Laser and Particle Beams (2006), *24*, 203–209. Printed in the USA.

DOI: 10.1017/S0263034606060289

Laser–induced film deposition by LIFT: Physical mechanisms and applications

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(RECEIVED 24 July 2005; ACCEPTED 18 January 2006)

Abstract

Peculiarities of the technique of the laser-induced film transfer (LIFT) are investigated. Possible mechanisms of tearing-off and transference of the films from the donor substrate (target) to the acceptor one are investigated. The main fields of LIFT applications are considered. One of the most interesting directions of LIFT applications—decontamination of radioactive surfaces—is investigated in detail. The main peculiarities and regimes of the processing are defined.

Keywords: Ablation; Deactivation; Film; Substrate; Transfer

1. INTRODUCTION

Fusion, especially inertial fusion is widely regarded as an important field to apply plasma physics methods. Especially in the direct driven scenarios, the surface condition of the target plays an important role (Canaud *et al*., 2004). The fast ignition of fusion pellets with superintense lasers (Mulser & Bauer, 2004) scenario depends on the acceleration mechanism of intense lasers with a solid surface. In many cases, the surface condition is controlled by laser ablation (Fernandez *et al*., 2005) or laser, and electrochemical methods (Gavrilov *et al*., 2004). Furthermore, the development of instabilities during the compression phase depends strongly on the target surface conditions (Fincke *et al*., 2005).

In this paper, we discuss the laser–induced film transfer (LIFT) method which was suggested some years ago (Veiko *et al*., 1970). This method is now commonly used for direct "writing" in microelectronic components, data storage devices, displays, etc. (Szörényi & Tóth, 1989; Fitz-Gerald *et al*., 2000; Lu & Ren, 2001; Piqué *et al*., 2003). Physical ideas of LIFT method have recently appeared to be advantageous also in laser cleaning of surfaces (Luk'yanchuk, 2002), up-to-date and perspective technologies. Laser cleaning of surfaces from particles and surface layers can be realized by laser heating and thermal expansion of the contaminations or evaporation. The method can be supplemented by collection of ablated material to transparent film covering the surface. In this case, processes of ablation and condensation of surface layer material correspond to LIFT physical processes. Application of this method for decontamination of radioactive surfaces is proposed in this paper.

The main variants of LIFT method employ either front or back transfer of donor material (Veiko & Metev, 1995; Mogyorósi *et al*., 1989) (Fig. 1). In the first case (Fig. 1a), laser beam is focused at donor film through its transparent substrate (DS); the film material is transferred to acceptor substrate (AS) in the form of melt or vapor or plasma, and condenses on the substrate surface. With back transfer (Fig. 1b), laser beam passes through transparent acceptor substrate to focus at the donor film free surface. The back transfer can be applied for opaque donor substrate as well as for bulk material as a donor.

According to experiments performed (see, e.g., Veiko *et al*., 1993), the following LIFT regimes may be distinguished depending on the laser intensity q:

- $q < q_{th}$—low-intensity regime, at which solid or melted film fragments are transferred to acceptor surface demonstrating a relatively weak adhesion (q_{th} is the threshold intensity corresponding to the boiling temperature of the donor film material at the normal pressure),

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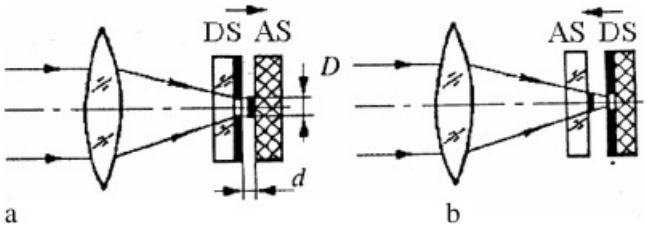

Fig. 1. Main variants of LIFT method: (**a**) front transfer, (**b**) back transfer (AS is the acceptor substrate and DS is the donor substrate).

- $q_{th} < q < 5q_{th}$—regime of "soft" evaporation of the film followed by condensation of the film material at the acceptor substrate (the evaporation LIFT regime),
- $5q_{th} < q < 10q_{th}$—regime of laser-induced ions and atoms implantation into the substrate,
- $q > 10q_{th}$—regime of deep implantation of the plasma products of film destruction into the AS resulting in considerable mechanical tensions in the substrate.

