

Combustion synthesis of NiAl/TiC composite from mechanically activated elemental powders

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Research Article

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Abstract

The aim of this study was the synthesis of a homogenous distributed NiAl/TiC composite by combustion synthesis of elemental powders. The effect of the mechanical activation process was evaluated. The phase characterization and evaluation of the microstructure during milling and combustion synthesis were carried on by XRD and SEM-EDS and TEM-EDS, respectively. The XRD result of as-mixed powder compact after combustion synthesis showed that in addition to $\text{TiC}_{0.75}$ and NiAl, some undesired phases like Ni_3Al and Ni_2Al_3 formed. While after combustion synthesis of 3h mechanically activated powder compact, only AlNi and TiC formed. The microstructural evaluations by SEM-EDS and TEM-EDS analysis confirmed the synthesis of truly homogenized NiAl/TiC composite. The mechanism of NiAl/TiC formation consists of dissolution of nickel, titanium, and graphite in molten aluminum and precipitation of TiC primarily and then the formation of NiAl. The value of microhardness of synthesized NiAl/TiC was 1070 ± 12 HV. The main reason for this result is a uniform distribution of spherical TiC reinforcement in the NiAl matrix. Therefore, the mechanical activation process facilitated the reactions and improved the mechanical properties of the final composite.

1. Introduction

Nickel aluminides, especially NiAl, have been attracted many interests in industrial applications due to their wonderful properties such as wide composition range, high melting point, high oxidation, and hot corrosion resistance at high temperatures and low density [1–5]. Also, NiAl has good tribological properties [6, 7]; however, sole NiAl materials contain low toughness and poor ductility at ambient temperature and poor strength and creep resistance at high temperatures which restricts its high-temperature applications [8, 9]. Based on previous studies, the reduction of crystallite and particle sizes of NiAl [4, 10–12] and fabrication of In-situ composite [13, 14] can improve the mechanical properties of NiAl. Many efforts have been devoted to developing a nano-crystalline reinforced nickel aluminide which one of them is reinforcing NiAl by a reinforcement(s) (as second phases) like ceramic materials. For instance, NiAl-WC [8], NiAl-WC- Al_2O_3 [13], NiAl-TiC [9, 14, 15], NiAl- TiB_2 [16], NiAl-ZrC [17], NiAl- Al_2O_3 [18], NiAl-TiN/ TiB_2 [19] composites have been studied. Because of low density, high hardness, high modulus of elasticity and high-temperature stability, titanium carbide (TiC) is a promising candidate as reinforcement [20–22].

Energy and time saving are the main properties of combustion synthesis which makes it become a considerable method to synthesize In-situ composite [1, 23–26]. The effect of many different parameters like heating rate, green density, etc. on the microstructure and properties of fabricated composite have been evaluated [27–29]. According to the author's knowledge, there is no study about the effect of mechanical activation on the synthesis of NiAl/TiC composite from elemental powders.

The mechanical activation is a short duration milling before the combustion synthesis [20, 24, 30]. Mechanical activation by the ball milling process is the simplest method for elemental and material dispersion. In this process, the structure and properties of powders modify and not only it decreases the

crystallite and particle sizes of powders, but also increases the surface area of powders, forms structural defects and porosity. Also, this process destroys layer oxide of the reactant powders such as aluminum powder. All of these effects increase the kinetics of reactions and consequently, more homogeneous powder mixture forms after mechanical activation which raises reactivity and kinetics of reactions during combustion synthesis [8, 20, 31]. The entire procedure is named mechanical activation combustion synthesis.

Saadati et al. [1], Rahaie et al. [9] and Zhu et al. [27] synthesized NiAl/TiC composite by combustion synthesis of elemental powders. Their results showed that heterogeneous composite with different phases such as Al_3Ni_2 , AlNi_3 , and Al_3Ni was formed. In this work, the effect of mechanical activation before combustion synthesis of elemental powders on the microstructure features of the synthesized NiAl/TiC composite was studied.

