Solid-flame: Experimental Validation

C. E. Shuck¹, K.V. Manukyan², S. Rouvimov³,
A. S. Rogachev⁴ and A.S. Mukasyan^{1*}

¹Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, IN 46556, USA

²Department of Physics, University of Notre Dame, Notre Dame, IN 46556, USA

³Department of Electrical Engineering, University of Notre Dame, Notre Dame, IN 46556, USA

⁴National University of Science and Technology MISiS, Moscow, 119049, Russia

* corresponding author: amoukasi@nd.ed

Abstract

The tantalum-carbon reactive system possesses a high energy of reaction with an adiabatic combustion temperature of 2743 K, which is significantly below the melting points of the reactants, as well as any intermediate phases and final products. It was suggested that a combustion wave could propagate in Ta+C mixtures solely owing to a solid-solid reaction. However, this combustion process has never been shown to occur without gas-assisted transport. Here, we report preparation of highly pure and pore-free Ta/C composite particles, which were used for experimental validation of the solid-flame. Preparation of these composite particles involves short term high-energy ball milling (HEBM) of tantalum and carbon powders. High-resolution microscopy coupled with three-dimensional reconstruction techniques were used to characterize the volume nanostructure of mechanically fabricated composite particles. It was quantitatively shown that the particles have nano-scale mixing of the reagents and possess high contact surface area between tantalum and carbon. Experiments revealed that the ignition temperature of as fabricated composite particles is 1243 ± 15 K and maximum combustion temperature was shown to be 2487 ± 50 K, which is well below any possible solid-liquid transitions. Utilizing results obtained with composite particles prepared under different HEBM conditions, it is shown that carbon diffusion through the tantalum grain boundaries and subsequent formation of a Ta(C) solid solution defines low temperature ignition of mechanically fabricated particles. The high surface area contact between the Ta and C nano-scale reagents allows the reaction to propagate in a self-sustained manner, owing solely to a solid-state diffusion mechanism.

Keywords

Combustion synthesis; Mechanical activation; High-energy ball milling; Self-propagating high temperature synthesis; Solid flame; Self-sustained Reaction; Solid-state Diffusion; 3D Reconstruction; Nano-structural Media; Heterogeneous Combustion

Introduction

Combustion synthesis (CS) or Self-Propagating High Temperature Synthesis (SHS) is a well-established non-traditional energy saving approach to produce a variety of materials, including metals and their alloys, ceramics and cermets, composites and functionally graded compounds, 2D-crystals, and nano-powders [1-6]. Although CS is attractive for many applications, there are still fundamental issues in solid physics that need to be resolved. One of these issues is the concept of a solid-flame [7]. This came about in 1967, when a group of Russian scientists from the Semenov Institute of Chemical Physics, headed by Prof. A. Merzhanov, discovered a wave localization phenomena for solid state self-propagating reactions [8], or in modern terms the *solid flame*. The fundamental basis for this phenomenon is the following: (i) in some exothermic reactive mixtures (e.g. Ta-C and Mo-B), the adiabatic combustion temperature, while being high, is still well below the melting points of the precursors, any intermediates, and final products (see Figure 1 for the Ta-C system); (ii) the amount of equilibrium gas phase products in these systems are negligibly small; (iii) it was experimentally shown that a combustion wave may propagate in a self-sustained manner along such reactive heterogeneous media with velocities ranging from 0.1-1 cm/s. Thus, it was suggested that a solid-state reaction, localized in the combustion front, may self-propagate in a heterogeneous gasless system, which leads to the concept of a solid flame.