The regime $q_{th} < q < 5q_{th}$ is commonly used in direct writing processes to provide a high-quality deposition of transferred material.

When the LIFT methods are applied for laser cleaning of solid surface with collection of removed contamination onto an adjacent acceptor surface, two of the above-mentioned regimes prove to be effective: $q < q_{th}$ (the regime corresponds to low-intensity cleaning), and $q_{th} < q < 5q_{th}$ (corresponds to evaporating cleaning) (Luk'yanchuk, 2002).

In what follows, physical mechanisms of LIFT are discussed, and a new example of LIFT ideas application to surface cleaning is presented.

2. PHYSICAL MODELS OF LIFT

2.1. Low-intensity mechanisms

Two possible causes for a film particle to be torn away from donor substrate surface are (Veiko *et al.*, 1993, 2002): (a) formation of a cavity between the film and the DS with a surplus pressure inside it (blasting mechanism; see Fig. 2a) and (b) thermo-mechanical processes in the film–DS system (Fig. 2b).

Blasting mechanism of the film tearing-off may be actuated by one of three following processes or their combination:

1. With a thermo–unstable DS (polymer, weak glass), gasification of the DS substance may take place. The donor film destruction by pressure of evaporation products of the donor substrate occurs if the evaporation (decomposition) temperature of the DS material is low enough. Solid film removal is possible if the gasification pressure p_g ($p_g = 0.5p_{sat}$, p_{sat} being the saturated vapor pressure) is larger than the pressure of the adhesion force.

2. For a thermo-stable DS (quartz glass, glass-ceramics), a surplus pressure may be a result of thermo-desorption of gases adsorbed by the DS surface defects. The desorbed gas pressure may be estimated at $p_d = kT/\sigma_0 r$, where k is the Boltzmann constant, σ_0 is the cross section of the adsorbed gas molecule, and r is the defect size. A special case is film tearing-off resulting from evaporation of thermo–unstable internal layer between donor film and DS. This is just the mechanism employed in laser micro-packaging (Holmes, 2002).

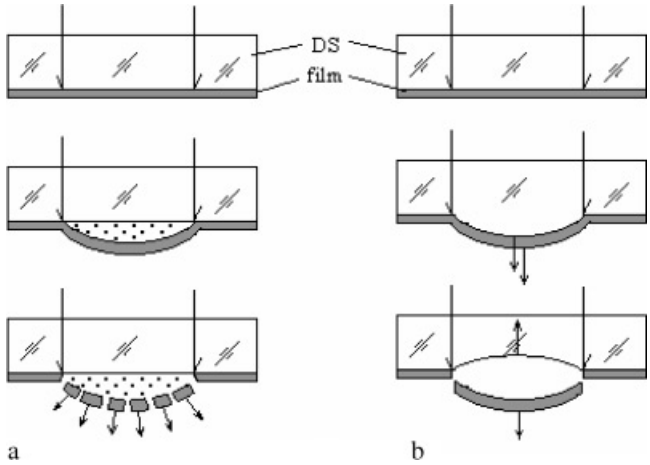

Fig. 2. Illustration of low-intensity mechanisms of film ablation: (**a**) blasting and (**b**) thermomechanical.

3. Film tearing-off can occur due to evaporation of the film material into a closed cavity at the film–DS interface. The pressure in the cavity is twice as large as that at the free film surface, and may cause an internal explosion in defects with sizes over a critical value r_{cr} in low intensity regimes of the LIFT if laser intensity is insufficient for film surface evaporation. As an example, $r_{cr} \approx 100$ nm for laser pulse duration $\tau \sim 10^{-8}$ s and film thickness $h \sim 100$ nm.