2. Experimental Procedure

The starting powder mixtures to synthesize NiAl/TiC composite contained nickel (99.99%, -45 μm), titanium (99.99%, -45 μm), aluminum (99.9%, -40 μm), and carbon (graphite) (99.0%, -10 μm). The powders were mixed with 1:1:1:1 atomic ratio and mechanically activated by high-energy milling in a stainless steel container by stainless steel balls (different sizes) at 350 rpm speed which took 3 h under argon atmosphere. The balls to the powder mixture weight ratio were 15:1. After mixing and mechanically activation, as-mixed, and 3 h milled powder mixtures were pressed into compacts using a uniaxial force of 50 MPa by stainless steel die, and both compacts were subjected to furnace under vacuum to begin and complete combustion synthesis process. To determine phase development during milling and combustion synthesis, all powder samples were characterized by a Philips X'PERT XRD (x-ray diffraction) equipment with CuK_α radiation (1.5404 Å). The morphology and microstructure of powder samples were studied by SEM (scanning electron microscopy, Philips XL30) equipped EDS (energy dispersive spectroscopy) and TEM (transmission electron microscopy, Tecnai G2 F20-200KV) equipped energy dispersive spectroscopy.

The synthesized NiAl/TiC sample was mounted in epoxy resin and polished for the microhardness test was carried out at 0.01N loading for 15 s. At least six tests were performed.

3. Results And Discussion

The combustion synthesis method consists of two different modes, TE (thermal explosion) which combustion is performed by heating the total compact at the desirable temperature [32, 33] and SHS (self-propagating high temperature synthesis) which combustion is performed by heating one side of the compact and propagated to whole it [23, 34]. To determine the kind of combustion synthesis mode, it must be calculated adiabatic temperature (T_{ad}) of reaction takes place during combustion synthesis. Previous studies [15, 25, 29] proposed that the T_{ad} must be more than 1800 K to yield SHS mode in a compact.

Eq. (2) can be used to calculate the T_{ad} of reaction (1) with considering short reaction time and no heat exchanges between environment and compact; therefore, total formation heat of final products can be consumed to raise the temperature of compact [1, 28]. (see Equations 1 and 2 in the Supplementary Files)

Where ΔH_{298}° is the whole formation heat of final products (reaction 1) at room temperature, and c_p is the heat capacity. Based on thermodynamic data and previous studies [1, 27], the adiabatic temperature of reaction (1) was about 1911 K which is higher than 1800 K. Consequently, during ignition of as-mixed Ni/Ti/Al/C powder mixture, reaction (1) happens in self-propagating combustion mode.

The XRD spectra of as-mixed powder before and after combustion synthesis are shown in Fig. 1. As can be seen, the main phases of an as-mixed powder mixture are nickel (Reference code: 01-087-0712), titanium (Reference code: 00-044-1294), aluminum (Reference code: 01-085-1327) and graphite (Reference code: 01-075-2078). However, new phases such as TiC_x , Ni_3Al , Ni_2Al_3 , and $NiAl$ formed after the combustion synthesis of as-mixed powder. The formation of undesired phases like Ni_3Al and Ni_2Al_3 has been reported by Saadati et al. [1], Rahaie et al. [9], and Zhu et al. [27].

As reported previously, TiC_x with the amount of x between 0.55 and 0.95 is a thermodynamically stable phase [14, 23, 35]. Based on lattice parameters, the amount of x can be calculated. The lattice parameter of synthesized TiC_x is 4.310\AA which means that according to Ref. [14], the amount of x is about 0.75.

The atomic ratio of Ni and Al was equal, however, the undesired phases Ni_3Al , and Ni_2Al_3 formed. Zhu et al. [27] and Ozdemir et al. [36] reported that during the combustion synthesis of $NiAl$ at $T < 1000^{\circ}\text{C}$, the reaction between Al and Ni is a dissolution of Ni in molten Al, with the formation of some intermediate phases such as Ni_2Al_3 and Ni_3Al at the liquid-solid interface. By increasing the temperature to 1300°C , all intermediate phases disappear and $NiAl$ solely forms.

Similar to the proposed mechanism by Curfs et al. [15] in the Ni-Ti-Al-C system, the reaction begins by melting of Al powder (because the selected temperature was 950°C). After that, Ti, C, and Ni dissolved in molten aluminum and TiC_x primarily appeared and after a moment, nickel aluminides appeared.

