

Review

Laser surface alloying: a bibliography

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Laser alloying is a material processing method which utilizes the high power density available from focused laser sources to melt metal coatings and a portion of the underlying substrate. Since the melting occurs in a very short time and only at the surface, the bulk of the material remains cool, thus serving as an intimate heat sink. Large temperature gradients exist across the boundary between the melted surface region and the underlying solid substrate. The result is rapid self-quenching and resolidification. Quench rates as great as 10^{11} K sec⁻¹ and concomitant resolidification velocities of 20 m sec⁻¹ have already been realized. What makes laser surface alloying both attractive and interesting is the wide variety of chemical and microstructural states that can be retained because of the rapid quench from the liquid phase. These include chemical profiles where the "alloyed" element is highly concentrated near the atomic surface and decreases in concentration over shallow depths (hundreds of nanometers), and uniform profiles where the concentration is the same throughout the entire melted region. The types of microstructures observed include extended solid solutions, metastable crystalline phases, and metallic glasses. This bibliography is a compilation of published work in this area.

1. Introduction

The use of laser sources to produce metal surface alloys has become a materials processing application receiving considerable attention. The concept of laser surface alloying (LSA) is schematically pictured in the sequential cross-section views in Fig. 1. In Fig. 1a the metal substrate (B) coated with a thin metal film (A) is irradiated with a laser pulse. A fraction of the incident laser light is absorbed by free carriers within the electromagnetic skin depth (10^{-6} to 10^{-5} cm). For metal surfaces and most laser wavelengths a significant fraction of the incident light will be specularly or diffusely scattered away (the reflectance process is not shown in the illustration). The absorbed energy is "instantaneously" (10^{-12} sec) transferred to the lattice. The near surface region very rapidly reaches the melting point and a liquid/solid interface starts to move (solid arrows) through the film as shown in Fig. 1b. In Fig. 1c the liquid/solid interface has swept through the original thin film/substrate interface. Interdiffusion of the film (solid circles) and substrate (open

circles) elements starts. The laser pulse is nearly terminated and the surface has remained below the vapourization temperature. In Fig. 1d the maximum melt depth has been reached. Interdiffusion continues. The resolidification interface velocity is momentarily zero and then rapidly increases. The resolidification interface then moves approximately half way back to the surface from the melt depth (Fig. 1c). Interdiffusion in the liquid continues but the resolidified metal behind the liquid/solid interface cools so rapidly that solid state diffusion may be neglected. In Fig. 1f the material is completely resolidified and a "surface alloy" of A in B has been produced. What makes laser surface alloying both attractive and interesting is the wide variety of chemical and microstructural states that can be retained because of the rapid quench from the liquid phase.

Fig. 2 is a bar graph of laser surface alloying (LSA) publications as a function of time. Much of the interest is rationalized from the standpoint of strategic element shortages and precious metal costs. All directed energy (laser, electron, and ion

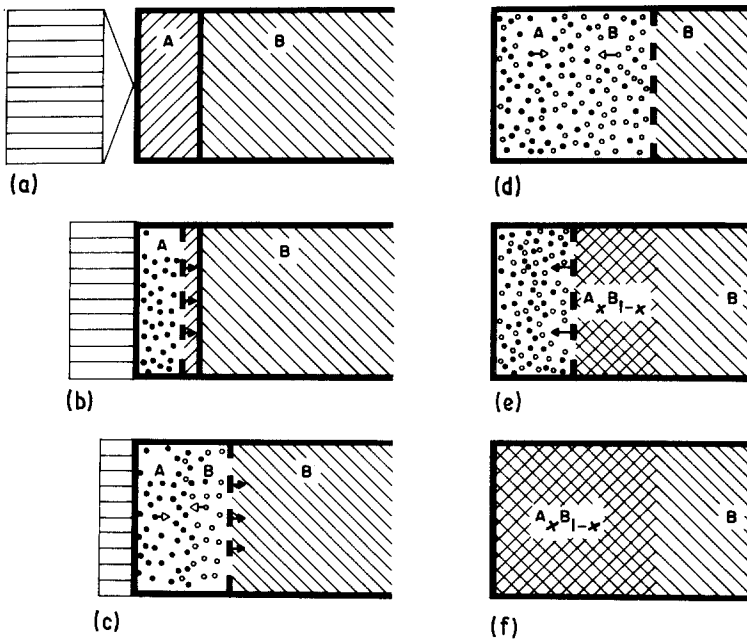


Figure 1 Sequential cross-section schematic illustration of mechanism of laser surface alloying from arrival of laser pulse to complete resolidification.

beam) methods of modifying surface composition are by their very nature materials conservation oriented. When an element is incorporated in a bulk alloy solely for the purpose of affecting the

alloys response to its interface environment, surface rather than bulk alloying is attractive.

