

Reactive Nanofolds for Joining Refractory and Dissimilar Materials

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Abstract—Heterogeneous nanostructured foils produced by magnetron deposition or mechanical processing represent a new class of reactive materials. They are composed of layers or clusters of different phases (typically with a size of 10–100 nm) that can react with each other with strong heat release. The reaction, being initiated locally, spontaneously propagates across the entire foil in the form of high-temperature wave. Some examples of promising practical applications of these foils in advanced technologies, such as joining dissimilar materials, were presented.

Keywords: combustion synthesis, reactive multilayer films, joining dissimilar materials, Ni–Al system, C–C composites

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INTRODUCTION

Reactive multilayer nanofolds (RMNFs) represent a new type of functional materials – thin films, foils or tapes – generating large amount of heat in exothermic self-propagating heterogeneous reaction [1, 2]. RMNFs are used for joining heat-sensitive, dissimilar or refractory materials. Nowadays, RMNF with a layer thickness of 5–100 nm and up to 1000 layers in a foil are produced by layer-by-layer magnetron deposition. This technique was used to produce mostly bimetal Me/Al foils (Me = Ni, Ti, Co, Pd, Pt, Cu, Nb, Ta) [2]. Some metal/nonmetal RMNFs have also been produced by magnetron sputtering, including Nb/Si [3] and Ti/2B [4] ones. A high production cost and relatively low productivity of the method restrict practical implementation of magnetron sputtering to fabrication of RMNFs. As an alternative, a multiple cold rolling technique [5–9], also named as accumulative roll bonding process [10, 11], has been proposed. The mechanical methods afforded the production of nano- and micro-scale laminates in the mixtures of incompatible nonreactive or reactive metals.

Still another approach to the problem is the combination of high-energy ball milling with cold rolling. The idea suggested in [2] has been accomplished as a technological process in [12]. The foils cold-rolled from ball-milled powders turned rather nonuniform, intermittent, and tortuous as compared to magnetron-deposited films. Nevertheless, the reactivity of foils is

known to depend on their thickness rather than on their shape.

In this paper, we report on some features of combustion reactions in mechanically prepared reactive nanofolds, with special emphasis on their possible application to combustion-assisted joining of materials.

EXPERIMENTAL

Commercially pure powders of Ni, Al, Ti, Si were used for the preparation of reactive mixtures. The mixtures were subjected to high energy ball milling (HEBM) in an Activator 2S planetary ball mill (Russia) that had an independent drive for sun wheel and jars, thus allowing variation in their rotation speed separately [12]. Changing the rotation speeds between sun wheel and jars, we could obtain different ball motions inside the jar. Therefore, at the initial stage the effect of different milling regimes on the target system was investigated (five regimes). According to X-ray data, optimal milling regime and time were chosen so that the reaction product could not be detected in the jar. SEM observations showed that such a treatment led to the formation of composite particles in which Si and Ti were present in the form of nanolayers. Under optimal conditions of HEBM, the reaction initiation temperature decreased by 300°C for Ni + Al and by 550°C for Ti + 0.6Si mixtures. Magnetron-sputtered multilayer reactive Ni/Al nanofolds with a total thickness of

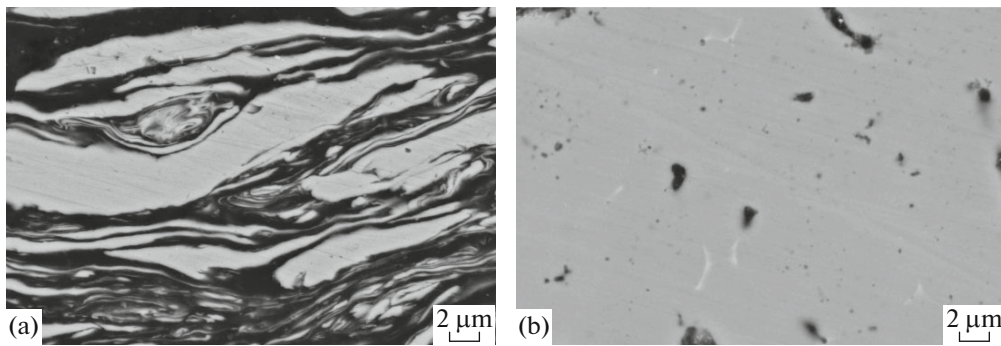


Fig. 1. SEM images of (a) reactive Ni–Al laminate after HEBM + CR and (b) NiAl intermetallic formed in combustion reaction. Here white corresponds to Ni, dark black to Al, and light grey to NiAl.