Thermo-mechanical film tearing-off mechanism may be induced by the following causes (Tóth *et al.*, 1999):

1. Film exfoliation may occur as a result of radial expansion of the film accounted for radial compression force, which causes transversal tension in the film. This force is normal to the DS surface, and film tearing-off happens if the force exceeds the adhesion threshold

$$F_{th} \leq \frac{Eh}{l}(\alpha_f \Delta T)^{3/2}, \quad (1)$$

where ΔT is the increase in film temperature, E is the elasticity module, l is the width of the defect on the DS surface, α_f is the thermal expansion coefficient, and h is the film thickness. For Al film at $\Delta T = 6 \cdot 10^2$ K (maximum temperature augmentation before melting), $h = 10^{-7}$ m, $l = 10^{-6}$ m, the adhesion threshold is $F_{th} = 4 \cdot 10^6$ Pa. Under these forces, the film fragment may be ablated and then fly away from the substrate with the velocity (before melting) (Tóth *et al.*, 1999)

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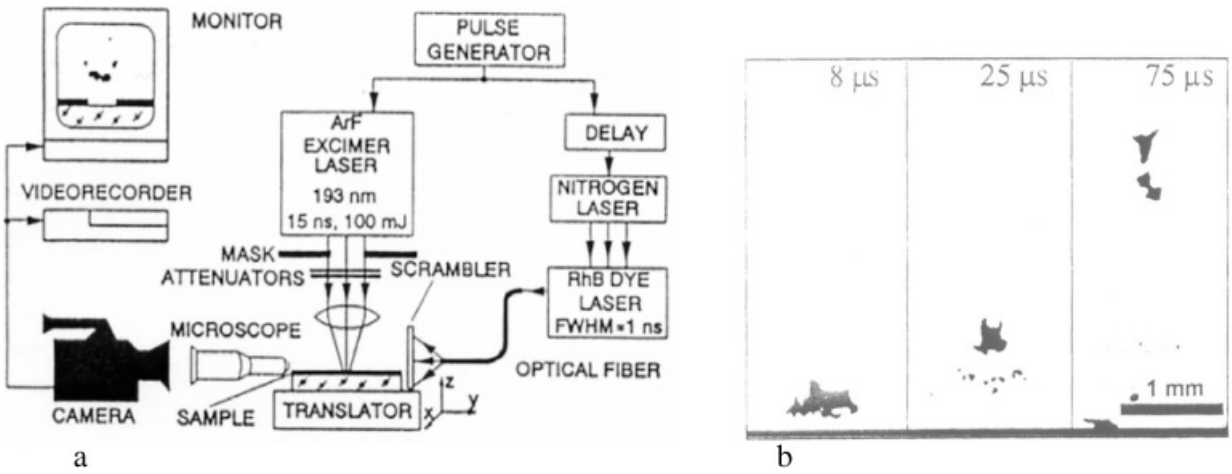

Fig. 3. Experimental study of ablation [17]: (**a**) experimental setup used for ablation process visualization and (**b**) ultra-fast images of separate solid pieces ablated by pulses of 70 mJ/cm^2 fluence at various time delays.

$$V = \frac{\alpha_f \varepsilon}{\rho c h}\sqrt{\frac{E}{\rho}} \tag{2}$$

(ε is the laser fluence, ρ is the film density, c and is the film heat capacity). Observation and measurement of big film fragments has been carried out (Tóth *et al.*, 1999) with the use of experimental setup presented in Figure 3. The light from operational excimer ArF laser (15 ns 100 mJ) is focused at the film sample by the lens. An independent visualizing optical system consisting of electronically delayed N_2-laser, pumped dye laser, the optical fiber, and the scrambler provides uniform pulsed illumination of the space above the sample. Images of film fragments ablated from the sample are obtained with a video system combined with a microscope. The ultra-fast photographs taken at fixed moments after the excimer laser pulse (Fig. 3b) show that film fragment ablated by thermo-mechanical mechanism (before evaporation) moves near the surface with a constant velocity, the velocity value may be derived from measured distances from the surface at the given time delays.

A typical curve of ablated film fragment velocity versus the operational laser light energy density at the film is presented in Figure 4. In regime 1, ablation of film fragment is caused by thermal expansion of the film during the pulse. In regime 2, film fragments tearing-off occurs before the pulse finishes, so that only a portion of the pulse energy is spent on film fragments movement. In regime 3, the main factors are the film expansion by melting and current of the melt.

