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A Simple and Complete Purification of Single-Walled Carbon Nanotube Materials**

By Anne C. Dillon, Thomas Gennett, Kim M. Jones, Jeffrey L. Alleman, Philip A. Parilla, and Michael J. Heben*

Carbon single-walled nanotubes (SWNTs) must be thoroughly purified if they are to be used in a variety of projected applications and basic studies. Here we describe a

non-destructive, scaleable, three-step purification process that produces materials with >98 wt.-% purity. A dilute nitric acid reflux digests, functionalizes, and redistributes the non-nanotube carbon fractions to form a uniform and reactive coating on the SWNTs. This coating is selectively removed by oxidation in air. Raman spectroscopy, inductively coupled plasma spectroscopy (ICPS), and thermogravimetric analysis (TGA) are used to evaluate the purity of the material at each step of the process, and illustrate that very few tubes are consumed. Such a purification technique is not currently available,^[1-4] and the use of TGA and ICPS defines a technique by which the components in raw and processed materials may be accurately determined on a weight percent basis.

SWNT materials were synthesized by a laser vaporization method similar to that of Thess et al.^[5] but a single Nd:YAG laser was used (~450 ns pulses at 1064 nm and 10 Hz, average power of 20–30 W/cm²). Material was produced at rates of 75–150 mg/h. The power density was selected to operate in a vaporization regime^[6] during synthesis so that graphite particles were not ejected from the target and incorporated into the vaporized soot. Graphite particles introduced by such “sputtering” are not removed effectively with the purification process described here. Targets were made by pressing powdered graphite (~1 μm particle size) doped with 0.6 at.-% each of Co and Ni in a 1 inch × 1/8 inch dye (1 inch = 2.54 cm) at 10000 psi (1 psi ≈ 6.89 kN m⁻²). Crude soot was produced at 1200 °C with 500 torr Ar flowing at 100 sccm. The transmission electron microscopy (TEM) image in Figure 1a reveals the components of the laser-generated material. Bundles of SWNTs span between large agglomerations of amorphous and nanocrystalline carbon and metal nanoparticles. Raw materials were estimated to contain ~20–30 wt.-% SWNTs by a detailed analysis of numerous different TEM images.^[6] ICPS was performed after complete air-oxidation of the carbon and thorough digestion of the residue in concentrated HNO₃. The same metal content was found in both the laser-generated crude material and the initial target (6 wt.-%).

Approximately 80 mg of crude material produced at ~20 W/cm² was refluxed in 60 mL 3 M HNO₃ for 16 h at 120 °C. The solids were collected on a 0.2 μm polytetrafluoroethylene-coated polypropylene filter in the form of a mat and rinsed with deionized water. After drying, an 82 wt.-% yield was obtained. The weight lost is consistent with the digestion of the metal and an additional ~12 wt.-% of the crude material. TEM showed that SWNTs were encased within the mat by a relatively thick and uniform carbonaceous matrix (Fig. 1b). The carbon matrix was completely removed by oxidation in stagnant air at 550 °C for 30 min, leaving behind pure SWNTs having a weight of ~20 % of the initial crude material (Fig. 1c). TGA in flowing air (100 sccm) showed the purified tubes to be quite stable (Fig. 2a). The decomposition temperature (T_d) at 735 °C, defined here as the inflection point during oxida-

[*] Dr. M. J. Heben, Dr. A. C. Dillon, Dr. T. Gennett,^[†] K. M. Jones, J. L. Alleman, Dr. P. A. Parilla
National Renewable Energy Laboratory
1617 Cole Boulevard, Golden, CO 80401-3393 (USA)

[†] On sabbatical from: Chemistry Department, Rochester Institute of Technology, 85 Lomb Memorial Drive, Rochester, NY 14623-5604, USA.