From the standpoint of conventional combustion, a solid flame is enigmatic, because it is hard to believe that solely solid-state diffusion may define the self-sustaining nature of the combustion process. It is a prevalent opinion that self-propagating combustion processes, such as metallothermic reactions, exist due to relatively fast mass transport in the liquid phase (diffusion and convection), therefore, melting of at least one component in the system is considered as a

necessary condition for combustion. Recently this requirement was even announced as a “new criterion” for sustainability of the combustion reaction: “*adiabatic temperature ... must be high enough to melt the lower melting point component*” [9]. On the other hand, the regularities of combustion in Ta+C mixtures were studied by Shkiro and co-authors assuming that the reaction proceeds completely in the solid state [10]. It was shown that the combustion wave in the Ta-C system can propagate in the pulsating mode at temperatures below the lowest melting or eutectic point. More careful experimental investigation of the combustion mechanism revealed, however, a gas-transport stage in the combustion reaction for this system [11]. In that article, based on detailed investigation of the structural transformations taking place in the combustion front, it was shown that impurities enhance mass-transport in the Ta-C system. Investigation of the microstructural transformations in the quenched combustion wave revealed that:

- The tantalum carbide (TaC) crystals that formed in the leading zone of the reaction front have columnar morphology and uniformly cover the particle surface, despite that, in the initial mixture, the reagents have only point contacts;
- The final combustion product consists of globular TaC particles.

These structural transformations led to the hypothesis that the existence of a gas phase and melt leads to enhanced reagent transport, and consequent product formation in this system. These phases form due to the impurities present in the initial reagents, such as oxygen, in a form of oxide film on the surface of the metal particles, and iron, introduced into the system during the mixing process. A variety of model experiments confirmed this hypothesis; small amounts of iron, distributed on the tantalum surface, accelerate combustion along with gas phase mass-transfer of carbon through the Bouduar-Bell cycle, which reduces the tantalum oxide film and leads to the formation of columnar structures on the surface of the metal particles. In 2001 the

term “*catalytically assisted combustion synthesis of tantalum carbide*” has been introduced by Kim and Wooldridge [12]. They indicated that the presence of iodine vapor and carbon dioxide significantly enhances the combustion synthesis process, leading to higher conversion efficiencies and influencing the product microstructure. There are also reports by Kashireninov et al. [13] and Egishyan et al. [14] on the leading role of gas phase reactions during combustion in the other thermodynamically solid flame system, i.e. the Mo–B mixture.

Thus the question still remains: can the solid-flame be accomplished in its pure form? A.G. Merzhanov suggested a term “*ideal solid flame*” for a combustion processes where all gas and liquid phases are absent, as opposed to real solid flames where impurities can promote the process through formation of small amounts of gases or liquids [7]. Till now, the existence of the ideal solid flame has not been proven. In order to assert the ideal solid flame in the Ta-C system, one has to form a pore-free media of tantalum and carbon, with no impurities that have melting points below the adiabatic combustion temperature. In this case, there is no space (pore-free) for gas transport and the melt cannot be formed in the combustion wave. In order to create such a media, advanced processing techniques must be utilized.

Short term (<10 min) high energy ball milling (HEBM) has received considerable attention in recent years as a method for fabrication of composite nano-structured reactive particles with oxide-free surface contact between solid reactants [15-18]. The mixture of powders, which initially only have point contact with each other, is subjected to the intensive (acceleration in the range 20-300G) mechanical treatment under an inert atmosphere in a ceramic (ZrO₂) milling jar. As a result, new, typically pore-free, composite particles are formed, which involve both elements, mixed on the nano-level with essentially oxygen-free contact surfaces. It is also important, as was shown for many systems, such structural transformations significantly

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4 decrease the reaction onset (ignition) temperature and accelerate the combustion front
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6 propagation [19,20].
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9 In this work we used short term (4 min) HEBM for preparation of composite Ta/C
10 particles. The internal nano-structure of these particles was investigated and characterized using
11 3D-reconstruction techniques. The ignition and combustion parameters of the reactive media
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13 were thoroughly studied. Analysis of the obtained data allows us to conclude that the solid flame
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15 phenomenon can be accomplished in such a system. To support this conclusion, a simple model,
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17 based solely on solid-state diffusion, is also proposed and discussed.
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26 **Experimental**