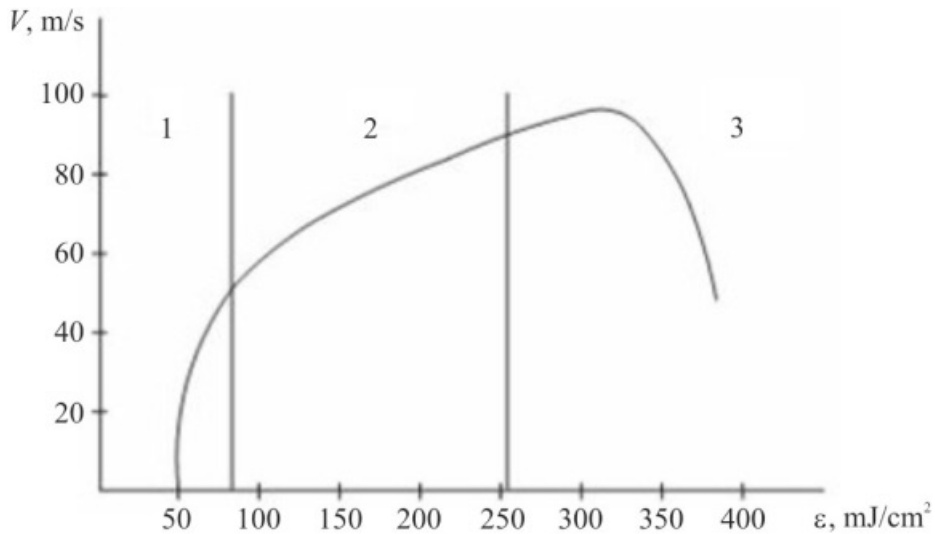

Fig. 4. A typical curve of film fragment velocity. Observed regimes of ablation: (**1**, **2**) film exfoliation is caused by its thermal expansion (tearing off occurs after (1) or before (2) pulse end), (**3**) film exfoliation is caused by expansion by melting.

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2. Film mass center displacement with its thermal expansion. Film tearing-off occurs if the specific inertia force exceeds the adhesion force. In this case,

$$F_{th} \le \rho h^2 \alpha_f \Delta T/\tau_m^2, \quad (3)$$

τ_m is the time of film heating up to the melting temperature. For Al film with the thickness of 10^{-7} m and $\tau_m \approx 2 \cdot 10^{-8}$ s, the threshold adhesion value is $F_{th} \approx 4 \times 10^2$ Pa. At the melting stage, F_{th} can be estimated by consideration of film expansion caused by melting.

3. Film tearing-off by means of shaking-off is caused by an inertial force arising from DS surface movement due to thermal expansion of heated layer x_0 ($x_0 \sim \sqrt{\alpha_{DS}\tau}$, a_{DS} is the thermal diffusivity of the DS). When compared with the effect of the above-mentioned film expansion, the relative magnitude χ of this effect is

$$\chi = \frac{x_0 \alpha_{DS} \Delta T_{DS}}{h \alpha_f \Delta T}, \quad (4)$$

where α_{DS} is the thermal expansion coefficient of the substrate, ΔT_{DS} is the average temperature augmentation of its layer x_0. For 10^{-7} m Al film on glass DS at $\tau = 2 \cdot 10^{-8}$ s, $\chi \approx 0.6$. Therefore, the effect related to substrate thermal expansion may be comparable to the effect of expansion of the film itself.

2.2. Evaporation front LIFT

If laser intensity q exceeds the threshold value q_{th}, the film temperature reaches and exceeds the boiling point T_b of donor material at normal pressure p_b. In this case, film tearing-off by the action of the blasting or/and the thermomechanical mechanisms are followed by evaporation from both sides of the film to chinks between the film and the both substrates (the DS and the AS), and there is partial vapor spreading outside the irradiated zone. During this process, vapor condensation at the substrate takes place inside and outside the irradiated zone. Film decreases in thickness and mass due to evaporation of donor material. This stage ends with complete evaporation of the film fragment or deposition of AS fragment. The difference between the initial film thickness (at the DS) and the deposited one (at the AS) is determined by evaporation of donor material during and after the transfer, and by the partial removal of the film material (vapor and melt) outside the irradiated zone. The processes of heating, evaporation, and hydrodynamical spreading of film melt layers transferred to the AS are analogous to ones described in (Veiko *et al*., 2002).

