforcement in aluminum matrix and produce BNNT reinforced aluminum matrix composite with casting method. Different methods were applied. Amongst the trials, the highest mechanical properties were achieved when BNNT was heated to 800°C and held for 30 minutes. UTS was increased approximately 20% from 288.5 MPa in base alloy to 334.9 MPa in the composite. Elongation at fracture was increased by 175% compare to the base alloy from 2% to 5.50% in the composite alloy. Similarly, toughness was increased from 500 J/m^3 to 1500 J/m^3 by three folds. Weibull analysis showed that the scatter of the results was too low, thus the reproducibility and reliability of the castings were found to be high with survivability plots showing the same trend as the base alloys. Addition of BNNT through vibrating table into the sprue, mixing the BNNT in the crucible and purging BNNT through the degassing lance revealed the highest tensile properties.

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