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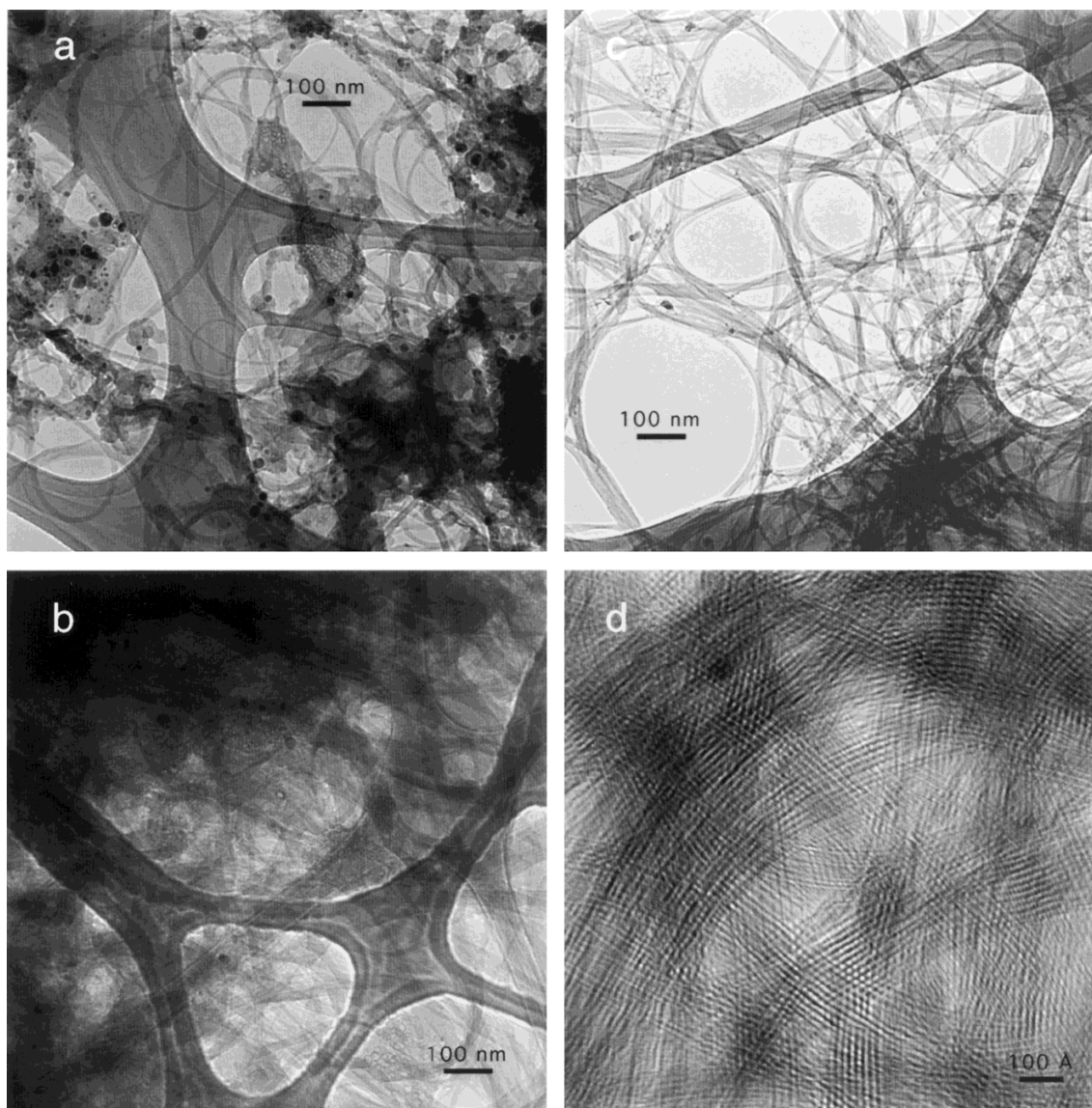


Fig. 1. Transmission electron microscopy images of: a) as-produced ~ 20 W/cm² laser-generated SWNT soot, b) crude material that was refluxed for 16 h in 3 M HNO₃, c) purified SWNTs produced by oxidizing the acid-treated sample for 30 min in air at 550 °C, and d) purified SWNTs at high magnification following a brief 1500 °C vacuum anneal.

tion of the tubes, is ~ 125 °C higher than a recently reported value.^[1] This result indicates a lack of dangling bonds, defects, and residual metal at which oxidation reactions can be initiated.^[7] Since less than 1 wt.-% is consumed below 550 °C, and less than 1 wt.-% remains above 850 °C, the final purity is conservatively estimated to be >98 wt.-%. The TGA technique establishes these SWNTs as the purest reported to date. Furthermore, the metal content was measured by ICPS and found to be the lowest reported to date at 0.2 wt.-% for both Ni and Co. The as-purified tubes were difficult to image at high magnification since tubes were left disordered within bundles after the acid reflux. However, a brief anneal to 1500 °C in vacuum was sufficient to re-order the tubes. The TEM image in Figure 1d reveals the high-quality, final material after the vacuum anneal.

