Stability of multilayers for synchrotron optics

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The temperature stability of metal (W, WRe, Co, Cr)-carbon multilayers has been studied using x-ray diffraction (θ–2θ and Debye–Scherrer) and electron microscopy. The results show that in all cases a crystallization occurs in the temperature range 650–750 °C. As a consequence of this crystallization, the layered structure is destroyed, the surface of the film becomes rough, and the x-ray reflectivity is considerably reduced. These results imply that efficient cooling or new multilayer structures will have to be developed for use at high temperatures or under high x-ray incident flux.

The appearance of new synchrotron sources with electron energies above 5 GeV requires the development of beamline components for high thermal loads as well as techniques to handle such heat loads. Candidates for synchrotron dispersing elements become rare when one tries to satisfy the requirements of thermal expansion, radiation damage, and vacuum stability.

As wide bandpass elements, metallic multilayers could be used as soft[1] or hard[2] x-ray monochromators. For the soft x-ray range, they present better stability than organic crystals.[3] At higher energies (>800 eV) narrow bandpass natural crystals are used. However, natural crystals require an alignment precision of a few seconds of arc.[4] Under intense beams, in a double crystal monochromator, the first crystal lattice expands, which mismatches with the second crystal and leads to errors in energy calibration.[5] Presently, crystals are at the limit of handling the thermal loads at SSRL with available power densities.[5] A 6-GeV undulator would produce 71 W/mm^2 at 50 m and 18 W/mm^2 at 100 m.[6] The SSRL beamline Wunder undulator producing 4.1 W/mm^2 at 10 m would increase the temperature of a grating above 500 °C.[7] Therefore, it is quite likely that the 6-GeV undulator will increase this temperature considerably above 500 °C. A solution to this problem would be to use multilayers to filter out most of the heat load that could reach a double crystal monochromator. Even this way efficient cooling schemes have to be designed; rotating optics, liquid metal cooling, or grazing incidence have been proposed.[8] In addition, the highly nonuniform power distribution and the use of shutters cause large stresses and thermal distortions.[9] Since the multilayer optical elements, used as reflecting elements in soft x-ray optics,[10–12] will be subject to high intensity radiation and/or temperature, it is of importance to study their temperature stability in detail.

We present here a study on the stability of W-C, WRe-C, Co-C, and Cr-C multilayers up to ~ 1000 °C using standard θ–2θ diffraction, Debye–Scherrer scattering, and electron microscopy. The results show that in all cases studied a crystallization occurs between 650 and 750 °C in one of the components. As a consequence the layered structure is destroyed, the film surface roughness increases, and the x-ray reflectivity decreases considerably. This implies that new, more stable multilayered systems will have to be developed

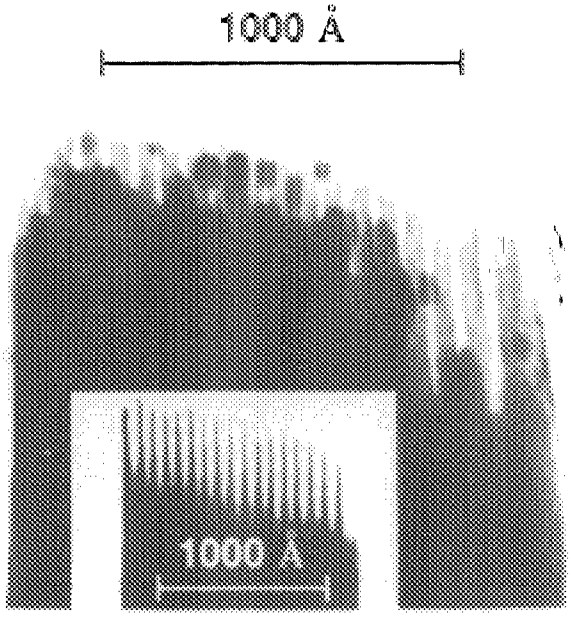

FIG. 1. Microcleavage electron microscopy cross section of a WRe-C multilayer at room temperature (dark area, WRe, light area, C). The inset shows the same multilayer after annealing at 750 °C. The contrast has been changed to emphasize the C. Notice that part of the heavy Z component is agglomerated.