27 *Initial Reactants*

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29 Tantalum powder (Materion Advanced Chemicals, 99.9% pure, <44 μm) and carbon
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31 lampblack (Fisher Scientific, 99% pure) were used as precursors. EDS analysis reveals less than
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33 1wt. % of oxygen in the initial powder mixture. The microstructures of these powders are shown
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35 in Figure 2. It can be seen that the metal particle sizes range from 5 - 20 μm (Fig.2a) and are
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37 comprised of irregular planar geometries (Fig.2b), while carbon lampblack particles (Fig.2c) are
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39 agglomerations of much smaller (<2 μm) spherical carbon particulates (Fig.2d).
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45 *High Energy Ball Milling*

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47 HEBM was performed by a PM100 (Retsch, Germany) planetary ball mill in a 500 mL
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49 zirconium oxide jar with 1 mm balls of the same material. The jar was filled with 99.999% pure
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51 argon. The ball to powder ratio was 40:1. A rotational speed of 650 RPM was used, with the
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53 internal rotation speed being 1300 RPM. For each treatment, 6 g of an equiatomic (1:1) mixture
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55 of Ta + C was subjected to dry milling (DM) for a duration of up to 12 min.
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Material Characterization

The particle microstructures were examined by a field emission scanning electron microscope Magellan 400 (FEI, USA), which has a spatial resolution 0.9 nm at an acceleration voltage of 20 keV and is equipped with a Bruker (USA) energy dispersive X-ray spectroscopy (EDS) analyzer. Analysis of the material structure on the atomic level was performed with a transmission electron microscope (TEM) Titan 80–300 (FEI, USA) at 300 kV, using both high resolution TEM (HRTEM) and STEM modes at 2 Å (point-to point) and 1.34 Å resolution, respectively. The composition analysis at nano- and sub-nano spatial resolutions was performed with EDS using Oxford Inca spectrometer with energy resolution of 133 eV.

A Nanolab 600 Helios Dual SEM/FIB (FEI, USA) was used to prepare TEM samples, as well as for 3D reconstruction studies. In the former case, the specimens (about 1 x 12 x 6 μm in size) were first cut by ion beam from the surface of the ball-milled powder particulates, placed on half-moon TEM copper grid, and then further thinned and polished by use of ion beam to remove any residual surface damage to a thickness of ~ 70 nm. In the latter case, the slice and view (S&V, FEI) software package was used to collect a series of 110 images with ~8 μm width and ~7 μm depth, with a voltage of 30 kV and with milling current of 28 pA. These images were taken in series after milling 25 nm in between each image, leading to a particle volume of ~150 μm³ imaged throughout and prepared for layer characterization. 3D reconstruction was completed with the AvizoFire (FEI) software package. The series of images collected from the FIB S&V were first shear corrected, contrast normalized and then aligned using a least-squares method. The image stack was stitched together with linear interpolation methods. The reactants were isolated by use of contrast thresholding. 3D skeletonization using a distance-ordered

thinner was used in conjunction with a 3D chamfer distance map to obtain nano-structural characteristics of the media, e.g. metal layer thickness, of the composite particles.

The overall phase composition of the materials was also determined by X-ray diffraction (XRD) analysis with Ni-filtered Cu-K α radiation (D8 Advance, Bruker) operated at 40 kV and 40 mA. Step-scan data (step size of 0.02° and counting time of 5 sec) were recorded over a range of 20-90° (2 θ).