TGA was also used to evaluate the crude and acid-refluxed materials to illuminate the key features of the purification process. The data for the crude soot (Fig. 2a) shows a slight increase in weight at low temperatures due to oxidation of the Ni and Co metals. The carbonaceous fractions begin to combust at ~ 370 °C and are mostly removed by oxidation below 600 °C. A small final weight loss beginning at ~ 650 °C is due to the oxidation of surviving SWNTs (~ 4 wt.-%). Consistent with observations by others,^[4] the majority of SWNTs are combusted at lower temperatures concurrently with other carbonaceous materials. The weight remaining at 875 °C corresponds to the weight expected for the oxidized metals (~ 8 wt.-%).

The TGA data from acid-refluxed material is also displayed in Figure 2a. A first thing to note is that refluxed

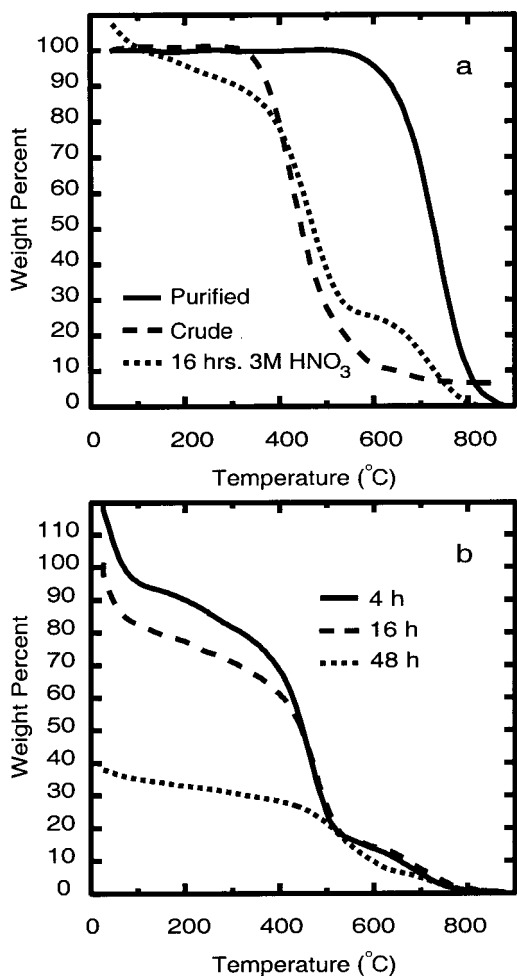


Fig. 2. Thermogravimetric analysis of 1–2 mg samples ramped from 25–875 °C at 5 °C per min in a platinum sample pan under 100 sccm flowing air. a) Materials produced at a laser power of ~20 W/cm², purified crude soot, and crude soot after a 16 h reflux in 3 M HNO₃. The data for the refluxed material was normalized to 100 wt.-% at 100 °C to compare dry weights. b) Materials produced with 30 W/cm² of laser power. Samples were refluxed in 3 M HNO₃ for 4, 16, and 48 h. These curves were normalized to 100 wt.-% at 100 °C to compare dry weights, and then re-normalized to account for the different weight losses in the HNO₃ refluxes.

samples getter as much as 10 wt.-% water from lab air, while purified and crude samples remain relatively dry. More importantly, non-nanotube decomposition occurs at a lower temperature and is completed before the onset of SWNT combustion. A plateau extending from 550 to 650 °C is evident in the TGA data because oxidation now occurs in two separate regimes. The sample weight is reduced to approximately zero by 850 °C since all carbonaceous materials have been removed and very little metal is left (0.4 wt.-%). The acid treatment not only removes most of the metal but also produces carboxyl, aldehyde, and other oxygen-containing functional groups^[7] on the surfaces of the non-nanotube carbonaceous fractions. As a result, the coating is extremely hygroscopic and reactive towards oxidation.