There are two causes of melt transfer to AS surface leading to crater-like deposit formation. First, if the melted film has not completely evaporated during its transfer from the DS to the acceptor one, it is flattened under the action of inertial forces after striking at the AS surface. The velocity of the melted deposit movement near the edges of deposited fragment may approach the speed of sound in the melt. This leads to non-linear spatial distribution of velocity in the melted material, accumulation of donor material along edges of the deposited fragment, and to formation of a crater-like structure. Another cause of melt redistribution at AS is vaporization of the condensate at the AS. The melt is forced out of irradiated zone under the action of vapor pressure and hardens in the form of a roller.

Melt movement from the condensate center to its periphery is also possible under the action of surface tension forces, if a pinhole is formed in the condensate layer and its diameter b exceeds the critical value b_{cr}. If the film is ideally adhesive to the AS, $b_{cr} = 2h$. The final diameter of the hole in the condensate is determined by correlation between the rate of the melt gathering at the periphery condensed fragment, and duration of the melt hardening.

Several peculiarities in the condensate structure (for example, formation of halo around the deposit at the AS) may be explained also by the consideration of gas flow in the chink-like channel between the two substrates.

The considered model of LIFT process makes it possible to explain and describe quantitatively a number of effects accompanying LIFT, including different mechanisms of low intensity transfer, crater and halo formation, and structural peculiarities in condensed material. Calculations carried out on the base of the proposed model provide a mean for optimization of LIFT technological parameters.

3. LIFT APPLICATIONS

The LIFT method extends the capabilities laser micromachining as compared with traditional approaches based on local laser evaporation of thin films. Combination of the LIFT method with the method of local laser evaporation (which can be carried out with the same setup due to similarity of corresponding physical processes) may build up universal laser micromachining processes.

The main applications of LIFT method encompass the following fields (Veiko *et al*., 2002):

- Correction of microelectronic component parameters.
- Saving of photo-masks and integral circuits.
- The process of an integral circuits or a photo-mask saving consists in removing excess fragments of the film by laser evaporation, and filling pinholes by the LIFT method. Effectiveness of the approach is confirmed by experiments (Veiko *et al*., 2002) carried out with Cr film covered photo-masks and with integral circuits of Cu wires.
- Direct film topology formation.

One of the typical problems of this kind is formation of resistors. High film adhesion to the substrate, sufficient resolution (down to 5 μm), and electrical resistance lying within the required interval are necessary for resistor formation. In experiments (Veiko *et al*., 2002) formation of various elements in the form of strips 10–50 μm in width was

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carried out by the LIFT method with different films and substrates: Cr and Cr/Cu/Cr films on textolite substrates; Cr, Cr/Cu/Cr, Al and V films on polypore, glass and glass-ceramics substrates and on a SiO_2 layer covering a Si plate; and PbS films on glass-ceramics, glass, polypore, and textolite substrates.

- Decorative drawing deposition.
  Experiments on decorative drawing deposition were carried out with YAG:Nd laser and computer-operated mirror scanner. Patterns of rastered portraits and outlined drawings on glass, glass-ceramics, article stones and jewelry stones were obtained.
- Optical recording.

The important problem of laser synthesis of optical image is to obtain not only perfect contrast elements, but also half-tints. It seems to be too difficult to solve this problem by means of deposition and subsequent local removing of a film. The LIFT method provides an easy way to do it by creation of various details of the image from deposited materials with different optical properties. Metal oxide deposits can be obtained if film transfer is carried out in oxygen environment.

4. DECONTAMINATION OF RADIOACTIVE SURFACES

One of the most important problems is recycling of equipment, contaminated with radioactive products during the process of nuclear fuel cycle. Nowadays a considerable quantity of spent equipment made of expensive materials is kept at atomic stations. The equipment is stored at specially conditioned sites that considerably increase ecological pressure on environment and expenses for safety. Investigations carried out make it quite clear that radioactive contaminations are largely accumulated in thin oxide film covering inner surfaces of equipment which contact with cooling agent—water, for instance. The films are composed predominantly of products of corrosion, compounds of calcium, and other elements with radioactive inclusions. The films are formed by sedimentation of radioactive products from cooling fluid during the process of corrosion.