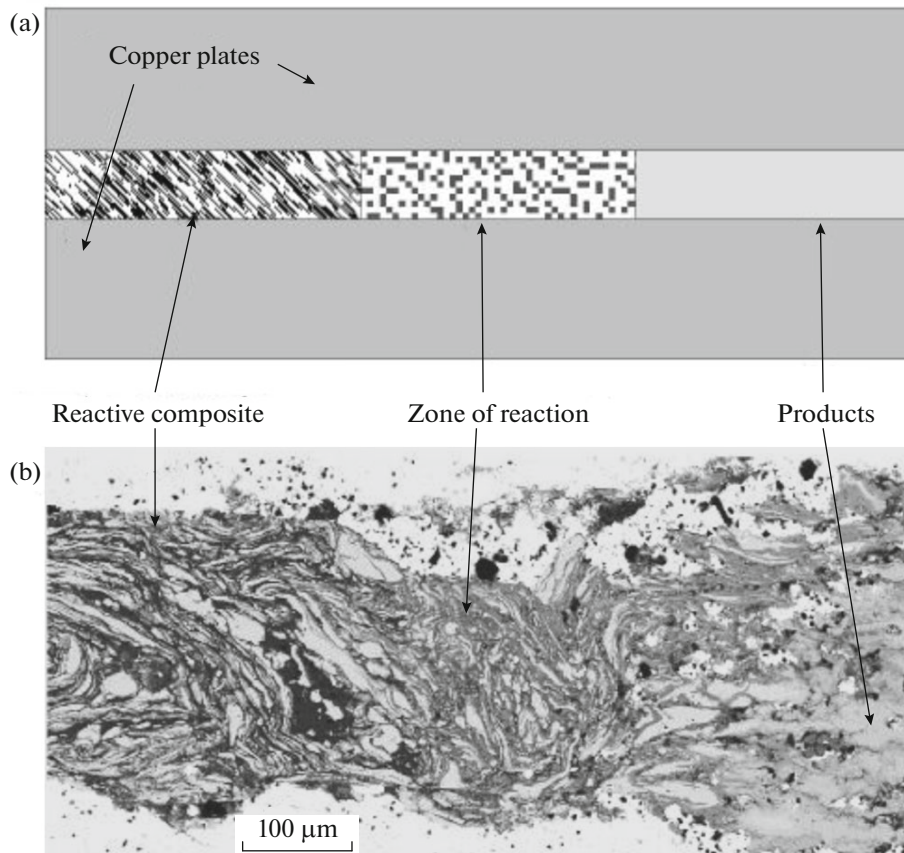


Fig. 2. (a) Schematic of quenching experiments and (b) SEM micrograph of the arrested reaction front in the Ni–Al laminate (b).

60–80 μm (purchased from Indium Co.) were used for initiation of the process at even lower temperatures.

Thus obtained powder mixtures were then subjected to cold rolling (CR) in laboratory-scale rollers to fabricate reactive tapes with a thickness of around 250 μm.

Reaction mechanism was investigated the quenching technique using two massive copper blocks at room temperature. Joining experiments were performed on the example of carbon materials (graphite and C/C composites) by placing a tape between two

carbon-based materials and installing this stack between the electrodes of a spark plasma sintering device (SPS) for heating up to 1600°C at a heating rate of 100 deg/min under applied force (3 kN).

RESULTS AND DISCUSSION

Figure 1 shows the SEM images of (a) reactive Ni–Al laminate after HEBM + CR and (b) NiAl intermetallic formed in combustion reaction. Upon combustion, a heterogeneous lamellar structure composed of