The combustion of non-nanotube carbons in the refluxed soot is essentially complete at the inflection point in the

TGA curve at 560 °C. At this temperature, the sample contains pure SWNTs amounting to ~26 wt.-% of the dry refluxed material, or ~21 wt.-% of the pre-reflux weight. This latter value is in excellent agreement with the yield after refluxed material was heated to 550 °C in stagnant air (~20 wt.-%), and considerably higher than the tube content determined by TGA analysis of the crude material (~4 wt.-%). The quantitative agreement between the bulk oxidation in stagnant air and the TGA measurements under dynamic conditions suggests that neither route consumed SWNTs appreciably. In fact, neither longer times in stagnant air at 550 °C (up to 1 h) nor holding the temperature at 550 °C during TGA experiments produced further significant weight loss. Since the weight loss proceeds as expected for oxidation of a single phase above 550 °C, and the TEM images of Figure 1c and d show only SWNTs, we can conclude that the final product is pure. It is important to note that the yields determined by purification and analysis are reproducible within two percentage points both for samples from a given laser run, and for samples produced in different runs using the same target/laser parameters.

To determine the extent to which tubes are damaged or consumed during the acid reflux, TGA was performed on materials refluxed for 4, 16, and 48 h in 3 M HNO₃ (Fig. 2b). Materials for these experiments were produced with 30 W/cm² of laser power. The data were adjusted for the dry weight lost during reflux so the y-axis represents the weight percent remaining of the initial crude material. The data for the 4 and 16 h refluxes overlay at temperatures above ~450 °C, and a plateau associated with SWNT stability is observed at 540 °C and a SWNT content of 17 wt.-%. It is striking that the TGA curves are virtually identical at the higher temperatures considering the difference in masses consumed during the reflux step. Since the SWNT content is determined to be the same in both cases, it is likely that neither reflux consumed a significant number of tubes. As discussed earlier, tubes are evidently not consumed by oxidation below 550 °C, so the 17 wt.-% value can be taken as an accurate assessment of the SWNT content in the crude soot. Once again, this value is in good agreement with the yield obtained from batch oxidation at 550 °C after a 16 h reflux in 3 M HNO₃. Unlike the 16 h process, the 4 h reflux did not always permit good purification by oxidation. In these cases a TGA curve very similar to that of the crude material was observed.

The oxidation reactions are no longer well-separated after a 48 h reflux (Fig. 2b) as only a small SWNT stability plateau is seen at ~625 °C. The affinity for water is considerably less than in either the 4 or 16 h samples. The uniform, hydrophilic carbon matrix produced after 16 h of refluxing was not observed by TEM. Instead, a patchier coating was observed along with occasional agglomerations, and some tubes appeared clean and uncoated. The TEM images revealed the non-nanotube carbons to be distributed much like as in the crude soot. Unlike the crude material, however, some of the tubes were observed to be

sharply angled, cut, and damaged. The extended reflux apparently digests most of the non-nanotube carbon and also begins to attack the SWNTs. These cut and defective tubes are then more susceptible towards air oxidation such that only ~8 wt.-%, or <50 % of the tubes known to be present remain at the onset of the SWNT stability plateau (Fig. 2b).

The Raman spectra of the purified and crude materials (Fig. 3) show two strong features at ~1593 and ~1567 cm^{-1}

