

Additive Manufacturing of Copper/Diamond Composites for Thermal Management Applications

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Supporting Information

I. Titanium oxide-titanium carbide (TiO₂-TiC) coating on diamond particles by a molten salt process

Diamond (D) particles with an average diameter of 105 μm (Eastwind Diamond Abrasives) were used in this study. The molten-salt synthesis was carried out as schematically illustrated in **Figure S1**. For that, potassium chloride salt (KCl) (Rectapur[®] 99%) and metal titanium (Ti) powders (99.5%, mean diameter 40 μm) were homogeneously mixed using an agate mortar. Next, D particles were added to the mixture and manually shaken for a few seconds to disperse them in the salt mixture at a molar ratio of KCl:D:Ti = 0.64:1:0.04. Later, the mixture was air dried for ~16 h (overnight) at 60 °C. The mixture was then heated in an alumina (Al₂O₃) crucible under flowing argon (Ar) at 850 °C for 4 h. Once the synthesis was done, the crucible containing the final product was immersed in boiling distilled water until the salt was fully dissolved. The coated Ds were filtered from the solution and flushed with deionized (DI) water and air dried in an oven at 60 °C for 1 h.

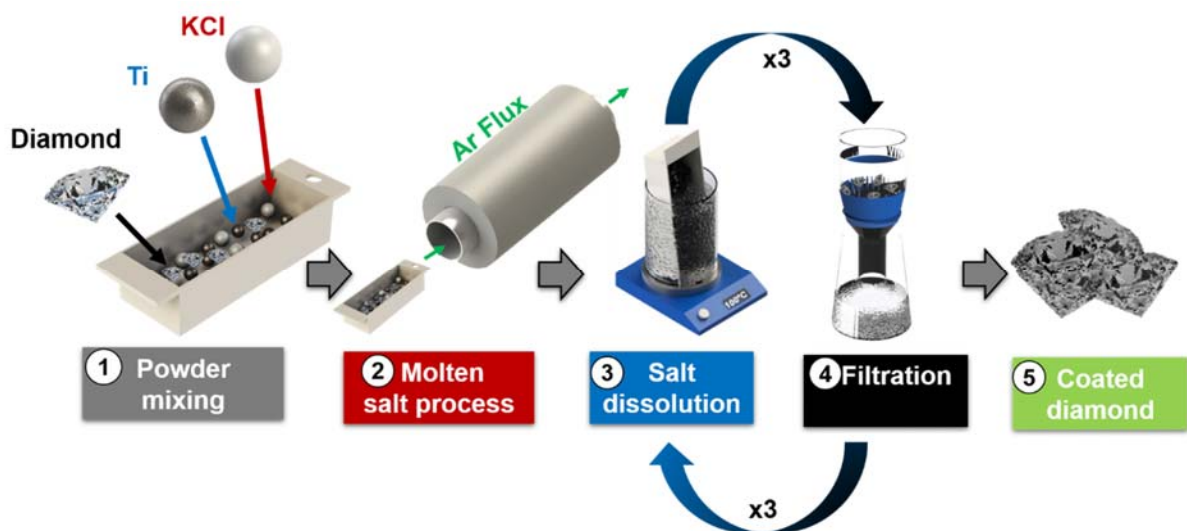


Figure S1. Schematic illustration of the molten salt coating process.

II. Laser engineering net shaping and printing parameters

The Optomec, Inc., LENS 3D Hybrid Machine Tool System is equipped with a continuous wave (CW) fiber laser (Nd: YAG, $\lambda=1064$ nm) with a maximum power of 2000 W. The laser beam has a focal point of 600 μm . In this study, the laser power was varied from 700 to 1000 W. The scan speed was tuned from 4.25 to 21.7 mm/s. The hatch distance and layer thickness were kept constant to 0.30 and 0.20 mm, respectively (**Figure S2 (a)**). The scan strategy was a meander, as illustrated in **Figure S2 (b)**, with a rotation of 90° between each layer.