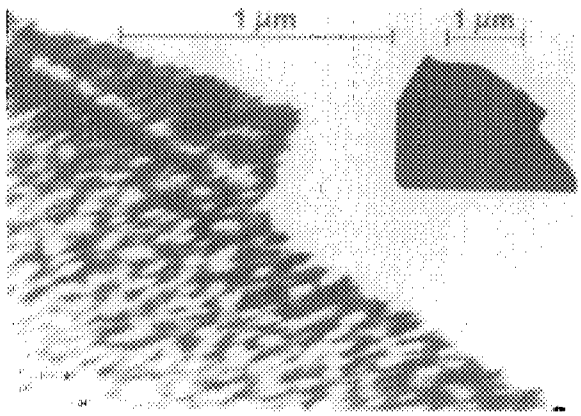

FIG. 2. Inset is a top view of the same film shown in Fig. 1 at 700 °C just below the crystallization point. This shows the same film at 900 °C clearly showing the formation of hillocks and roughening of the surface.

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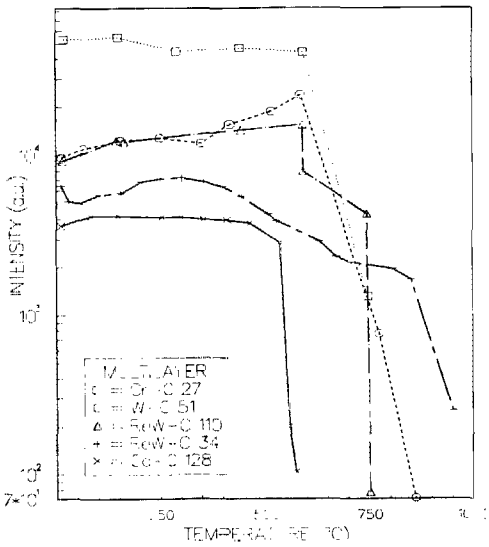

FIG. 3. Intensity of the first order Bragg peak as a function of temperature.

for uses at high temperatures or under intense radiation, such as a synchrotron.

The WRe-C, Co-C, Cr-C, W-C multilayered samples were prepared using a multisource evaporation system described earlier.[13,14] Briefly, the materials are evaporated on Si substrates using electron beam guns in an ultrahigh vacuum environment. Typical evaporation rates were 0.5 Å/s and typical vacuum during evaporation was in the 10^{-6} Torr range. Some W-C multilayers were also prepared using a multisource hot filament magnetically enhanced triode sputtering technique. Here the float glass or Si substrate rotates on a planetarium and the sputtering targets are commuted to produce the layers. Standard θ–2θ [15] and Debye–Scherrer x-ray diffraction (Cu K_α radiation) and electron microscopy measurements were performed in temperatures up to ~1000 °C. The cross sections for electron microscopy have been prepared using a microcleavage technique[16] described earlier.

The inset in Fig. 1 shows a transmission electron micrograph of an as-prepared WRe-C microcleaved cross section at room temperature. The micrograph clearly indicates the existence of well formed layers with reasonable uniformity along and across the stack. The dark area in this case is the high Z WRe constituent. A WRe/C multilayer kept at 730 °C for more than one hour showed no signs of deterioration. On the other hand, after approximately 5 min of heating at 750 °C the WRe agglomerates and some moves out from in between the C layers as shown in Fig. 1, thus showing the phenomena to occur abruptly around 750 °C. The contrast here has been changed in order to emphasize the C layers.

Below ~700 °C a top view of the same WRe-C multilayer shows the film to be featureless which indicates that the surface is smooth on a scale less than 50 Å (inset Fig. 2). On the other hand, a top view (Fig. 2) at 900 °C shows clearly the formation of hillocks showing that the surface has become considerably rough.