Reaction Characterization

The thermal ignition and combustion characteristics of the reactive media were studied using two methods. To determine the combustion propagation velocity, the sample was pressed into a 10 mm diameter pellet of 20 mm height (65 theoretical maximum density (TMD%)). The sample was placed into a sealed reaction chamber in a 99.999% pure argon atmosphere. A coiled tungsten wire was placed above the pellet and resistively heated, leading to ignition of the sample. A high-speed infrared thermal imaging system (SC6000, FLIR Systems, MA, USA) was used to explore the ignition characteristics of the process. This was repeated five times. With FLIR's ExaminIR software, thermal images and videos can be captured over several different temperature ranges up to 2300 K. Depending on the frame size and temperature range of the investigation, frame rates upward of 15,000 fps are achievable. The use of a high-speed IR camera obtains temperature distributions on the sample surface during the heating process, making it convenient for monitoring ignition and combustion wave propagation processes. Additionally, a C-type thermocouple was used to monitor the temperature inside the pellet during propagation.

In order to study the thermal ignition temperature (T_{ig}), eight cylindrical samples, 3 mm in diameter and 0.5 mm height, were pressed to 70% TMD. These samples were individually

placed into a boron nitride crucible and were then heated with a carbon strip heater in a 99.999% pure Ar atmosphere at a rate of 500 K/s. A 100 μm W-Re thermocouple was used to measure the time-temperature profile of the sample. Ignition was determined when a sharp increase in temperature occurred, corresponding to an increased temperature derivative.

Results and Discussion

Typical powder microstructures of the initial Ta+C mixture and the 4 min dry HEBM (DM) are presented in Figure 3. Comparing Fig.3a and c reveals that the average particles size increases after HEBM. Additionally, after HEBM, the composite particles have plate-like shapes. These structures are formed from the repeated shear forces applied during intensive milling. The average size of the media after HEBM is on the order of 30 μm . Also, the composite mechanically-induced Ta/C particles have a more textured surface (Fig.3d), including sphere and disc-like structures, this is in contrast to the initial Ta particles (Fig.3b and Fig.2b), which have sharp features and interfaces. This indicates that cold welding plays an important role in formation of the Ta/C composite particles (Fig.3c) during HEBM.

XRD patterns for the initial equiatomic Ta+C powder mixture, and the Ta/C particles after different DM times (4, 6, 12 min) are shown in Figure 4. The Ta-C system, unlike Ni-Al [17,21] and Ti-C [22], does not undergo a spontaneous complete reaction at some critical milling time. Instead the carbide phase starts to form after ~ 6 min of DM with conversion increasing with milling time, which is qualitatively similar to the Si-C system [23]. Additionally, the Ta peak both shifts to the left and broadens significantly with increased milling time, specifically after 4 min. This indicates that the lattice constant of the Ta crystals are increasing due to the formation of a solid solution between Ta and C (Fig.1), while the overall crystalline size of the