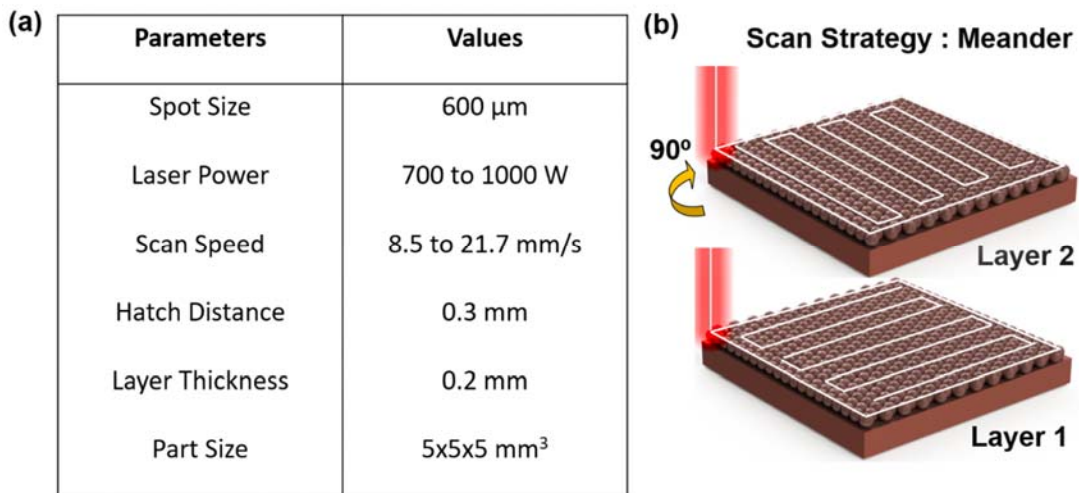


Figure S2. (a) Table of printing parameters and (b) schematic illustration of the meander scan strategy.

III. X-ray diffraction of the coated diamond powder

The phase and crystallinity of the coated D particles were analyzed by X-ray diffraction (XRD). Diffraction peaks at $2\theta = 35.9^\circ$, 41.7° , 60.5° , 72.4° , and 76.1° can be observed in **Figure S3**, corresponding to the face-centered cubic TiC phase. Also, diffraction peaks at 39.8° and 60.6°

confirmed the presence of the tetragonal TiO_2 phase. In addition, peaks at 43.9° and 72.3° can be attributed to the face-centered cubic diamond phase.

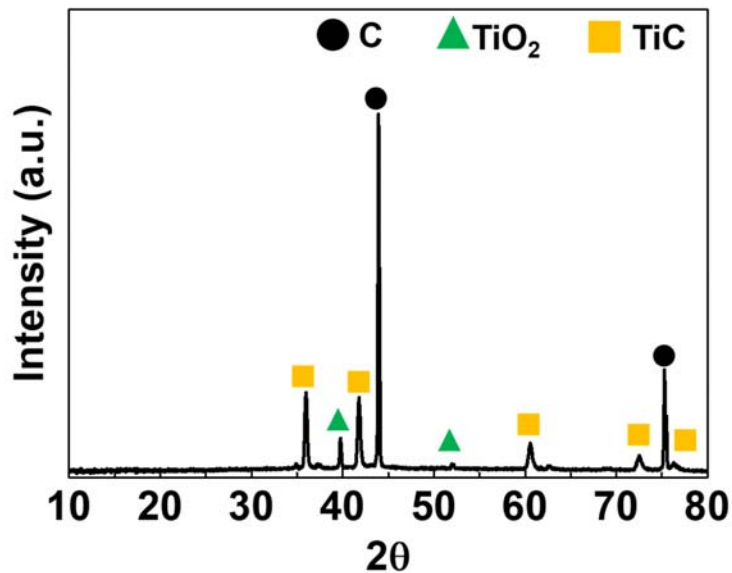


Figure S3. X-ray diffraction of coated D with TiO_2 -TiC layers.

IV. Thermal conductivity measurements

The periodic photothermal radiometry (PPTR) technique [1], used to measure the thermal diffusivity (TD) of a printed copper (Cu)-D composite material, is schematically illustrated in **Figure S4 (a)**. The setup is equipped with an Nd: YAG laser (Coherent MATRIX Q-switched diode-pumped laser, $\lambda = 1064 \text{ nm}$) that delivers a 40 ns pulse duration each period T_R with a maximum power of 10 W (**Figure 4 (b)**). The laser beam (diameter 0.55 mm) is directly focused on the sample; an infrared (IR) detector (Teledyne Judson Technologies J15D12) is used to monitor the temperature change at the rear face of the sample. Also, a photodiode was set to trigger the acquisition after each laser pulse. An averaging process was performed over the period of repetition that allows to increase the signal noise ratio of the response by the detector. The TD of

the sample is identified by minimizing the gap between the experimental response and the theoretical one calculated using an analytical solution of the heat transfer within the material. The minimization is achieved by implementing a nonlinear least square algorithm (the Levenberg-Marquardt technique).