Fig. 2 shows an SEM image for an as-mixed Ni-Ti-Al-C powder compact after combustion synthesis. As can be seen, some of the grey TiC_x particles have precipitated in nickel aluminides matrix. But, the main characteristic of this microstructure is thick layer structure which EDS analysis confirmed that the white layer is nickel-aluminum rich and grey one is titanium-carbon rich. Due to short reaction time and heterogeneous distribution of elements, precipitation of $NiAl$ and TiC , as one of the main parts of the mechanism, was not complete and therefore, the heterogeneous structure formed. These results (formation of $TiC_{0.75}$ and intermediate phases of the Ni-Al system) confirmed the heterogeneous distribution of elemental powders. Similar results have been reported by the previous studies [1, 27]. Also, the size of starting powders must be reduced to increase the surface contact area between powders and reduce diffusion paths because the reaction time during combustion synthesis is too short and no enough time exists to elemental diffusion. To overcome this problem, the starting powder mixture was

milled for 3 hours and mechanically activated. The mechanical activation of powders reduces crystallite and particle sizes of powders, fractured the oxide layer on the powders (especially Al powder) and increases surface contact area and interfaces between powders. These effects cause a more homogenous distribution of powders. Fig. 3 shows XRD results of 3 h mechanically activated powder mixture before and after combustion synthesis.

XRD spectrum of 3 h milled powder shows the formation of no new phases and only the height of diffraction peaks reduced and peak broadening happened. These changes without new phase formation are related to crystallite size reduction and the increase of lattice strain of powders. However, after combustion synthesis of 3 h milled powder compact, new phases NiAl and TiC_y formed. The amount of TiC_y lattice parameter was 4.316 Å and according to Ref. [14], the amount of y is about 0.95. The $\text{TiC}_{0.95}$ formation confirmed that the mechanical activation process caused better mixing of the elements and increases the contact of graphite and titanium particles. Therefore, more carbon reacted with titanium and $\text{TiC}_{0.95}$ formed. Also, no intermediate nickel aluminide phases were formed which these results confirmed that mechanical activation not only caused the homogeneous distribution of powders but also destroyed the oxide layer around Al powders which both of these effects facilitated NiAl-TiC composite formation.

The proposed mechanism is the molten Al distributed around more homogenized and fine nickel, titanium, and graphite powders uniformly and dissolved titanium, nickel, and graphite. Ni and Al reacted and NiAl formed. This reaction released energy and promote the reaction between Ti and graphite and consequently $\text{TiC}_{0.95}$ formed.

The microstructure of the 3 h milled powder after combustion synthesis was presented in Fig. 4. The matrix (NiAl) phase looks brighter than the TiC as reinforcement. The synthesized TiC is spherical and distributed homogeneously. The TiC boundaries indicate that TiC was formed before the NiAl formation. During the TiC formation, the heat released by this reaction can help further the formation of NiAl. Hence, TiC spherical particles surrounded NiAl. Also, the TEM image of the synthesized NiAl/TiC with chemical analysis in different points is shown in Fig. 5. These results also confirmed that the homogeneous distribution of NiAl and TiC phases was formed.

The hardness of the sole NiAl phase was reported between 330 $\text{HV}_{0.2}$ (synthesized by casting) [10] and 475 HV_1 (synthesized by sintering PPS)[37]. The addition of TiC (3000 HV)[38] increases the hardness of synthesized NiAl/TiC and Saadati et al.[1] reported that the hardness value of NiAl: TiC 1:1 composite is 838.5 HV. While the value of microhardness of synthesized NiAl/TiC composite in the present work was 1070 ± 12 HV. The main reason for this difference is a uniform distribution of spherical TiC reinforcement in NiAl matrix in the present work. Therefore, the mechanical activation process facilitated the reactions and improved the mechanical properties of final composite.

4. Conclusion

In this work, a homogenous NiAl/TiC composite was successfully synthesized after the mechanical activation of elemental powders. The product of combustion synthesis of a sample without mechanical activation was $\text{TiC}_{0.75}$ and NiAl, some other nickel aluminides such as Ni_2Al_3 and Ni_3Al formed. But, after the combustion synthesis of 3 h milled powder compact, no undesired phases were formed and a composite with a homogeneous distribution of NiAl and TiC was synthesized. The mechanical activation process decreased the crystallite and particle sizes of reactant powders which increased surface contact area of reactants and homogeneously distributed elements. The homogeneous distribution of TiC improved the mechanical properties of NiAl/TiC composite. The value of microhardness of synthesized NiAl/TiC composite was 1070 ± 12 HV.