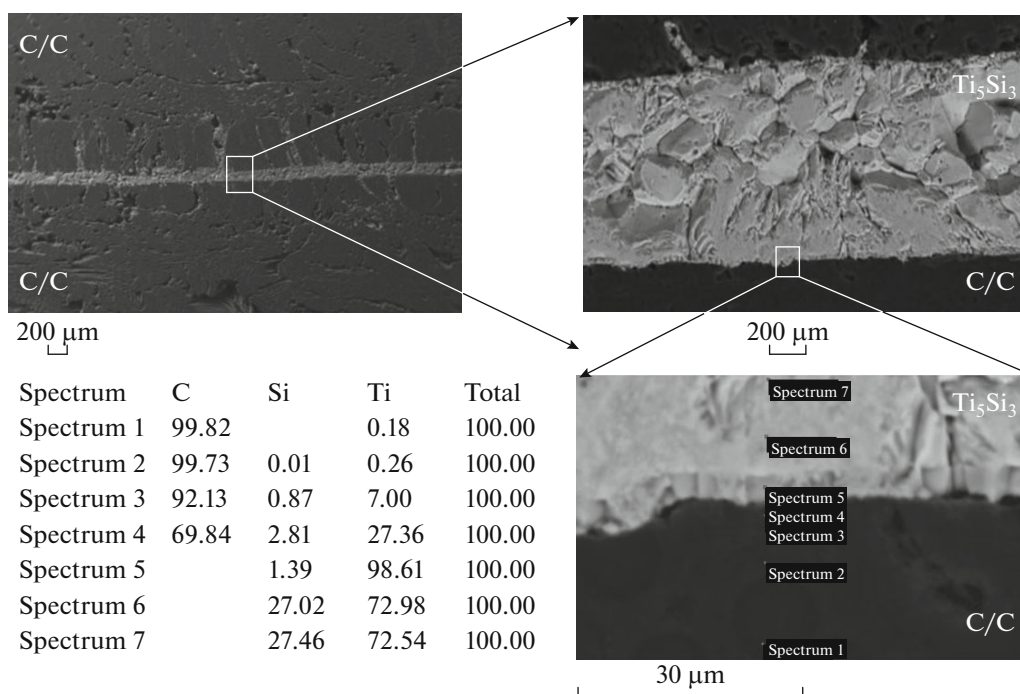


Fig. 3. SEM image and EDS results for the cross section of the C/C joint obtained via reactive Ti + 0.6Si tape.

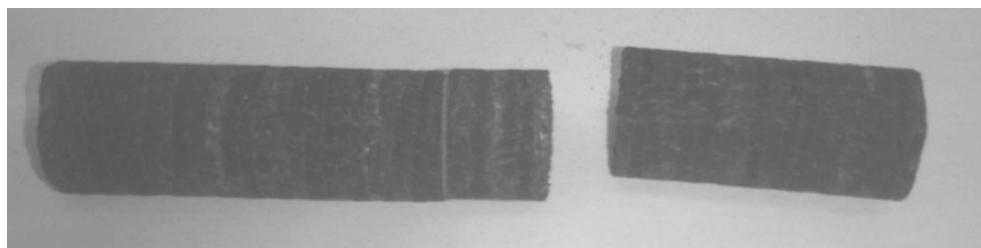


Fig. 4. Broken sample of combustion-joined C/C composites after tensile strength test. Breakage occurred over the bulk material, while the joint is seen to remain intact.

the Ni and Al layers (Fig. 1a) transforms into uniform NiAl intermetallic. A heat release of about 120 kJ/mole is sufficient for self-sustained propagation of combustion wave. The temperature of products attains a value of 1640°C for Ni + Al and 2600°C for Ti + 0.6Si mixtures, which is sufficient for melting metal constituents and hence for combustion-assisted joining.

The dynamics of chemical and phase transformations in the Ni–Al laminate are illustrated by Fig. 2. When the heat losses exceed some critical value (e.g. when the reactive foil is placed between two Cu plates, Fig. 2a), the propagating reactive front is arrested, and some features of the process can be revealed.

The width of the reactive zone is about 0.3 mm, which corresponds to the time of transformation around 0.1 s or less. The first product phase registered in the reaction zone was NiAl₃ which immediately converted into NiAl in reaction with residual Ni. It can be seen that the Ni layers probably remain solid during

reaction (they lost their shape gradually by dissolving in aluminum). Therefore, the combustion temperature was lower than the melting point of Ni. In order to decrease the reaction time and increase the temperature, for joining carbon-based materials we used Ti + 0.6Si mixtures with a higher reaction heat. An additional reason was better wettability of carbon with liquid Ti and Si.

Figure 3 presents the SEM image and EDS results for the cross section of the C/C joint obtained via reactive Ti + 0.6Si tape. The reactive tapes were placed in between the cylindrical samples of C/C composite, and the uniaxial pressure was applied along the cylinder axis in an evacuated SHS reactor. Reaction was initiated by Joule heating with current pulses. The Ti foils were placed between the reactive ribbon and the carbon materials in order to improve the wettability and adhesion. It can be seen in Fig. 3 that a crack-free transition layer 150–200 μm thick was produced. Also,

it was observed that part of the resulting interlayer infiltrated 100 μm deep into the porous skeleton of the C/C composite. In addition, the bonding of C/C composites was facilitated by applied pressure, which favored the formation of dense, pore-free, and crack-free transition zone.

Figure 4 shows the overall view of the broken sample of combustion-joined C/C composite after tensile strength test. Breakage occurred over the bulk material; while the joint is seen to remain intact.

CONCLUSIONS

The results of this work show that combination of the HEBM and subsequent cold rolling is a promising method for producing reactive nanostructured foils (ribbons) that can be used as energy source during joining different materials.

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