Much of the more recent work explores the use of Q-switched laser sources to achieve high velocity regrowth (10 m sec^{-1}) of the molten metal interface. Extended solid solutions, metastable crystalline and amorphous surface alloys have all been formed by LSA processing. Other reasons for the growing interest are the wider availability of laser sources capable of energy fluences sufficient to melt metal surfaces and some spin-off from the immense push into semiconductor laser annealing.

Whatever the reason, it appears that there will continue to be significant growth in the literature on LSA, and that a timely updated bibliography [108] will aid interested parties in keeping abreast of current research activity.

In assembling Tables I and II, we have attempted to include all scientific literature that falls within the scope of laser surface alloying. Search methods included computer keyword and author searches of chemical, physics, metals, and laser abstracts. Written correspondence and telephone conversations with other active researchers in this field have been very helpful.

Table I is a chronological list of the laser surface alloying literature. No attempts have been made to order the literature on a finer scale than the year of publication. Table headings include the reference year, type of laser sources, alloying specie/host substrate, post-irradiation analysis of

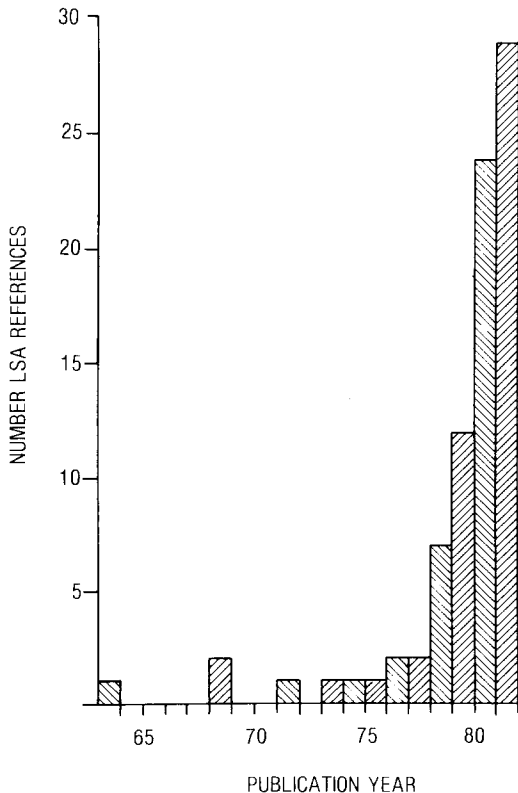


Figure 2 Growth in the LSA literature over the last 20 years.

TABLE I

Year	Laser	Alloying specie/substrate	Analysis	Reference
1964	P-Ruby	C, W, TiC, WC/St	COMP:EPM SSB:MH	[1]
1969	P-NdG	C/Fe	STRT:XRD SSB:MH	[2]
1969	P-Ruby	WC/St	STRT:OM COMP:EPM SSB:WR	[3]
1972	P-Ruby	C, WC/St	STRT:XRD SSB:MH	[4]
1974	P-NdG	C, W/Fe; Co/W; Al/Nb	STRT:OM, XRD	[5]
1975	CW-CO ₂	Cr, Ni, W, V, Mn, C/St	COMP:ANG SSB:MH	[6]
1976	P-NdG	Ni, Mo, Ti, Ta, Nb, V/Fe, St	SSB:MH	[7]
1977	P-NdG	Mo, Nb/Fe	COMP:ANG	[8]
1977	P-NdG	Mo/Fe	STRT:OM, XRD COMP:XRD SSB:MH	[9]
1978	P-NdG	V/Fe	STRT:OM, XRD COMP:XRD SSB:MH	[10]
1978	CW-CO ₂	CrC, WC/St	STRT:OM COMP:ANG SSB:MH	[11]
1979	CW-CO ₂	Cu/Ag-7 Cu	STRT:OM, SEM	[12]
1979	Q-Ruby	Cu/Al	STRT:CHN COMP:RBS	[13]
1979	CW-CO ₂	Cr, Ni/St	STRT:OM COMP:EPM, EDX	[14, 15]
1979	CW-CO ₂	Cr-C-Mn-Al/St	STRT:OM COMP:EPM SSB:MH	[16, 20]
1979	P-NdG	Fe/Nb	STRT:MOSB, XRD	[17]
1979	CW-CO ₂	Mo-Ni-Si-Cr-C/St	STRT:OM, XRD COMP:EDX, ANG SSB:MH, HTS, WR	[18]
1979	CW-CO ₂	Cr/St	STRT:OM, SEM COMP:EPM, EDX, AES	[19]
1980	CW-CO ₂ , Q-NdY	Au, Ag, Pg, Sn, Ta/Ti	STRT:OM, SEM, CHN COMP:RBS, EDX, AES, ESCA SSB:MH, COR	[21]
1980	CW-CO ₂	TiC, WC/Ti-6Al-4V, Inconel	STRT:OM, SEM	[22]
1980	Q-NdG	Sb/Al	STRT:OM, XRD COMP:RBS	[23]
1980	Q-Ruby	Ni/Fe/SiO ₂	STRT:TEM SSB:MAG, OPT	[24]
1980	CW-CO ₂	TiC, WC/St, Ti-6Al-4V, Inconel, 5052Al	STRT:OM SSB:MH	[25]
1980	P-NdY	Re/Fe-50 Ni	STRT:OM, SEM COMP:EPM	[26]
1980	Q-NdY	Au, Ag, Ta/Ni	STRT:OM, CHN COMP:RBS	[27]
1980	Q-dNdY	Pb/Cu	STRT:CHN COMP:RBS	[28]
1980	CW-CO ₂	TiC/304 St	STRT:SEM SSB:MH	[29]
1980	Q-Ruby	Cu, Pb/Al	COMP:RBS	[30]
1980	CW-CO ₂	B, FeB/St	STRT:OM, SEM, TEM SSB:MH, HTS	[31]