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Figures

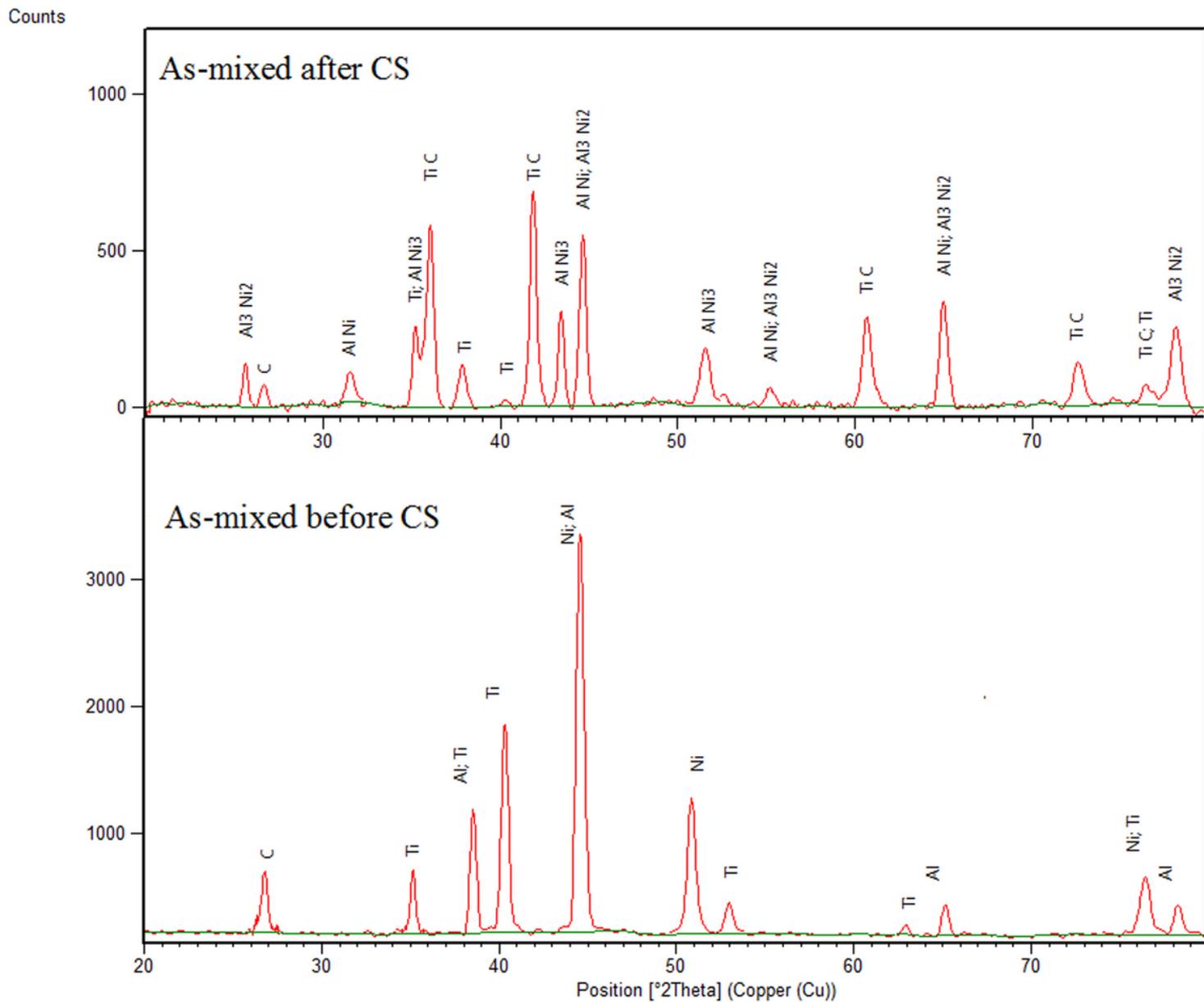


Figure 1

XRD patterns of as-mixed Ni-Ti-Al-C powder compact before and after combustion synthesis (CS).