As a rule, the film has two-layer structure formed by a dense thin solid layer adjacent to the metal base, and a porous external one. The external layer is of utmost importance in forming and accumulation of radioactive sources because of its high absorbing capability. Besides, cooling agent penetrates into the porous layer, dissolves and washes out soluble fractions of corrosion products and replaces them with insoluble compounds.

Nowadays the most commonly used methods are chemical and electrochemical ones. Their disadvantage is a large quantity of liquid radioactive waste products, the products processing being very labor consuming and expensive. On the contrary, laser-based deactivation obviates the employment of chemical reagents. Application of nanosecond pulses of AIG-Nd laser makes it possible to remove surface corrosion films up to 200 μm thick without melting or evaporation of base material (Nikishin & Smirnov, 2001; Veiko *et al*., 2001). The authors proposed a laser-based deactivation method on the base of LIFT with laser beam operating through a transparent (at 1.06 μm) polymer film, the film separating "dirty" operation zone from "clear" one and accumulating deactivation products due to a glue deposited on the "dirty" film surface (Fig. 5). With this method, wastes are utilized along with the film. .

An investigation of mechanisms of oxide film material transfer from metal surface and its sorption was carried out in (Smirnov *et al*., 2005) with the use of 1.06 μm 10 ns Nd:YAG laser and 20 $\times$ 20 mm corroded steel plates as

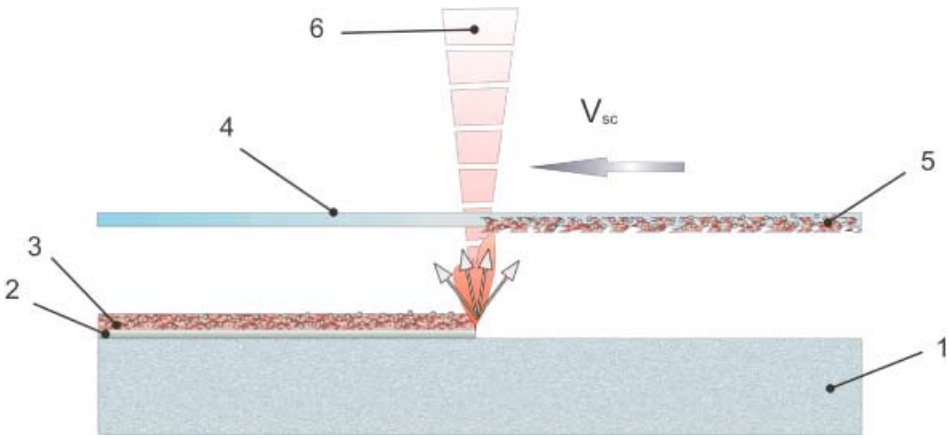

Fig. 5. Structure of radioactive contamination of a surface and schematic of laser cleaning of the contaminated surface: (**1**) base material, (**2**) dense thin oxide layer, (**3**) corroded porous thick layer, (**4**) transparent film collector, (**5**) ablated and collected products (radioactive contaminates), (**6**) scanning laser beam.

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samples. The experimental schematics of LIFT process and transferred material collection at a transparent film situated near by the contaminated surface corresponded to Figure 5. Difference in sample mass before and after processing let define the ablated layer thickness. Results of the experiments presented in Figure 6 show that there are two important regimes:

- $q < q_{th}$ ($q < 170$ MWt/cm^2)—thermo-mechanical ablation, with characteristic sound, and
- $q > q_{th}$ ($q > 170$ MWt/cm^2)—evaporation ablation.