TABLE I Continued

Year	Laser	Alloying specie/substrate	Analysis	Reference
1980	CW-CO ₂	Cr/4815 St; Ni, Cr, Si/Al	STRT:OM	[32]
1981	Q-Ruby	VC, WC/St; TiC/Ti; Ni-CrAl/St	COMP:ANG	
		Zn, Sb/Al	STRT:TEM, CHN	[33]
			COMP:RBS	
1981	Q-NdY	Au, Ag, Pd, Ta, Sn/Ni	STRT:TEM, CHN	[34]
			COMP:RBS	
1981	Q-Ruby	Ni/Au/Ni	STRT:TEM	[35]
			COMP:RBS	
1981	Q-Ruby	Mo, Cd/Al	STRT:CHN	[36]
			COMP:RBS	
1981	Q-NdY	Hf/Ni	STRT:CHN, PAC	[37]
			COMP:RBS, AES	
1981	Q-NdY	Pd/Ti	COMP:RBS	[38]
			SSB:COR	
1981	CW-CO ₂	TiC, WC, WC-Co/St	COMP:EDAX	[39]
			STRT:SEM	
			SSB:MH	
1981	CW-CO ₂	Cr, Cr-Co/St	STRT:OM	[40]
			SSB:MH	
1981	CW-CO ₂ , Q-NdY,	Au/Ni	STRT:CHN	[41]
	Q-dNdY, Q-Ruby		COMP:RBS	
1981	CW-CO ₂ , Q-NdY,	Au/Ni	STRT:CHN	[42]
	Q-dNdY		COMP:RBS	
1981	Q-Ruby	Cr, Sb/Al	THRY:HF, MT	[43]
1981	Q-dNdY	Ag/Ni	STRT:CHN	[44]
			COMP:RBS	
1981	CW-CO ₂	TiC/Ti-6Al-4V, 304 St, 4340 St, 5052 Al, Al-bronze	STRT:OM, SEM	[45]
1981	CW-CO ₂	Si, TiC/5052 Al	STRT:OM, SEM	[46]
1981	CW-CO ₂	Fe-B/Fe-12Cr-2C, Ni ₆₀ Nb ₄₀ /Nb	STRT:TEM, OM	[47]
			SSB:MH, HTS	
1981	CW-CO ₂	Cr/St	SSB:COR	[48]
1981	Q-NdG	Cr/Al	COMP:RBS, SEM	[49]
1981	CW-CO ₂	Cr, Cr-Ni/St	COMP:ANG	[50]
		Cr-W-TiC/St	SSB:MH	
1981	CW-CO ₂	TiC, WC/Ti-6Al-4V, 304 St, 5052 Al	STRT:SEM	[51]
			SSB:MH, WR	
1981	Q-Ruby	In/Al	STRT:PAC	[52]
1981	CW-CO ₂	Cr/Fe	COMP:EPM	[53]
			STRT:OM	
1981	Q-NdG	Sb/Al	COMP:RBS, SEM	[54, 55]
1982	Q-Ruby	Zn/Al, Au/Ni	THRY:MT	[56]
1982	Q-NdG	Pb/Al	COMP:RBS, SEM	[57]
1982	Q-Ruby	Cu/Al	Soret coeff.	[58]
1982	Q-Ruby	V-Si/sapphire	COMP:RBS	[59]
			STRT:XRD	
			SSB:SC	
1982	Q-Ruby	Cr/Al	COMP:RBS	[60]
			STRT:CHN, SEM	
1982	Q-Ruby	Cr, Mo/Al	COMP:RBS	[61]
			STRT:CHN	
			SSB:HTS	
1982	Q-Ruby	Eu, Ni/Ni	COMP:RBS	[62]
			STRT:CHN, SEM	
1982	Q-Ruby	Sb/Al	STRT:CHN, TEM	[63]
1982	Q-dNdY	Cr, Ni/Cu	COMP:RBS, AES	[64]
			STRT:CHN, TEM, XRD	
			SSB:COR	
1982	Q-dNdY	Zr/Ni, Al, Fe, Ti, V Cu/Zr	COMP:RBS	[65]
			STRT:TEM, CHN, XRD	