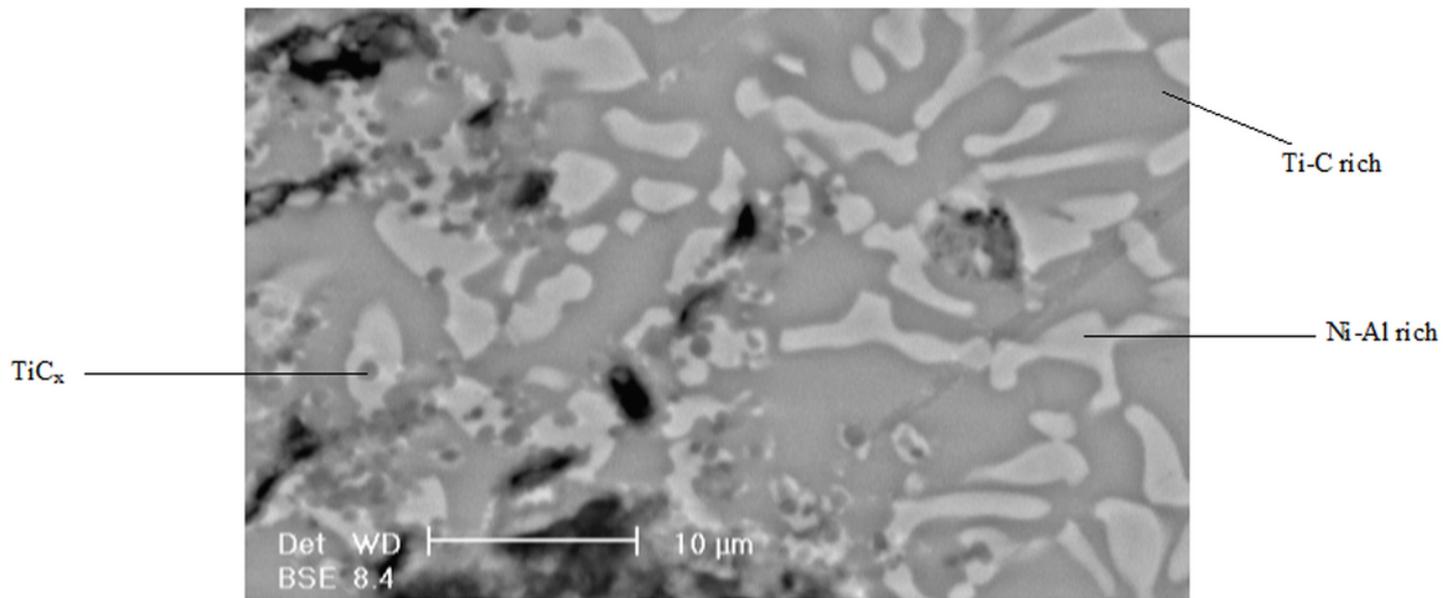


Figure 3

The microstructure of as-mixed powder compact after combustion synthesis.