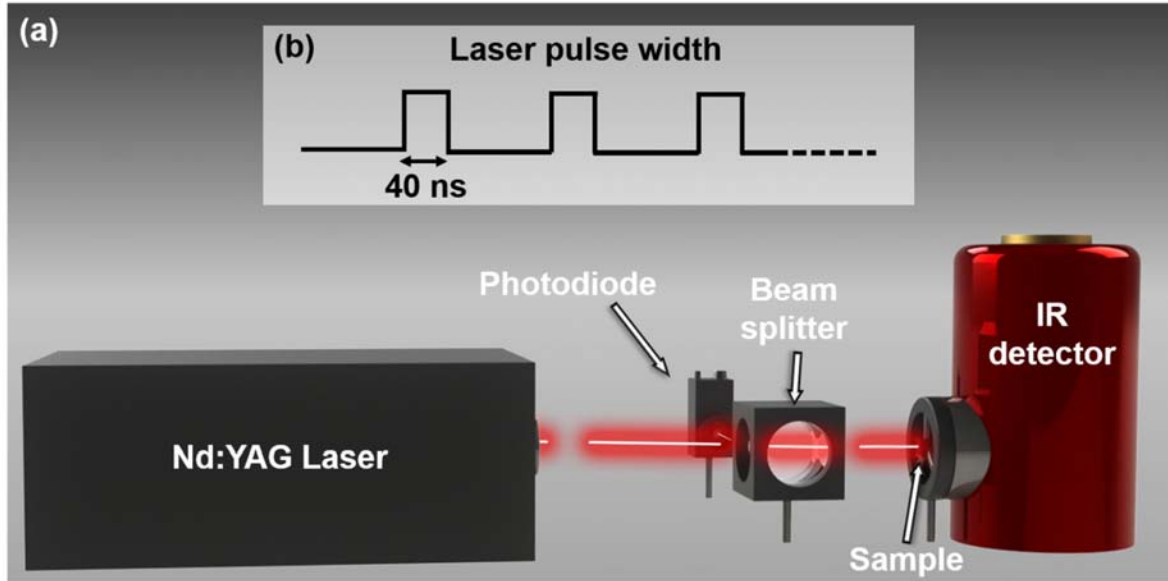


Figure S4. (a) Schematic illustration of the laser flash setup for thermal conductivity measurement and (b) illustration of the laser pulse width.

Figure S5 (a) and **(b)** presents the experimental data of the TD measurement using the PPTR method. A close correlation between the experimental curve and the model can be observed in **Figure S5 (b)** with a low residual signal. The measured diffusivity was $104.573 \text{ mm}^2/\text{s}$ for a laser frequency of 9 Hz. The thermal diffusivity was converted into thermal conductivity (TC) using the following equation:

$$k(T) = a(T) \times C_p(T) \times \rho(T), \quad (1)$$

where k is the TC (W/m.K), a is the TD (mm^2/s), ρ is the density, and C_p is the heat capacity of the sample (J/Kg.K). The C_p of the diamond, TiC, and Cu used in this study are 630, 190, and 392 J/Kg.K, respectively [2], leading to the thermal conductivity of 329.419 W/m.K.

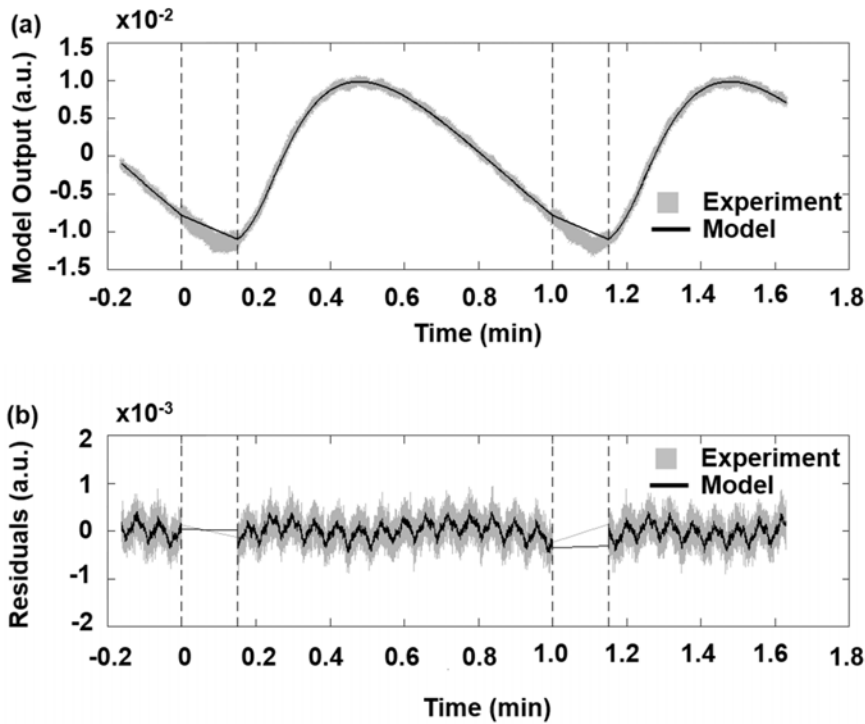


Figure S5. (a) Plot of the measured (grey dots) and theoretical temperature with the identified thermal diffusivity vs. time and (b) residuals vs. time.

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- [1] E. Ruffio, C. Pradere, A. Sommier, J.-C. Batsale, A. Kusiak and J.-L. Battaglia, Signal noise ratio improvement technique for bulk thermal diffusivity measurement. *International Journal of Thermal Sciences* 2018;129:385–95. <https://doi.org/10.1016/j.ijthermalsci.2018.03.011>.
- [2] Azina C, Cornu I, Silvain J-F, Lu Y, Battaglia J-L. Effect of titanium and zirconium carbide interphases on the thermal conductivity and interfacial heat transfers in copper/diamond composite materials. *AIP Advances* 2019;9:055315. <https://doi.org/10.1063/1.5052307>.