TABLE I Continued

Year	Laser	Alloying specie/substrate	Analysis	Reference
1982	Q-Ruby	Cu, Zn/Al	THR Y:MT	[66]
1982	CW-CO ₂	Au/Ni WC-Co, WC-Co-Mo, WC-Ni-Cr-B-Si/St	COMP: ANG STRT:OM, SEM SSB:MH	[67]
1982	CW-CO ₂	Fe-B/St	STRT:XRD, TEM	[68]
1982	CW-CO ₂	Cr/Fe-0.2C	COMP:EDAX STRT:OM, TEM	[69]
1982	CW-CO ₂	FeTi, FeCr, FeCb, FeV, FeSi/St	STRT:OM, SEM	[70]
1982	CW-CO ₂	8Al-12Mo-3Ta-Ni 12Al-15Mo-Ni	COMP: ANG STRT:TEM SSB:TNS	[71]
1982	CW-CO ₂	Mo/St	COMP:EDAX STRT:SEM SSB:MH	[72]
1982	CW-CO ₂	Ni/Fe	COMP:EPM THR Y:MT	[73]
1982	Q-NdG	Sn/Al, W/V, Pb/Fe, W/Cr	COMP:RBS STRT:CHN	[74]
1982	CW-CO ₂	Cr/Fe	COMP:EPM STRT:SEM	[75]
1982	Q-NdG	Pb/Al	COMP:RBS STRT:SEM	[76]
1982	ML-NdY	Fe ₃ B/Fe	STRT:TEM	[77]
1982	CW-CO ₂	Al, Cr, Ni/Fe	COMP:EPM THR Y:MT	[78]
1982	CW-CO ₂	Ni/Fe	COMP:EDAX STRT:TEM, OM	[79]
1982	CW-CO ₂	Cr/St	COMP:EDAX STRT:TEM, SEM SSB:MH	[80]
1982	CW-CO ₂	Cr-Ni/4140 St	COMP:EPM SSB:COR	[81]
1983	Q-Ruby	Cu/Au/Co Cu/W/Cu	COMP:RBS	[82]
1983	Q-NdY	Hf/Ni	COMP:RBS STRT:PAC, CHN	[83]
1983	CW-CO ₂	Mo/6150 St	COMP:EDAX STRT:SEM, TEM SSB:MH	[84]
1983	CW-CO ₂	Ni/1020 St Cr/1018 St	THR Y:MT COMP:EPM	[85, 88]
1983	CW-CO ₂	Cr, Ni/Fe; Cr/Ni/Fe	COMP:EDAX STRT:OM, SEM, TEM	[86]
1983	CW-CO ₂	Cr/Fe	COMP:EDAX STRT:OM, SEM, TEM SSB:MH	[87]
1983	ML-NdY	Fe ₃ B/Fe	STRT:TEM	[89]
1983	P-CO ₂	Ar, N ₂ , CO ₂ , C ₂ H ₄ /St	STRT:OM SSB:MH	[90]
1983	Q-Ruby	N, C/Mo	COMP:RBS SSB:SC	[91]
1983	Q-NdG	Ni ⁶² /Ni, Fe ⁵⁵ -Fe ⁵⁹ /Fe	COMP:RI THR Y:MT	[92]
1983	CW-CO ₂	Cr/St	STRT:OM, SEM, TEM SSB:MH	[93]
1983	CW-CO ₂	Cr, WC-10Co/St	STRT:OM SSB:MH	[94]
1983	CW-CO ₂	TiC/6061 Al	STRT:SEM SSB:WR	[95]