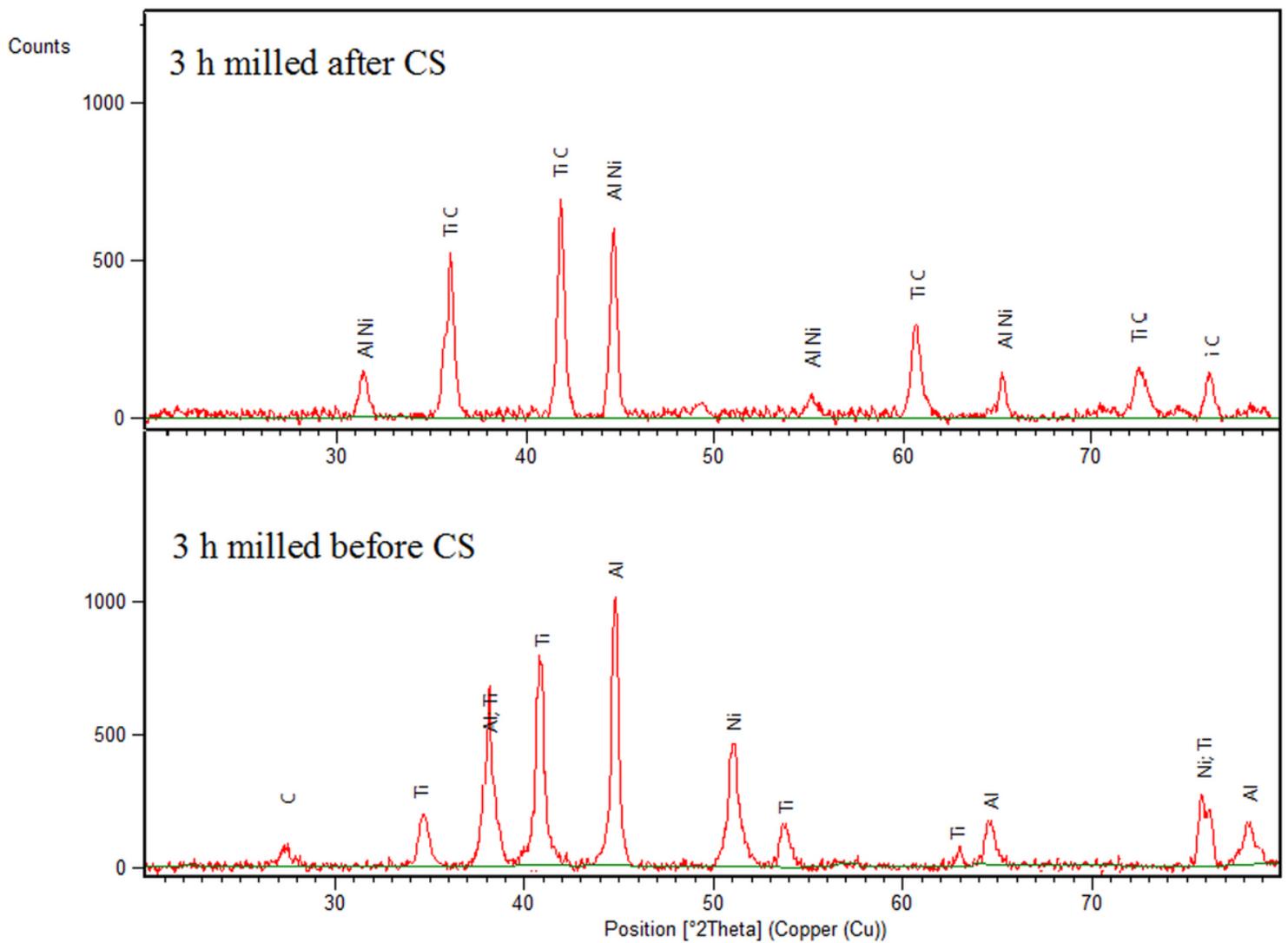


Figure 5

XRD patterns of 3 h milled Ni-Ti-Al-C powder compact before and after combustion synthesis (CS).