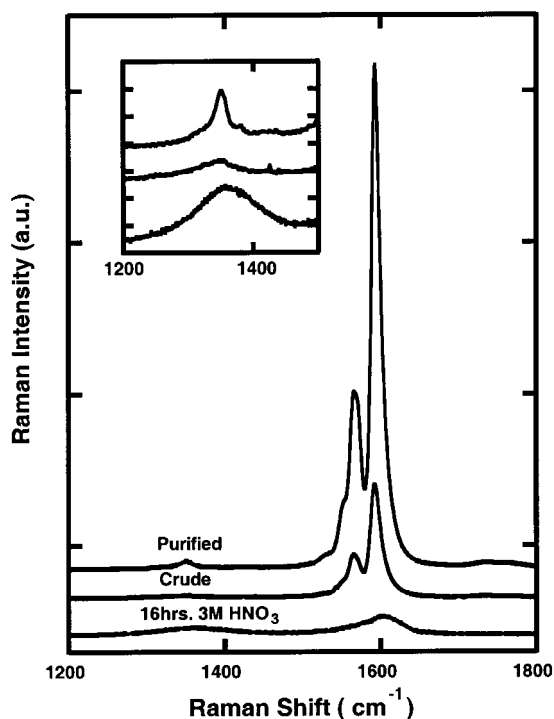


Fig. 3. Raman spectra obtained at 488 nm with a resolution of 2–6 cm^{-1} for purified, crude, and crude material that was refluxed for 16 h in 3 M HNO_3 . Inset: Magnification of the range between 1200 and 1500 cm^{-1} .

as expected for the 1.2 to 1.5 nm diameter SWNTs produced here.^[8] The features are approximately four times stronger after purification, consistent with the improvement in SWNT content. The origin of the slight blue shift of the signal from the pure relative to that of the crude material is not completely understood, and currently under study. The inset of the figure shows the region from 1200 to 1500 cm^{-1} at an amplified intensity scale. The feature at 1349 cm^{-1} in the crude material is due to the disordered sp^2 carbon “D-band” of non-nanotube graphitic components^[9,10] and indicates the presence of impurities. The D-band is more intense and narrower in the purified material, as observed by others,^[2] presumably due to curvature-induced enhancement of electron–phonon coupling.^[11] The Raman spectra of material midway in the purification process, i.e., after the 16 h reflux and before oxidation, is also shown in Figure 3. Here, the D-band is moderately intense and very broad, indicating a large disordered nanocrystalline graphite component^[12] (Fig. 3). Also, a signal derived from the fundamental E_{2g} mode of disordered graphite is

observed instead of the intense SWNT modes. The disordered graphite^[13,14] matrix formed by the reflux prohibits observation of the E_{2g} modes of the SWNTs despite the fact that the SWNT content in the 16 h HNO_3 -treated sample is actually higher than in the crude material. However, as seen in Figure 1b, the coating is somewhat transparent to electrons at the edges.

The 16 h 3 M HNO_3 reflux produces a uniform coating of disordered carbon on the SWNTs without introducing damage. Reactive functional groups are also introduced onto the surfaces of the non-nanotube carbon material during the process.^[7] These two effects combine to maximize the surface area and oxidation kinetics of the non-nanotube carbon. The functionalized coating is oxidized at lower temperatures and, since it is evenly distributed, the heat generated by exothermic oxidation does not initiate combustion of the SWNTs. In contrast, SWNTs in raw materials are consumed simultaneously with impurities because the oxidation of carbonaceous and metallic agglomerations generates local “hot spots”. The combination of the high surface area, reactivity, and uniformity of the carbon film produced by the 16 h 3 M HNO_3 reflux allows non-destructive purification of SWNTs by air oxidation.

A comparison of our results with those obtained by others is not straightforward since different production methods applied in different laboratories produce materials with SWNTs, non-nanotube carbon, and metal fractions that vary in type and configuration. For instance, when we applied our method to materials produced by arc-discharge^[15] we found tube contents below 5 wt.-% by TGA and a metal content greater than 25 wt.-% by ICPS in the final purified materials. Thus, the methods reported here were not sufficient to remove the majority of the metal from this commercial material. Some tubes may have been consumed during the purification process as it was more difficult to find SWNTs using TEM after purification. These materials are reported to contain 50–70 vol.-% SWNTs by the manufacturer. Our weight percent findings cannot be rationalized with reported volume percent values since there is currently no direct method to translate between the two. This is especially true when reported volume percent values are actually areal percent values obtained by scanning or transmission electron microscopy examinations.