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4 Ta crystals is being reduced. After only 4 min DM, neither shifting of the Ta peaks, nor
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6 formation of the TaC phase (within method sensitivity) can be detected.
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9 A typical cross section of the mechanically induced (4 min) composite Ta/C particle is
10 shown in Fig. 5a. Using contrast thresholding during 3D analysis, the internal porosity of the
11 samples was measured to be minimal (<2%). The composite particles are essentially pore-free,
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13 fine lamellar internal structure, which involves Ta (light) and C (dark) phases. Obtaining the
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15 stack of images by FIB S&V, as described above, and utilizing the 3D reconstruction package
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17 AvizoFire, a 3D representation of the reactive media can be obtained (Fig.5b, gray area
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19 represents the Ta phase, the C phase is represented as void space). Utilizing skeletonization and
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21 distance mapping, the characteristic scale of the diffusive layers (Ta and C) and their size
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23 distributions can be determined, as shown in Figure 5c. For the 4 min DM composite particles,
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25 the average Ta layer thickness is 560 nm with σ of 232 nm, and the average carbon thickness is
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27 659 with σ of 290 nm, where σ represents the dispersion of the histogram. As milling time is
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29 increased, both the Ta and C layer thicknesses decrease. EDS analysis reveals that the main
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31 impurity in the powder is ZrO_2 , which has a melting point of 2998K, well above adiabatic
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33 combustion temperature of the system. Essentially no iron contamination was found and the total
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35 oxygen concentration does not increase compared to the initial mixture. Thus, short term HEBM
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37 allows fabrication of relatively large pore free nanostructured Ta/C composite particles, which
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39 do not contain any impurities that have low melting temperatures. A representative time-
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41 temperature profile of the pressed thin disks on the BN-hot plate is shown in Figure 6. It was
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43 calculated that the self-ignition temperature is 1243 ± 15 K for the 4 min DM sample. A similar
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45 result was obtained using a resistively heated W-coil to radiatively heat the sample. For this
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47 ignition scheme, the reaction onsets at $T_{ig} = 1281 \pm 14$ K. Utilizing the high-speed IR camera, it
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4 was determined that this reaction propagated with a combustion velocity of ~ 0.7 cm/s, as shown
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6 in Figure 7. Using a C-type thermocouple, the maximum combustion temperature was measured
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8 as $T_c = 2487$ K. It is important to note that neither the initial Ta+C mixture, nor the composite
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10 particles after either 6 or 12 min DM, could be ignited under the same heating conditions.
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14 Thus, the composite nanostructured Ta/C particles, mechanically induced under optimum
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16 conditions, may combust in an inert atmosphere in a self-sustaining manner. The measured
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18 characteristic temperatures (T_{ig} , T_c) are well below the melting points of the precursors,
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20 impurities, and any intermediate or final phases. To support the solely solid-state mechanism of
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22 reaction, it is important to examine the structural transformation, which takes place in the
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24 combustion wave.
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29 The XRD patterns for the composite powders both initially and after combustion are
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31 shown in Figure 4. The data indicate that the reaction leads to the formation of the TaC phase to
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33 almost full conversion, i.e. only traces of the Ta phase can be detected.
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37 SEM images illustrating the morphology of the combustion product are shown in Figure
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39 3e,f. Comparing these images with Figure 3c,d shows that the particle morphology, on the
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41 micron-scale, remains essentially unchanged. This suggests that the combustion wave occurred
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43 without melting or gasification processes, otherwise if melting or gasification had occurred,
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45 significant change of the particle morphology would be apparent [1].
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49 In order to understand the structural transformations that are occurring, TEM studies have
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51 been performed on the composite particles of varying milling times (4, 6, 12 min). The TEM
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53 images in Figure 8 show the internal nanostructure of the composite particles before and after
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55 combustion. A comparison between the pre- and post-combusted nanostructures reveals that they
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57 both have lamellar structures of approximately the same thickness. In the pre-combusted
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4 samples, the lamellas are comprised of nanocrystals, with relatively narrow size distribution.
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6 These polycrystalline structures are completely pore-free and have intimate contact between the