Our investigations (Nikishin & Smirnov, 2001; Smirnov *et al.*, 2005) demonstrated that laser processing of contaminated metal samples resulted in considerable reduction in

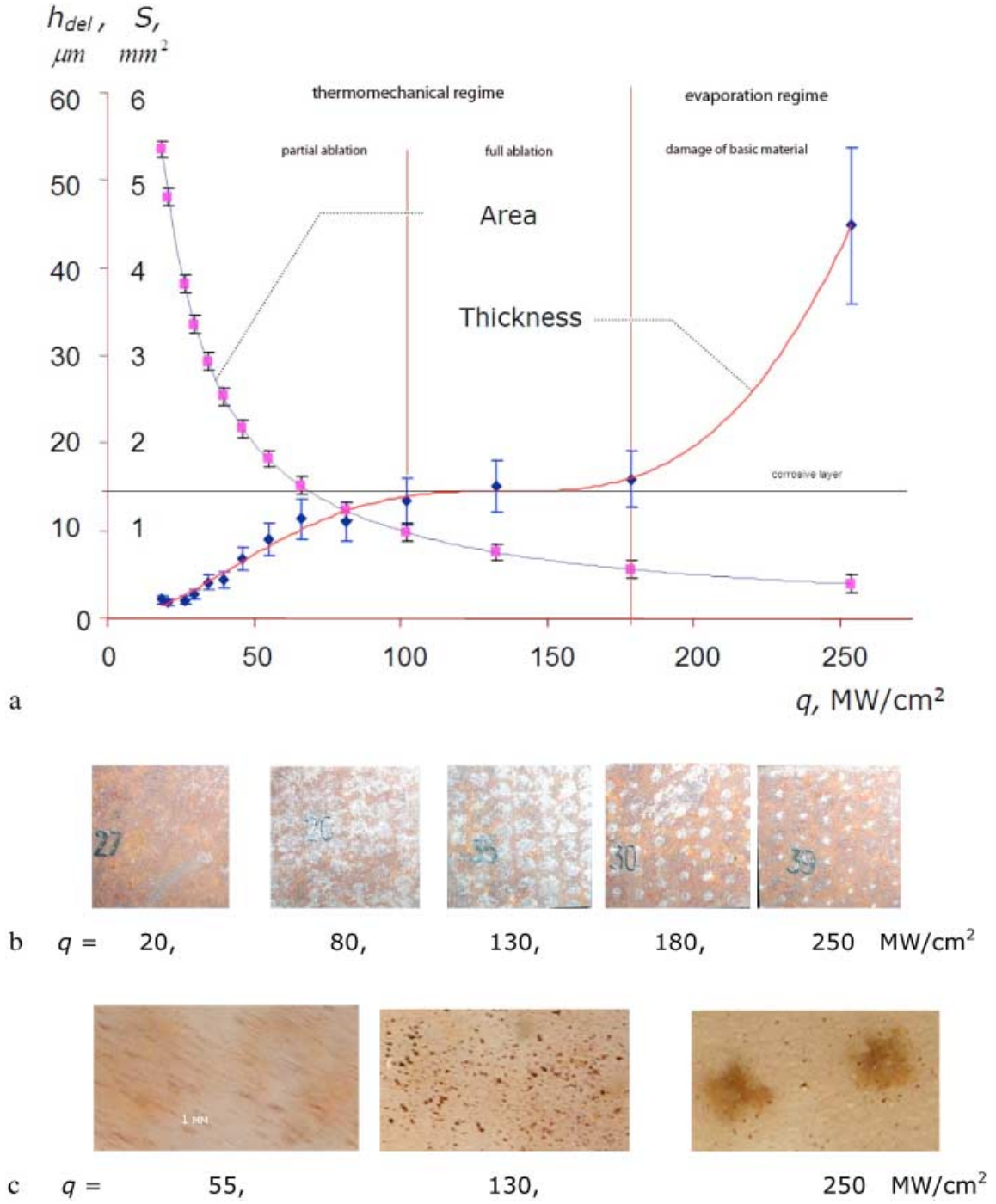

Fig. 6. Results of laser ablation of corroded layer and condensation of ablation products at collector film: (**a**) ablated layer thickness h_{del} and processed surface area S versus intensity q of applied laser radiation; (**b**) images of sample surface after processing with a given laser intensity power, (**c**) images of collecting film with deposited material.

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their radioactivity. For alfa-emitters the background radiation levels were reached after laser-based deactivation, and for beta-emitters initial activity was reduced by a factor of tens or more.

ACKNOWLEDGMENTS

This work was supported by a grant from the President of the Russian Federation (HW-1370.2003.8), grant RFBR (04-02-166-11a), and Russian Government grant 02.445.11.7108.

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