Similarly, purification strategies developed by others may not be applicable to our materials. We applied a method utilizing a 4 h reflux in concentrated HNO_3 followed by several dilution/centrifugation/decanting cycles^[3] in an attempt to purify our SWNTs. The resulting SWNTs appeared pure when analyzed by TEM, but TGA studies indicated a purity of only 50 wt.-%. After oxidation in air the tubes were very pure and exhibited a T_d of 785 °C by TGA. However, TEM of the purified material revealed degradation after prolonged exposure to air, which might be due to defects introduced by the process. A weight loss totaling ~10 wt.-% below 550 °C was observed by TGA after 7 days in air. Approximately 3 wt.-% was lost at 100 °C and another 3 wt.-%

% decrease was observed at 200 °C with a final 4 wt.-% loss observed above 350 °C. In contrast, the SWNTs purified by the 16 h 3 M HNO₃ reflux and air oxidation remained >98 % pure in lab air for several weeks without any degradation. Unfortunately, no detailed analysis was performed to determine the composition of the final sample in the previous study.^[3] Consequently, we cannot conclude whether their final material really was impure, or if our materials are not sufficiently stable to be purified by concentrated HNO₃. Regardless, a rigorous comparison is difficult to make without accurate weight-based determinations.

Another comparison can be found in the procedure described by Rinzler et al.^[1] Here, a process involving numerous steps yields ~10–20 wt.-% of the initial material at the end. The final purity is not quoted but can be estimated from the TGA data reported by Rinzler et al.^[1] to be ~90 wt.-% from weight loss before 550 °C and the weight remaining after combustion of the SWNTs. The first step (45 h reflux in 2–3 M HNO₃) is similar to one that damaged our SWNTs (Fig. 2b). The TGA data^[1] shows a T_d that is 125 °C less than the 735 °C value reported here under the same experimental conditions. The lower T_d is consistent with either the presence of defects, metal impurities,^[7] or enhanced oxidation thermodynamics associated with higher SWNT curvature. Raman spectroscopy of the radial breathing modes (not shown) indicated a slightly larger mean diameter for our materials. Other explanations might involve differences in non-nanotube carbon content and configuration.

The dilute HNO₃ reflux/air oxidation procedure described here appears to be a simpler and more effective purification process than any previously reported. The reflux must be performed for sufficient time to produce a carbon coating on the SWNTs that can be removed by oxidation. However, exposures that are too extensive or in concentrated acids result in damage and digestion of SWNTs. Also the initial material should not contain relatively large graphite particles, large concentrations of metal, or graphite-encapsulated metal particles. Presently, there is not a good method by which initial and final SWNT content may be readily evaluated. Most estimates are now based on determining areal densities observed in selected TEM images, or observing the intensity of characteristic Raman signals. The techniques described here should provide a reliable means by which SWNT content may be determined on a weight percent basis in both crude and purified materials, enabling new comparisons.

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Direct Relationship Between Shape and Size of Template and Synthesis of Copper Metal Particles

By Marie-Paule Pileni,* Barry W. Ninham,
Tadet Gulik-Krzywicki, Judith Tanori, Isabella Lisiecki,
and Arlette Filankembo

Over the last few years, many groups concerned with membrane mimetic systems and nanoparticles have assumed that a first key step in the control of biomineralization must be the self-assembly of an appropriate microstructured template.^[1,2] Then, under controlled conditions, minerals of a required structure and shape might be induced to form within that space. However it has been clearly shown that, for example, production of calcium carbonate assemblies with structures similar to those occurring in nature do require the presence of some polypeptides, proteins, or other specific compounds.^[3,4] This indicates that the simple templating scenario is by no means obvious and remains unproven.

In many attempts using organized media as templates to mimic biomaterial synthesis of calcium carbonate and other materials, there has been no clear connection between product and the initial supposed microreactors.^[5,6] One reason is that the template structure changes due to

[*] Prof. M.-P. Pileni, Dr. J. Tanori, Dr. I. Lisiecki, Dr. A. Filankembo
Laboratoire SRSI, URA CNRS 1662
Université P. et M. Curie (Paris VI)
B. P. 52, 4 Place Jussieu, F-75231 Paris Cedex 05 (France)
and
CEA-CEN Saclay, DRECAM-SCM
F-91191 Gif-sur-Yvette Cedex (France)
Prof. B. W. Ninham
Department of Applied Mathematics
Australian National University
PO Box 4, ACT Canberra, 0200 (Australia)
Dr. T. Gulik-Krzywicki
Centre de Génétique Moléculaire-CNRS
F-91190 Gif-sur-Yvette Cedex (France)