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8 crystals within the lamellas. Investigating the nanostructure of the post-combusted particles, it is
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10 seen that the TaC crystals are unequivocally comprised of roughly spherical nanocrystals. These
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12 nanocrystals are ordered in a lamellar structure. Additionally, the nanocrystals have a relatively
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14 narrow size distribution; there do not appear to be any large, thick single crystals. A significant
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16 difference is observed within the lamellas in the post-combusted samples; surrounding every
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18 TaC grain, there is a shell of amorphous carbon. This occurs regardless of the location of the
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20 TaC grain, whether it is on the edge of a lamellar structure or in the center of it. Each particle is
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22 separated from other TaC grains by this amorphous layer of carbon. The existence of this layer
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24 indicates that grain diffusion dominates over volume diffusion.
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31 Further interpretation of the data allows us to suggest a more complete picture. We are
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33 able to propose a reaction mechanism that occurs in the solid combustion wave. This mechanism
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35 is wholly supported by the data and is non-trivial. As shown in Figure 5, the average thicknesses
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37 of the Ta and C lamellas after 4 min HEBM are 560 nm and 659 nm, respectively. From Figure
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39 8, we also can see that the average TaC grain size in the reaction product is much smaller than
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41 the lamella. The TaC grains, instead, are similar in size to the Ta crystal grains in the bulk of the
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43 metal lamella within the initial composite particle. Finally, TEM images reveal that carbon is
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45 located around the formed TaC grains. These three observations allow us to conclude that the
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47 first stage of the reaction involves carbon diffusing along the Ta crystal grain boundaries,
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49 throughout the volume of the metal lamella.
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55 Next, as can be seen in Figure 8, the average size of the 6 min HEBM lamellas is smaller
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57 than the 4 min HEBM case. Under identical heating conditions, a reaction could not be initiated
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4 in the 6 min HEBM sample. XRD and TEM analysis shows that the main qualitative distinction
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6 between the particles fabricated under the two milling times is that, after 6 min of mechanical
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8 treatment, the carbon solid solution has already been formed in Ta lamella.
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11 Table 1 shows how the lattice parameter of the crystal changes with increased milling
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13 time. This implies that there is an amount of carbon being dissolved in the Ta grains. Based on
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15 this result, we may conclude that the formation of a solid solution is an important part of the
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17 combustion mechanism. Solid solution formation is an exothermic process and, in accordance to
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19 the equilibrium phase diagram (Figure 1), up to 7 at. % of carbon can be dissolved in metal.
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21 Thermodynamic estimations show that this may lead to a significant temperature increase of
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23 approximately ~ 400 K under adiabatic conditions.
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29 The dissolution of carbon in tantalum is relatively fast; the reported carbon diffusion
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31 coefficient through Ta at ~1300 K (initiation temperature; see Fig 9) ranges from $\sim 10^{-7}$ [27] to
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33 10^{-9} [26] cm²/s. It means that, with a characteristic time of 0.4 s, carbon can diffuse from of ~500-
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35 5000 nm, which is well above the average Ta grain size. It was experimentally shown that carbon
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37 diffuses in these distances through Ta wires [30]. It appears that this rapid release of energy
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39 converts the reaction into the self-sustained thermal explosion mode [1]. If this stage is
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41 eliminated (by longer HEBM times), the slower reaction, owing to diffusion of carbon through
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43 the formed tantalum carbide layer (Fig.9), does not allow for the self-sustained mode of reaction
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45 to occur at this relatively low temperature. In order to initiate the combustion wave in the 6 min
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47 HEBM, the temperature must be significantly increased (>1800 K) for initiation. This illustrates
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49 the importance of carbon dissolution into the Ta crystals; this feature is paramount to initiation at
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51 low temperatures owing to the difference in diffusion coefficients between Ta and TaC.
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Furthermore, because the final product consists of spherical TaC grains with sizes comparable to the initial Ta crystals, it is logical to suggest that, at this stage, the reaction takes place via carbon diffusing through the tantalum carbide layer immediately following solid solution formation. This process being relatively slow (Fig. 9), but still requires only 0.16 s to diffuse on the length of the radius of the Ta grains in the mechanically induced nanostructured media.