TABLE I Continued

Year	Laser	Alloying specie/substrate	Analysis	Reference
1983	EB	Ag/Al, Mn/Al, As/Si	THRY:MT	[96]
1983	ML-NdY	Fe ₃ B/Fe	STRT:TEM	[97]
1984	ML-NdY	Fe ₃ B/Fe, Ni/Nb	STRT:TEM	[98]
1984	CW-CO ₂	Mo/Ni, Mo/Co, Nb/Co	COMP:EDAX	[99]
1984	Q-Ruby	Cr/St	STRT:SEM, TEM	
1984	Q-Ruby	La, Eu/Ni	COMP:RBS	[100]
1984	Q-Ruby	Pb/Ni	STRT:CHN	
1984	Q-Ruby	Au, Co, Cr, Zr/Ti	COMP:RBS	[101]
1984	Q-dNdY	Cu/Zr	STRT:SEM	
1984	Q-dNdY	Ni-8V/Cr/Cu-2Sn	STRT:XRD	[102]
1984	CW-CO ₂	Cr/St	STRT:XRD, TEM	[103]
			COMP:AES, RBS	[104]
			COMP:EDAX	[105]
			STRT:OM	
			SSB:MH	

structure, composition or modification of surface sensitive behaviour, and finally the numerical reference. Abbreviations used are explained in Table II. Take for example the 1978 Kovalenko work [10]; samples of iron coated with vanadium were alloyed with a pulsed neodymium:glass laser. Alloyed samples were analysed structurally using optical microscopy and X-ray diffraction. Compositions are given based on lattice parameter changes in the X-ray diffraction patterns. The microhardness of the irradiated sample is reported.

Table I does not provide insight into how the alloying elements were put down prior to laser irradiation. The methods used are numerous and

include vacuum evaporation, electroplating, electroless plating, powder coatings, thin-foil application, and ion implantation. In some cases the element is injected into the melt region in the form of particles or wire. These last two methods of introducing the alloying specie are the most attractive from a commercial standpoint. When coupled with repetitive laser scanning it has been demonstrated that one can build up the surface alloyed region. This is the basis of the United Technologies effort in a commercial processing method called "layer glazing". A sizable body of that work has been summarized [112, 113].

There have been several short review type articles which may be useful to the reader interested

TABLE II Abbreviations used in Table I

Laser

P, pulsed

CW, continuous

Q, Q-switched

ML, mode locked

CO₂, carbon dioxide

NdY, neodymium yttrium aluminium garnet

NdG, neodymium glass

dNdY, frequency doubled NdY

Ruby, chromium doped aluminium oxide

EM, electron beam

EX, excimer

Alloying specie/substrate – elemental symbols

St, steel, cast irons, general designation for Fe alloy substrates

Commas (,), denote individual experiments

Dashes (-), denote a mixture of elements

Semicolon (;), denote individual experiments (added clarity)

For example, C, WC/St denotes carbon film and tungsten carbide film on a steel substrate

Re/Fe-50Ni denotes a rhenium film on an iron-50 wt % nickel alloy substrate

Analysis

STRT, structure
SEM, scanning electron microscopy
TEM, transmission electron microscopy
OM, optical microscopy
XRD, X-ray diffraction
CHN, channelling
MOSB, Mössbauer spectroscopy
PAC, perturbed angle correlation
COMP, composition RBS, Rutherford backscattering spectroscopy
EPM, electron probe microanalysis
EDAX, energy or wavelength dispersive X-ray, elemental mapping
ESCA, electron spectroscopy for chemical analysis
AES, Auger electron spectroscopy
RI, radioactive isotope analysis
SSB, surface sensitive behaviour
MH, microhardness
COR, corrosion
WR, wear
TNS, tensile properties
HTS, high temperature stability
SC, superconductivity measurements
MAG, magnetic
OPT, optical
ANG, results quoted by analytical method not given
THRY, theory
MT, mass transport
HF, heat flow

in more details on the general topic, mechanisms and methodology of laser surface alloying. These are listed at the end of the Bibliography under [106–111].

We hope that this bibliography will serve several purposes. The first is to assemble in one place references of work in this area of research. The manner in which Table I has been constructed allows one to quickly extract the alloying elements, substrates, laser sources, and analyses performed for each reference. It is hoped that the authors will be made aware of both references missed and those forthcoming in order to be able to update the tabulated material presented here.

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