The intensity of the first order Bragg peak as a function of temperature is shown in Fig. 3. Care was taken to assure that indeed the first order Bragg peak intensity is changing and the effects are not due to changes in the diffractometer alignment as a function of temperature. Most samples were annealed *in situ* in an Ar environment in order to avoid oxygen contamination. In this case, a correction was applied to account for changes in the absorption due to the Ar gas. Some samples were also annealed in air in order to study their behavior in a room environment. Figure 3 shows that the intensity of the first order peak in all cases is relatively constant up to the crystallization temperature and then precipitously decreases below detectable limits. Electron diffraction [Fig. 4(a)] experiments on the crystallized samples showed the existence of a large number of crystalline rings. In order to identify accurately the crystallites that are formed, we performed Debye–Scherrer diffraction[17] on the sample scraped off the substrate. In some cases one of the disordered components of the multilayer crystallizes other times a crystalline compound is formed. In all cases we were

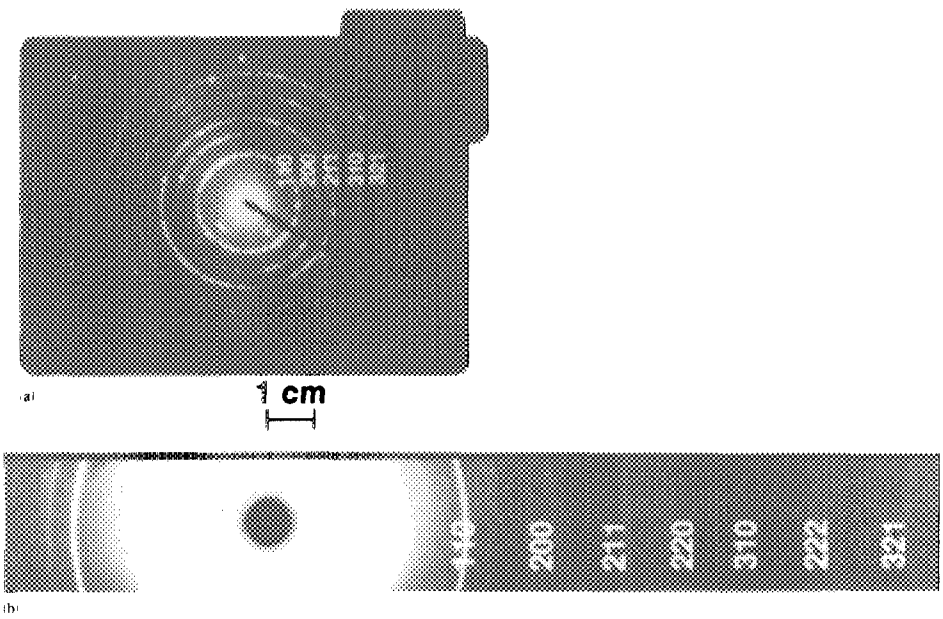

FIG. 4. (a) Electron diffraction picture of a W-C multilayer after crystallization showing seven distinct tungsten rings. (b) Corresponding Debye–Scherrer picture, showing also some weak Si or unidentified additional rings.

TABLE I. Parameters for the samples measured in these experiments.

High Z material (d = period)	Number of layers	Gas environment	Crystal formed	Annealing temperature (°C)
ReW ($d = 77$ Å)	34	Ar	hexagonal not indexed	950
ReW ($d = 36$ Å)	110	Ar	Re	750
		Air	WO_3 + Re	500
W ($d = 32$ Å)	51	Ar	W	770
			WO_3	630
Co ($d = 32$ Å)	128	Ar	not indexed	575
Cr ($d = 71$ Å)	27	Ar	Cr_2O_3	745

able to substantiate these claims by indexing a large number (up to seven) of Debye–Scherrer rings [Fig. 4(b)]. Table I shows the parameters used in the experiments and summarizes the results. Notice that for the case of W-C contrary to our expectations and earlier claims[18] we have *not* observed the formation of a carbide but rather the recrystallization of the disordered W. We would like to point out that earlier annealing studies[19] at 400 °C, for 4 h in vacuum for ReW/C, Cr/C, V/C, and Ru/C multilayers showed no appreciable changes in x-ray reflectivity, while Ni/C, Co/C, and Fe/C multilayers crystallized and lost their reflectivity.

In summary, we have performed extensive structural studies of a variety of metal-carbon multilayers which are potential candidates for x-ray optics in the 10–100 Å range. We find that for all systems studied a recrystallization occurs in one of the components which destroys the layered structure, roughens the surface, and considerably decreases the x-ray reflectivity.

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