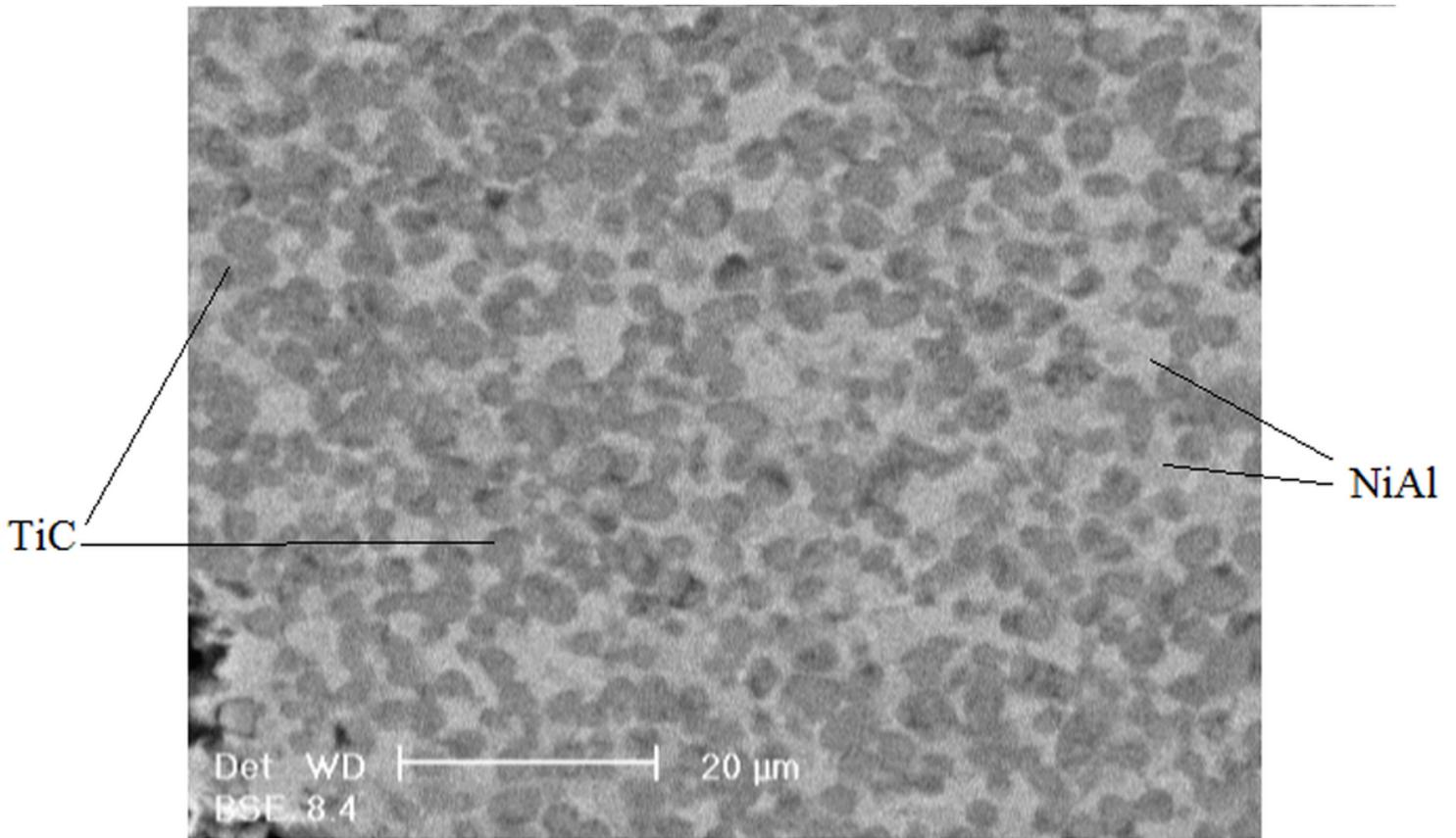


Figure 7

SEM microstructure of 3 h milled powder compact after combustion synthesis.

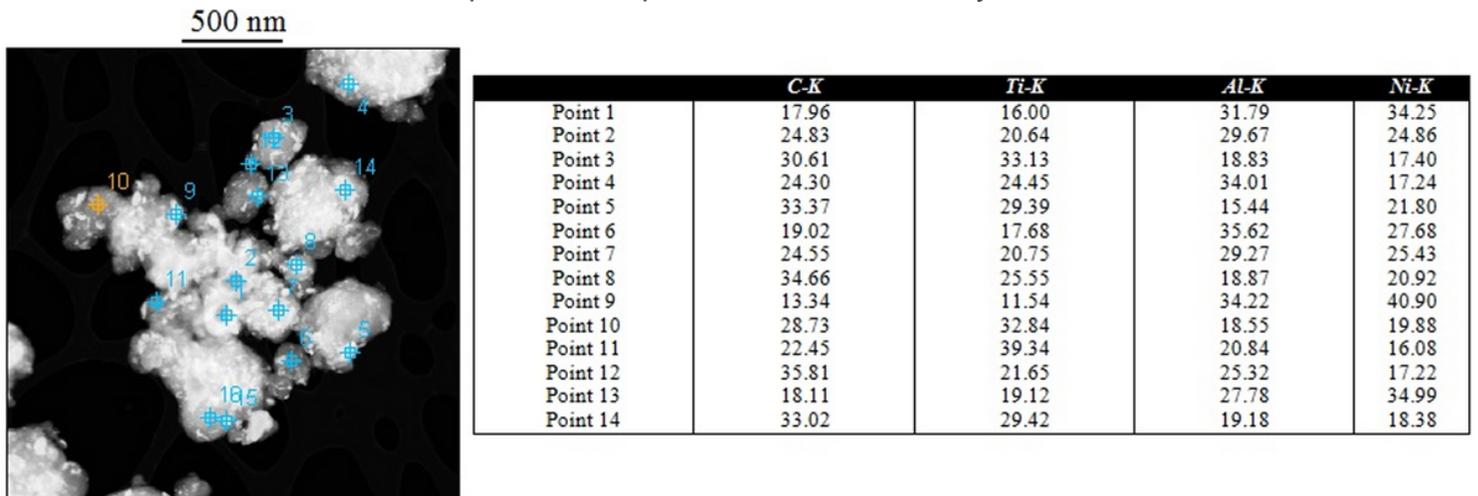


Figure 9

TEM image of NiAl/TiC composite synthesized after combustion synthesis of 3 h milled powder compact with chemical analysis of its different points.

Supplementary Files

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