The above mechanism is schematically summarized on Fig, 10. The reaction occurs, first, by surface diffusion leading to the intimate coverage of the Ta grains by C. Then is followed by volume diffusion of C into the Ta grain, leading to the formation of a solid solution within the crystal. A thin layer of TaC forms on the outer surface of the Ta crystal. Carbon diffuses through the TaC layer, into the unreacted Ta+C solid solution, and reacts, thickening the TaC layer until the solid solution is completely converted into pure TaC. This mechanism also directly explains why the pre- and post-combusted samples have the same external particle morphology (Fig. 5) and internal nanocrystal size and shape (Fig. 8).

Conclusion

By ensuring that the reactants were sufficiently free of any contaminants that would lead to the formation of low temperature eutectics and conducting all experiments in a truly inert atmosphere, it has been shown that the solid-flame phenomenon definitively occurs. The solid-flame phenomenon was made possible by use of advanced processing techniques, specifically HEBM, which created oxygen-free composite particles with significantly enhanced reactivity. Ignition experiments have shown that this reaction initiates at 1281 K, and consequently propagates at a rate of 0.7 cm/s, forming TaC completely within the solid phase without any liquid or gas-phase transport occurring. This reaction was further studied using XRD, SEM, and

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4 TEM analysis of the samples at differing milling times, in addition to after reaction initiation, to
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6 determine the reaction mechanism. The internal nanostructure was investigated and analyzed
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8 using 3D reconstruction techniques to ensure that the solid-state diffusion mechanism is
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10 theoretically valid, based on the thermal diffusivity of the mixture in conjunction with the mass
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12 diffusivity.
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16 Furthermore, this study illustrates how significant solid solution formation is for
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18 reactivity. In the 4 min DM sample, T_{ig} was multiple hundreds of degrees lower than the 6 min
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20 DM sample, even considering that the 6 min DM sample has lower diffusive distances and
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22 significantly increased surface area contact between the reactants. This parallel can be extended
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24 to other systems. For systems where a solid solution exists between the reactants, such as in the
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26 Ni-Al system, combustion is comparatively easier to initiate than in systems, like Si-C, that do
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28 not have solid solutions. In exploratory systems, the presence of a high reaction ΔH coupled with
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30 a solid solution may indicate a system that can undergo a combustion reaction more readily than
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32 just a high enthalpy of reaction.
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42
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44
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Figure Caption 1: Phase diagram of the Ta+C system. The adiabatic temperature is indicated. The maximum possible temperature is well below any eutectic or phase transition point.

Figure Caption 2: SEM images of the initial reactant morphology. The tantalum is plate-like and flat, between 5-40 μm . The carbon is amorphous and roughly spherical, consisting of large agglomerations of particles, individually under 2 μm .

Figure Caption 3: SEM images showing contact between reactants. In the unmilled samples, only point contact occurs. In the dry milled (DM) samples, there is intimate contact between the Ta and C, additionally, after combustion synthesis (CS) the particle size and shape remains unchanged.

Figure Caption 4: XRD patterns of the initial Ta+C composite, Ta/C composites for each milling time, and the combustion products.

Figure Caption 5: a) Sample cross section of the 4min DM Ta/C composite particle b) AvizoFire 3D image of analyzed Ta volume, C is voidspace. c) Computed Ta and C layer thickness distributions using distance-ordered thinning.

Figure Caption 6: Typical time-temperature profile used to define ignition temperature, with temperature derivative inset.

Figure Caption 7: Set of IR images of the combustion front propagation for the sample compacted from 4 min DM Ta/C composite particles, the emissivity is set to 0.6 for these images.

Figure Caption 8: TEM images showing the composite particle nanostructure. The diffraction insets show the crystallinity of the samples. As the milling time is increased, TaC crystals simultaneously become apparent, while the Ta crystal size decreases.

Figure Caption 9: Mass diffusivity data for the diffusion of C through Ta and TaC [24-29].

Figure Caption 10: Mechanism of the Ta+C reaction. First the carbon diffuses along the grain boundaries. This leads to a solid solution of Ta+C forming along the boundaries. Then volume diffusion occurs, causing the entire Ta crystals to be solid solutions of Ta+C. The heat released from the solid solution forming leads to the formation of TaC.

Table Caption 1: Lattice Constant ($\text{\AA}$) calculated from XRD patterns.

Table1

Time (min)	Lattice Constant (Å)
0	3.30
4	3.30
6	3.31
12	3.33

Figure1

Figure2

Figure3

Figure4

Figure5

Figure6

Figure7

Figure8

Figure9
[Click here to download Figure: Figure9.pdf](#)

Figure10